OPTICAL HOLOGRAPHY IN DICHROMATED GELATIN.

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ABSTRACT.

Coupled wave theory is used to investigate the properties of high quality holograms recorded in dichromated gelatin. Initially, the properties of planar reflection gratings are studied in order to characterize the behaviour of the recording material in detail. Dichromated gelatin is then used to fabricate more sophisticated holograms for several applications.

Chapter 1 contains an introduction to holography, holographic optical elements and coupled wave theory. Chapter 2 contains a detailed review of dichromated gelatin based on published literature and on experimental observations made during work for this thesis. In Chapter 3 the non-linearity of the recording characteristic and changes in the bulk properties of the material during processing are determined. In Chapter 4 the non-uniformity of planar gratings is investigated by making a detailed comparison with a coupled wave theory in which the grating parameters are functions of depth. The effect of different processing procedures on these non-uniformities is demonstrated. In Chapter 5 the mechanism behind the formation of real-time gratings in dichromated gelatin is determined by applying a dynamic form of coupled wave theory. The source of diffraction is found to be a pure absorption grating. In Chapter 6 the use of dichromated gelatin as a medium for copying holograms and for recording efficient noise gratings is demonstrated. In Chapter 7 a theoretical model of holographic optical elements based on vectorial coupled wave theory is presented and is used to investigate the effects of polarisation. In Chapter 8 an optical method for the measurement of distance is developed using the dispersive properties of an on-axis holographic lens. Conclusions are then presented in Chapter 9.
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CHAPTER 1.

INTRODUCTION.

The fundamental principle of optical holography is the recording and reconstruction of both the amplitude and phase of an optical wavefront. Since in normal circumstances it is only possible to detect the intensity of light it is not possible to record the phase of the wavefront directly. However, the spatial intensity variation due to the interference between a mutually coherent object wave and reference wave contains both amplitude and phase information. If we record this interference variation, and are aware of the nature of the reference wave, we have all the information necessary to reconstruct the original object wavefront.

1.1 THE DEVELOPMENT OF HOLOGRAPHY.

The principle of wavefront reconstruction was proposed by Gabor in 1947 [GAB48]. His original aim was to improve the resolution of electron microscopy which was limited by the spherical aberration of magnetic electron lenses. To do this he intended to use a lensless imaging technique whereby the scattered field from an object illuminated by electrons was recorded and then reconstructed later, using visible light. This idea was stimulated by the Bragg X-ray microscope which utilised similar principles [BRA39]. However, Gabor realised that recording the interference pattern of any two object waves would produce a complex
grating or hologram which, when illuminated by one of the original recording waves, would reconstruct the other wavefront by diffraction.

Unfortunately, the low coherence of the light sources available to Gabor forced the use of an in-line geometry which led to annoying spurious diffraction orders. Another problem was that the silver halide recording materials available at the time were used to produce absorption gratings, which led to low efficiency.

It was not until 1962 that significant advances in holography occurred. The appearance of the laser allowed an off-axis reference beam to be used [LEI62, LEI64]. As a result, the orders were spatially separated and the power in the higher orders was significantly reduced.

At about the same time another recording configuration was investigated by Denisyuk [DEN62] which had similarities to Lippman colour photography [LIP94]. In this configuration the reference and object wave were contra-propagating and were incident from opposite sides of the photographic plate. As a result the diffracted wave appeared to be reflected by the hologram rather than transmitted. The high spatial frequency of these reflection holograms meant that higher order diffraction was almost totally eliminated and, because they were wavelength selective, replay with white light was possible [STR66].

Further significant advances in holography were then mostly concerned with recording materials. Cathey obtained high diffraction efficiencies by converting the absorption modulation in silver halide emulsions to phase modulation using a bleaching process [CAT65]. Phase holograms recorded in dichromated gelatin were then reported by Shankhoff [SHA68]. This new material was found to have ideal properties for recording high efficiency, low noise phase holograms.

Since this early work holography has continued to excite both scientific and public interest. However, despite this interest,
holography has not yet achieved its full potential in scientific and artistic terms. The major reason for this is the restrictions and difficulties imposed by the available recording materials and coherent light sources.

1.2 DICROMATED GELATIN.

The function of a holographic recording material is to record the spatial intensity variation of light, due to the interference between the object and reference wave, as a spatial variation of refractive index or absorption. The performance of a recording material can therefore be expressed in terms of its resolution, noise and modulation capacity.

In these terms dichromated gelatin is a good recording material. The spatial resolution is almost uniform between 100 and 5000 lines/mm. Unlike silver halide materials, where the light sensitive agent is in the form of fine crystal grains, dichromated gelatin is homogeneous and therefore the level of scatter is very low and a high signal to noise ratio can be achieved. The level of refractive index modulation that can be obtained is also high (8% or more) and, with the low level of scatter, this can lead to diffraction efficiencies as high as 98%.

Dichromated gelatin is also a very flexible recording material. It can be used in both thin (<1 μm) and very thick (>100 μm) layers, unlike silver halide recording materials which are limited to thicknesses below 20 μm by processing difficulties. It is also possible to adjust the tuning (of the Bragg condition) and the bandwidth of dichromated gelatin holograms by altering processing procedures and if necessary it is possible to reprocess holograms until the desired properties are obtained. These features will be described in more detail in Chapter 2.

However, despite the performance and flexibility of the material, the use of dichromated gelatin is not as widespread as might be expected.
This is due to the fact that there are still several problems associated with its use.

The first problem is that of reproducibility. At many stages of the dichromated gelatin process the material is sensitive to humidity and temperature and therefore the control of the environmental conditions is essential. The material also has a limited shelf life, due to a slow dark reaction, and reproducible results will only be obtained if the timescale of exposure and processing is fixed. Considerable effort was required in the initial stages of work for this thesis to obtain acceptable and reproducible results for the quantitative studies in Chapters 3 and 4.

The second problem is that the exposures required for dichromated gelatin are typically 1000x greater than those required for silver halide recording materials. The sensitivity increases from green to ultraviolet but is negligible in the red region of the optical spectrum unless the added complication of dye sensitivity is used. Since the amount of light available is limited it is inevitable that long exposures are needed, particularly for large holograms or when uniform diffuse illumination is required, and this places stringent requirements on the stability of the recording arrangement.

1.3 HOLOGRAPHIC OPTICAL ELEMENTS.

Holograms are not limited in function to the storage and display of information; they are also capable of behaving as optical elements. Consider a hologram recorded with a simple plane wavefront and a spherical wavefront. At replay the hologram can transform either of these wavefronts into the other. Transmission holograms recorded in this way can therefore perform an identical optical function to a conventional lens using diffraction rather than refraction. Reflection holograms can
perform an identical optical function to curved mirrors using diffraction rather than reflection.

These holographic optical elements (HOEs) have several unique properties which may be used in novel applications. For example, they are highly dispersive. This property will be utilised in Chapter 9, but in general it limits the use of HOEs to monochromatic systems. They also display angular and wavelength selectivity. The optical function of HOEs is independent of the shape of the recording material and they can be recorded on any surface, such as a curved visor or on a conventional lens. Finally, it is possible to superimpose several HOEs in one emulsion by making several exposures. The ease with which holographic elements can be fabricated and replicated on thin, light weight substrates contrasts strongly with the difficult grinding processes required for conventional optics.

Holographic optical elements have been reviewed by Close [CL075]. Transmission elements have included lenses [CHA68, RIC74, SYM83, ONO86], and multiple lenses [LIA83, SLI85]. More specific applications have been couplers for fibres [SOA81] and waveguides [NIS75], and scanners for bar code readers [LEE77] and laser printers [IWA86]. Reflection elements have included parabolic [MIN75], ellipsoidal and hyperbolic mirrors [GUP77], scanners [ISH84] and a highly successful application has been in head-up displays for aircraft [COO77]. The higher selectivity of reflection gratings has also led to their use for wavelength demultiplexing [DUN85] and makes them more useful in white light systems.

At present the use of holographic optical elements is confined to applications where their unique properties have particular advantages (such as head-up displays) but more widespread use may be found in the future.
1.4 COUPLED WAVE THEORY.

Throughout this thesis extensive use is made of coupled wave theory. In this theory it is assumed that there is a continuous exchange of energy between the waves propagating inside the hologram which are coupled together by the grating. This can be represented mathematically as a set of coupled differential equations which can be derived from the wave equation.

The coupled wave approach was first used by Raman and Nath in an analysis of the diffraction of light by ultrasonic waves in liquids [RAM35]. However, the first application of coupled wave theory to holography was made by Kogelnik [KOG69] who was able to account for many of the properties of volume gratings. His one-dimensional analysis has since been extended to treat many other problems in holography and the widespread use of coupled wave theory has been reviewed by Solymar and Cooke [SOL81] and others [RUS81, GAY85].

This chapter will initially follow Kogelnik's approach and the same basic limitations of one-dimensional theory will be imposed on the system under consideration. We shall assume that a uniform plane grating is contained within a parallel sided slab which extends to infinity in the transverse directions and is surrounded by a medium of the same average dielectric constant. At replay an infinite plane wave of uniform amplitude is incident and we will impose the restriction that the electric field inside the hologram is perpendicular to the plane of the grating, which will allow the use of the time-independent scalar wave equation.

1.4.1 Hologram recording.

At recording, two infinite plane waves are assumed to be incident on the photosensitive material as shown in Fig. 1.1 (a) for reflection
Fig. 1.1. Recording configuration and Ewald sphere representation for (a) Reflection holograms and (b) Transmission holograms.
gratings and (b) for transmission gratings. The intensity in the material will be given by

\[ I(\mathbf{r}) = I_1 + I_2 + 2|I_1||I_2|\cos(K_0 \cdot \mathbf{r}) \quad (1.1) \]

where \( I_1 \) and \( I_2 \) are the intensities of the two incident waves, \( \mathbf{r} \) is the position vector and \( K_0 \) is the latent grating vector defined by

\[ \mathbf{K}_0 = \mathbf{\hat{p}} - \mathbf{\hat{\sigma}} \quad (1.2) \]

where \( \mathbf{\hat{p}} \) and \( \mathbf{\hat{\sigma}} \) are the propagation vectors of the incident waves. After processing the recording material this cosinusoidal variation of intensity will be converted to a periodic variation in permittivity. We will assume for this analysis that the response of the recording material is linear. Within the parallel boundaries of the material, which are defined by the planes \( x=0 \) and \( x=d \), the complex permittivity \( \varepsilon(\mathbf{r}) \) is now given by

\[ \varepsilon(\mathbf{r}) = [\varepsilon'_0 - j\varepsilon''_0] + [\varepsilon'_1 - j\varepsilon''_1]\cos(K \cdot \mathbf{r}) \quad (1.3) \]

where \( \varepsilon'_0 \) and \( \varepsilon''_0 \) are the real and imaginary parts of the average permittivity, \( \varepsilon'_1 \) and \( \varepsilon''_1 \) represent phase modulation and absorption modulation and where \( \varepsilon'_1, \varepsilon''_1, \varepsilon''_0 \ll \varepsilon'_0 \). \( \mathbf{K} \) is the grating vector after any thickness change during to processing and is given by

\[ \mathbf{K} = \frac{T_i}{T_f} \mathbf{k}_0 \mathbf{\hat{i}}_x + \mathbf{k}_0 \mathbf{\hat{i}}_y \quad (1.4) \]

where \( T_i \) and \( T_f \) are the initial and final grating thickness of the recording material.

Equation (1.4) is based on the assumption that because the recording material is restrained in the transverse directions by the substrate, a thickness change will only affect the \( x \) component of the grating vector. During work for this thesis on very thick gelatin layers
some evidence has been seen of sideways shearing, but in most circumstances this assumption is reasonable.

1.4.2 Hologram replay.

At replay an infinite plane wave is incident. The electric field within the grating is defined by the scalar wave equation

\[ \nabla^2 E + (\beta^2 - 2j\alpha_0\beta + 4\kappa\beta\cos(K_y \cdot \hat{r})) E = 0 \quad (1.5) \]

where the propagation constant, \( \beta \), and the absorption constant, \( \alpha_0 \), are given by

\[ \beta = \frac{2\pi(\epsilon'_0)}{\lambda} \quad \alpha_0 = \frac{\beta \epsilon'_0}{2 \epsilon'_0} \quad (1.6) \]

and where we have defined a coupling constant, \( \kappa \), as

\[ \kappa = \beta \left[ \frac{\epsilon'_1 - j\epsilon'_{1'}}{4 \epsilon'_0} \right] \quad (1.7) \]

We now assume that the electric field inside the hologram is of the form

\[ E = \sum_{n=-\infty}^{\infty} A_n(x) \exp(-j\rho_n \cdot \hat{r}) \quad (1.8) \]

where \( A_n(x) \) are the scalar amplitudes of a set of waves with propagation vectors \( \rho_n \) and \( n \) is the order of diffraction. Because the amplitudes \( A_n \) are \( x \) dependent, the \( y \) components of the set of wavevectors, \( \rho_{ny} \), are related by the expression

\[ \rho_{ny} = \rho_{0y} + nK_y \quad (1.9) \]

where \( \rho_{0y} \) and \( K_y \) are the \( y \) components of the replay wavevector and the \( K \) vector respectively. At this point Kogelnik (who was considering only the 0th and -1th order) chose his diffracted wavevector to be defined by
\[ \rho_n = \rho_0 + n\hat{K} \quad (1.10) \]

This is referred to as 'K Vector Closure'. These vectors are shown in Fig. 1.2 (a) and in general are not of modulus \( \beta \). A more intuitively pleasing set of wavevectors, which are identical to the wavevectors of the diffracted orders in an external medium of the same average permittivity, are given by the Beta Value construction shown in Fig. 1.2 (b). The \( x \) components of these wavevectors may be obtained from the expression

\[ \rho_{nx}^2 + \rho_{ny}^2 = \beta^2 \quad (1.11) \]

It has been shown that better agreement with experimental results is obtained with the wavevectors defined from the Beta Value construction than from K Vector Closure [SYM82] and therefore they will be used in this thesis wherever possible.

Substituting (1.8) into the scalar wave equation (1.5) we obtain

\[ \sum_n \exp(-j\rho_n \cdot \hat{r}) \left[ \frac{d A_n}{dx} \frac{d A_n}{dx} + 2j\rho_{nx} \frac{d A_n}{dx} - 2j\alpha_0 \beta A_n \right. \\
\left. + 2\kappa \beta \left\{ \exp(j\hat{K} \cdot \hat{r}) + \exp(-j\hat{K} \cdot \hat{r}) \right\} \right] = 0 \quad (1.12) \]

If we assume that the energy exchange between the diffracted orders is slow then \( A_n \) will be slowly varying functions and we may neglect the second orders above. Substituting for \( \hat{K} \) in (1.12) and equating separately the coefficients of the exponentials with zero we obtain an infinite set of coupled differential equations

\[ \begin{aligned}
\rho_{nx} \frac{d A_n}{\beta \, dx} + \alpha_0 A_n + j\kappa \left\{ A_{n+1} \exp[j(\psi_{n+1} - \psi_n)x] \\
+ A_{n-1} \exp[j(\psi_{n-1} - \psi_n)x] \right\} = 0
\end{aligned} \quad (1.13) \]
Fig. 1.2 (a). An Ewald sphere diagram of the diffracted waves in the hologram specified by $\bar{K}$ Vector Closure theory.

Fig. 1.2 (b). An Ewald sphere diagram of the diffracted waves in the hologram specified by $\beta$-value theory.
where the scalar quantities $\psi_n$, termed the dephasing parameters, are given by

$$\rho_{nx} = \rho_{0x} + nK - \psi_n$$

(1.14)

The coupled equations describe the interchange of energy between the diffracted orders as they propagate through the hologram. The rate of change of the amplitude of each diffracted order, $A_n$, is related by the coupling constant to the amplitudes of adjacent orders $A_{n+1}$ and $A_{n-1}$, and to the absorption constant.

1.4.3 Optically thick holograms.

The coupling between adjacent orders is also dependent on the value of the exponential terms in (1.13). If the difference between the dephasing parameters of two orders is zero then they satisfy the Bragg condition for the grating and the coupling between them will be strong. For some holograms the exponential terms of higher orders may be large and therefore the coupling to these orders will be very weak. Holograms for which this occurs are termed optically thick or volume holograms. In these circumstances it is valid to neglect higher orders and to consider a smaller, finite set of coupled wave equations.

Several criteria have been assessed which help define whether a hologram is optically thin or optically thick [KOG69, MOH78, BEN80]. We discuss here the dimensionless parameter $\Omega$ defined as

$$\Omega = \frac{K^2}{2\beta\kappa}$$

(1.15)

It has been shown that a hologram for which $\Omega \gg 1$ will be optically thick [BEN80, SOL81]. The value of $\Omega$ is strongly dependent on the modulus of $\tilde{K}$. In general this means that holograms where the angle between the two recording waves is large, as in reflection holograms, will be optically thick. Because the power in all orders other than the incident
Oth order and the -1th diffraction order will be very small these holograms can be described adequately by two coupled differential equations.

1.4.4 Analytical solution for reflection holograms.

In the following Chapters the behaviour of optically thick planar reflection holograms is considered. It is therefore useful to derive the analytical solution to the coupled wave equations which can be obtained in the two wave case. Neglecting all higher orders the equations (1.13) reduce to

\[ \frac{\rho_0 x}{\beta} \frac{dA_0}{dx} + \alpha_0 A_0 + j \kappa A_{-1} \exp(j\psi_1 x) = 0 \]

(1.16)

\[ \frac{\rho_{-1} x}{\beta} \frac{dA_{-1}}{dx} + \alpha_{-1} + j \kappa A_0 \exp(-j\psi_1 x) = 0 \]

We can obtain analytical solutions to these equations for both reflection and transmission holograms after specifying appropriate boundary conditions. For reflection holograms these boundary conditions are

\[ A_0(0) = 1 \]
\[ A_{-1}(0) = 0 \]

(1.17)

The transmitted wave amplitude \( A_0(d) \) and the diffracted wave amplitude \( A_{-1}(0) \) are thus

\[ A_0(d) = \exp \left[ -j \xi - \frac{\alpha_0 \beta d}{\rho_0 x} \right] \left[ \cosh \phi - j \frac{\xi}{\phi} \sinh \phi \right]^{-1} \]

(1.18)

\[ A_{-1}(0) = \left[ -\frac{\rho_{-1} x}{\rho_{-1} x} \right]^{1/2} \left[ \frac{\xi}{\mu} + j \frac{\phi}{\mu} \coth \phi \right]^{-1} \]

where

\[ \xi = j \frac{\alpha_0 \beta d}{2} \left[ \frac{1}{\rho_0 x} - \frac{1}{\rho_{-1} x} \right] + \frac{\psi d}{2} \]
and

\[ \mu = \frac{j\kappa \beta d}{\sqrt{\rho_0 x^p - 1x}} \quad \text{and} \quad \Phi = \left[ \mu^2 - \xi^2 \right]^\frac{1}{2} \]

1.4.5 Further developments.

In Chapter 4 we will extend the two wave model to allow the grating parameters to vary with depth in the hologram (Appendix II). It will not be possible to obtain analytical solutions to the modified coupled wave equations and a numerical method of solution will be necessary.

In Chapter 5 we will allow for both a time dependence and a spatial variation of the grating parameters for a pure absorption grating.

Finally in Chapter 7 it will be necessary to remove some of the geometrical restrictions imposed at the beginning of this analysis and to allow for the arbitrary polarisation of the replay wave. To do this a vectorial form of coupled wave theory will be required.

1.5 THESIS SUMMARY.

The central theme of this thesis is the recording material, dichromated gelatin. In Chapter 2, therefore, the dichromated gelatin process is described in detail, with reference to the published literature, and to observations and deductions made during the fabrication of holograms for this thesis.

In the two Chapters which follow, the properties of simple planar reflection gratings in dichromated gelatin are examined. In these Chapters we seek to deduce the internal structure of the holograms by
examining the emerging light field, in a manner analogous to X-ray crystallography.

Firstly, in Chapter 3, we ignore any spatial variation of the grating parameters and compare the characteristics of the hologram with the uniform grating, one-dimensional coupled wave theory developed in this Chapter. Although some disparity between theory and experiment is found, we are able to determine the recording response of the material i.e. the microscopic refractive index modulation that results from exposure to a spatially varying light field. This is essential information if the full potential of the recording material is to be realised in later Chapters. We also make a quantitative study of the effect of processing on the bulk properties of the recording material, which have a strong affect on the replay Bragg condition of a hologram.

Secondly, in Chapter 4, we improve the agreement between theory and experiment by extending the model to allow for a spatial variation of the grating parameters. We are then able to see the internal grating structure in more detail and can observe how changing processing procedures can affect the non-uniformity of the gratings.

In Chapter 5 we examine the formation of real-time gratings in dichromated gelatin. These real-time gratings have several potential applications but they can also be related to the properties of processed gratings. By applying coupled wave theory, we are able to determine unambiguously the source of the real-time behaviour which is observed.

Having obtained a good understanding of the basic recording properties and mechanisms, the later chapters then examine the applications of the dichromated gelatin. It is found in Chapter 6 that dichromated gelatin has ideal properties for hologram copying and that copying is a useful technique for generating more sophisticated dichromated gelatin holograms, without any of the problems which might
otherwise occur. The copying of several classes of hologram are attempted, with particular attention to the relative modulation and noise in the original and copy. The properties of a novel type of hologram, which are recordings of an external scattered light field, are examined and are named 'synthesised noise gratings'.

In Chapter 7 we develop a theoretical model for holographic optical elements which makes use of vectorial coupled wave theory. This allows us to examine the effect of polarisation in on-axis and off-axis lenses. In Chapter 8 we then construct an optical position measurement sensor which utilises the dispersive properties of an on-axis holographic lens. The model developed in Chapter 7 is used to optimise the design of the lens to suit this application.

Conclusions are presented in Chapter 9.
CHAPTER 2.

DICHROMATED GELATIN.

The photosensitivity of dichromated gelatin has been known since the beginning of the 19th century. Areas which are exposed to light become hardened and less soluble than unexposed areas. It is therefore possible to generate relief images by washing the exposed material in warm water and it behaves as a linear, negative photoresist. This mechanism was used extensively by the printing industry in the "carbon tissue" process, where carbon black is incorporated into the gelatin before exposure, and for photo-engraving [KOS65].

The use of dichromated gelatin to produce surface relief holograms followed logically from its use in printing. However, it was soon discovered that a large refractive index variation could be induced in the bulk of the material by first soaking in water and then by drying rapidly with a desiccant such as propanol [SHA69]. To understand the mechanism behind the formation of the modulation of the refractive index it is necessary to consider the nature of gelatin in detail.

2.1 PHYSICAL PROPERTIES AND STRUCTURE OF GELATIN.

Gelatin is derived from the collagen found in animal tissues. The production of gelatin involves breaking down the fibrous collagen into a colloid of long helical chains of irregular size, shape and structure. These consist of various amino acids linked together by polypeptide
links. The exact chemical composition is very dependent on the starting material but can be considered as:

\[
\begin{array}{c}
\text{R} \\
\text{N} - \text{C} - \text{C} \\
\text{H} - \text{N} - \text{O}
\end{array}
\]

where \( R \) represents an amino acid such as glycine \([\text{MEY77}]\). The chains have a springy nature and are typically 80 nm long in their unstretched state \([\text{MEE66}]\).

Solutions of more than 1% gelatin will set to a gel if left to stand below 30°C. In this state the gelatin is weakly bound by hydrogen bonding between the non-polar regions of the molecules and the gelation is easily reversed by warming in water. However the gel can be rendered insoluble by baking or by chemical hardening, a process referred to as tanning.

Gelatin has the ability to take up large quantities of water from humid air or when immersed which becomes hydrogen bonded to the polar regions of the molecule or sandwiched between layers of molecules. The hydrogen bonding between water and gelatin is stronger than that between individual gelatin molecules and as water is incorporated the gelatin can swell to several times its initial thickness.

The ease with which water and dissolved chemicals can penetrate the gelatin matrix and the good optical quality of thin films makes gelatin ideal for silver halide emulsions in photography and holography. In this application the role of the gelatin is a passive support. However the dichromated gelatin process makes active use of the gelatin itself.
2.2 THE PHOTOCHEMICAL REACTION.

When incorporated into the gelatin matrix the hexavalent dichromate ion \( \text{Cr}_2\text{O}_7^{2-} \) can be reduced to trivalent \( \text{Cr}^{3+} \) by the absorption of a photon of sufficient energy. The exact photochemical process and the final form of the \( \text{Cr}^{3+} \) ion is uncertain but a possible reaction \([\text{KOS65}]\) is

\[
\text{Cr}_2\text{O}_7^{2-} + 14\text{H}^+ + 6\text{e}^- + h\nu \rightarrow 2\text{Cr}^{3+} + 7\text{H}_2\text{O}
\]

(Orange) \hspace{1cm} (Green)

The evidence for this is that during exposure the pH can be seen to be changing rapidly and both the pH \([\text{MAZ82}]\) and the presence of electron donors \([\text{MAZ85}]\) affect the light sensitivity. It is thought that the reduction can also proceed by an indirect route via chromic acid. This could explain the fact that there is a reduction in sensitivity as the intensity of the exposure is increased \([\text{CHA79}]\), termed reciprocity failure. The colour change during exposure causes increased absorption of the recording beams and leads to real-time effects which are investigated in Chapter 5.

In particular positions along the gelatin molecule there are the following active groups:

- \(-\text{OH}\) Hydroxyl.
- \(-\text{COOH}\) Carboxyl.
- \(=\text{CO}\) Carbonyl.
- \(-\text{NH}_2\) Amino.

The \( \text{Cr}^{3+} \) ions produced in the photochemical reaction react with two separate carboxyl sites (either on different molecules of gelatin or at different points on the same molecule) to form a coordinated complex which crosslinks the two sites:
Unlike the hydrogen bonding between gelatin molecules these crosslinks are very strong and stable. They harden the gelatin and reduce its ability to absorb water. In sufficient quantity they render the gelatin insoluble and many of the physical properties of the gelatin become altered.

2.3 THE MECHANISM OF MODULATION FORMATION.

In the production of dichromated gelatin holograms the spatial intensity variation of a standing wave interference pattern is recorded in the sensitised gelatin as a hardness variation. In the case of surface relief gratings the initial gelatin is soft and unexposed regions can be dissolved away [SHA68, MEY71]. However, if the gelatin is given a uniform overall bias hardness it is still possible to produce efficient holograms with little or no removal of gelatin. To do this the hardness variation is converted into a refractive index modulation in the volume of the gelatin by rapid dehydration in propanol.

The precise mechanism which occurs during this procedure is not fully understood and is still the subject of some controversy. Early workers suggested that stresses, due to shrinkage during dehydration, caused the gelatin to tear in the softer (unexposed) regions [SHA68, CUR70] creating air filled cracks or voids [CAS76]. There is some evidence to suggest that this may occur when the initial bias hardness is inadequate but it is not able to explain all observations.
Other authors have suggested that the index modulation was due to the formation of a chromium-propanol-gelatin complex [MEY72, SJ081]. However, the maximum index modulation that would be expected is about 2% and this mechanism cannot not explain the successful use of other drying agents or the index modulation sometimes seen before dehydration.

More recently it has been suggested the processing cycle results in an increase in gelatin density in exposed regions and a decrease in unexposed regions due to a rearrangement of the gelatin molecules [CHA80, MCG80]. After exposure and the removal of excess chromium compounds the gelatin is soaked in water and swollen. The degree of swelling that occurs is proportional to the hardness and thus the exposure. It is quite possible to see this differential swelling at the edges of the exposed area or if there are wide fringes running across the hologram (due to inadequate index matching). In this state the density of the gelatin is obviously lowered in the swollen areas. If the gelatin is dried slowly this density variation disappears. However, if the water is rapidly dried in propanol some of this density variation is frozen, perhaps because gelatin loses its pliability in the absence of water. Further it has been suggested that transfer of gelatin may occur between exposed and unexposed regions during processing due to osmotic forces [MCG80, CUL82].

The density change hypothesis allows the explanation of many the factors which affect dichromated gelatin processing, such as the complex effect of the bias hardness and the drying rate.

2.4 PREPARATION OF GELATIN PLATES.

Gelatin coated plates suitable for the production of dichromated gelatin holograms are not available commercially. The holograms described in this thesis were made with gelatin plates produced by the
spin coating method unless otherwise indicated and were supplied under special arrangement by Pilkington Brothers PLC Research Laboratories.

Various authors have suggested removing the chemicals from commercial silver halide plates for dichromated gelatin use. These plates are reasonably uniform within one batch with respect to thickness and gelatin quality. From batch to batch however, the gelatin quality may change and processing procedures may have to be adjusted. KODAK 649F photographic plates have been found to give good results as they have the correct level of gelatin hardness after removal of silver halide with a hardening fixer [LIN69]. Olivia [OLI84] found that AGFA 8E75 HD could only be used if the hardness of the gelatin was reduced and suggested a useful method to quantify the hardness of gelatin coatings. The weight of gelatin plates was measured before and after they were soaked in water for a fixed period of 15 minutes at 18°C. A good indication of the hardness of the gelatin was found to be a swelling factor, defined as the percentage increase in mass of the coating after soaking. A swelling factor of about 200 is optimum and the gelatin hardness was reduced by soaking in water at 90°C, until this was achieved.

Alternatively, coatings can be made specifically for dichromated gelatin use. The properties of the gelatin chosen will affect the processing requirements and therefore it is essential to use the same batch to ensure uniformity in successive experiments. Glass substrates can be cleaned with dilute sulphuric acid and then treated with 10% acetic acid to improve adhesion. Gelatin will not adhere well to plastic surfaces (a property which can be used to produce separated gelatin films) unless the surface is first treated with a "subbing" solution [MCC73]. The substrate can then be coated either by spinning or dipcoating [BRA69], the doctor blade method [SJ081], casting [MCC73] or
simply by pouring gelatin onto a warm, levelled glass plate [CUL82]. Curved surfaces can also be coated by film transfer [MCC73].

The conditions under which the plate is then dried are critical to the mechanical properties of the gelatin film. If the drying temperature is above the melting point of the gel the molecular structure will be amorphous. If the drying temperature is very low the structure will consist of many small crystallites. Films dried under either of these conditions will have poor mechanical properties and will tend to become noisy during dehydration. There is an optimum drying temperature just below the melting point at which large and perfect crystallites are formed and films dried in these conditions will produce clearer holograms [CHA71].

The coating and drying technique may induce stresses in the film which can cause peeling and in some instances are capable of tearing glass slivers out of the substrate. These problems can be reduced by careful control of the drying process [DUN85] and by avoiding rapid changes in humidity. When the film is processed in aqueous solutions any stored stresses will be relieved and distortion may result. To avoid this the gelatin can be kept under high humidity conditions for a period before sensitising and exposure to allow these stresses to relax.

2.5 BIAS HARDNESS.

The final level of modulation and noise in the hologram is dependent on the overall hardness or bias hardness of the gelatin. If the hardness is low, high modulation will be achieved but there will be an increase in the level of scatter due to formation of small cracks or microfractures. In extreme cases the gelatin can become totally opaque, particularly in unexposed areas, demonstrating dramatically the hardening
effect of the exposure. Increasing the overall hardness reduces the final modulation but also reduces the level of scatter.

To optimise the modulation whilst keeping the noise level low, the gelatin hardness, exposure and the final processing temperatures must be manipulated. Better results appear to be obtained by using reasonably hard gelatin with high exposures and moderate processing temperatures rather than trying to use soft gelatin, low exposures and high temperatures.

The critical gelatin hardness is that of the film directly before the final wash and dehydration stage of the processing procedure. At any time before this stage, either before or after exposure, the hardness can be adjusted chemically. To maximise the number of carboxyl sites available to the Cr\(^{3+}\) ions it is desirable to use organic hardeners such as formaldehyde which only act on amino sites \([KOS65, MAZ82]\):

\[
\text{(CHAIN)}—\text{NH}_2 + \text{CH}_2\text{O} + \text{NH}_2\text{—(CHAIN)} \rightarrow \\
\text{(CHAIN)}—\text{NH—CH}_2\text{—NH—(CHAIN)} + \text{H}_2\text{O}
\]

This maximises the potential hardness differential whilst allowing the overall hardness to be set at an adequate level. There will also be contributions to the overall gelatin hardness from the initial baking, the dark reactions, and the uniform component of the exposure.

It is interesting to note that hardening the gelatin whilst wet and swollen causes it to resist shrinking to its original thickness during dehydration whereas hardening in the dry state, by baking or by exposure, causes it to resist swelling during processing. As a result it is possible to control the thickness change during processing (and therefore the replay wavelength) by altering the hardening method.
Coleman and Magarinos were able to induce a change in replay wavelength from 514.5 nm to 680 nm using this technique [COL81].

2.6 SENSITIZATION

Ammonium dichromate is the most popular sensitiser as it has a high solubility in water and can therefore be incorporated in gelatin films to high concentrations, up to 30% by weight using sensitiser solutions of 20 g/litre to 100 g/litre. It is also one of the most sensitive dichromates. This could be because ammonium dichromate solutions are typically pH 4.5 and hence there is a ready availability of $H^+$ ions for the photochemical reaction.

If the dichromate concentration is increased, the sensitivity will be higher. However, absorption at the recording stage will also increase and the recording beams will be attenuated, particularly at shorter wavelengths. In thick gelatin films the absorption could cause the effective thickness of the hologram to be smaller than the physical thickness of the gelatin and therefore the sensitiser concentration should be chosen to suit the thickness in use.

The ammonium dichromate occupies an appreciable volume in the gelatin matrix and affects the dried film thickness and refractive index. During processing almost all the dichromate is washed out of the gelatin and these effects are removed. Because of this, the sensitiser concentration has a considerable affect on the replay Bragg condition, and high concentrations result in a reduction of the shift to longer wavelengths that normally occurs during processing.

After sensitisation the plates were dried in rapidly circulating dry air at 20°C. for at least 4 hours. Draining the plates vertically for prolonged periods can lead to a concentration gradient of dichromate, particularly along the bottom edge of the plate. From the time of
sensitisation, to the removal of the chromium compounds during processing, a dark reaction takes place. This involves a slow reduction of the dichromate ion in the absence of light (via chromic acid) which reduces the useful shelf life of the plate. The rate of reaction is increased by increasing humidity, pH and temperature. Hence the plates should be used within a reasonable period and should be stored in cool conditions at low humidity (below 40%). The effect of the dark reaction is identical to a uniform exposure and the bias hardness of the gelatin is increased.

After exposure this dark reaction continues in addition to a second dark reaction which occurs because the \( \text{Cr}^{3+} \) ions created in the exposed regions do not necessarily form crosslinks immediately and may exist for some time in an intermediate ionic state. The dark reaction before exposure decreases the potential modulation whilst the dark reaction after exposure can increase the modulation by a factor of 2 over two days [CHA79]. For consistent results the environmental conditions must be carefully controlled and the delay between sensitisation and exposure, and the delay between exposure and processing, should be kept constant.

2.7 EXPOSURE.

Dichromated gelatin is most sensitive to blue and ultraviolet wavelengths but the sensitivity falls to zero above 580 nm. The exposure required varies considerably for different gelatins and different processing methods but is typically \( 200 - 800 \text{ mJ/cm}^2 \) at 514.5 nm and 20 - 100 \( \text{mJ/cm}^2 \) at 488.0 nm. Dye sensitisation can be used to extend the sensitivity to red wavelengths [GRA73, KUB76] but the concentration of the dye must be high, leading to considerable absorption, and the pH must be carefully controlled to maximise sensitivity [CHA86].
At high exposures the recording response of dichromated gelatin becomes non-linear and saturates. It is interesting to note that at the saturation energy there are still many unconverted dichromate ions available in the exposed regions [CHA79]. This suggests that the saturation level is determined by the availability of active sites in the gelatin and not by the number of chromium ions available for the photochemical reaction. A consequence of the non-linear behaviour is the generation of spatial harmonics and non-sinusoidal grating profiles. This results in multiple K-vectors within the grating and, in the case of multiple exposures, intermodulation effects (Sec. 3.6).

The sensitivity and saturation level are variable and are dependent on the sensitisier concentration, pH, humidity, and the source and history of the gelatin. The sensitivity can be increased by the addition of mild reducing agents such as hydroquinone or by lowering the pH, but both these methods increase the rate of the dark reaction.

The relative humidity at exposure can affect the final replay wavelength of the hologram because the gelatin absorbs water and swells if the humidity rises above 40% R.H. Humidity also affects the rate of the dark reaction and therefore controlling the environmental conditions at this stage is essential for consistent results.

During exposure a brown colouration develops, due to the formation of the Cr ions, which increases the level of absorption and energy is absorbed which does not contribute to the photochemical reaction. This dynamic absorption reduces the effective exposure and can alter the fringe contrast at different depths in the gelatin, depending on the recording geometry and film thickness. Because of this it may be advantageous to operate in the saturated region to obtain a more uniform modulation. As the exposure is not uniform with depth the final degree
of swelling or shrinkage is not uniform. The effect this may have on the uniformity of the grating is investigated in Chapter 4.

The exposure energy used does not only determine the final modulation - the hardening effect of the exposure reduces the swelling which occurs during processing and therefore the replay Bragg angle and wavelength.

2.8 PROCESSING.

The processing procedure used for the holograms recorded in this thesis is similar to that described by Chang and Leonard [CHA79] and is shown in Table 2.1. For some holograms changes were made which are discussed separately where relevant.

TABLE 2.1 STANDARD PROCESSING PROCEDURE.

<table>
<thead>
<tr>
<th>STEP</th>
<th>TEMP.</th>
<th>TIME.</th>
<th>PROCESSING SOLUTIONS (all 200 ml).</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>20°C.</td>
<td>5 min.</td>
<td>Sensitise raw gelatin plate in 2% Ammonium Dichromate + 1/200 Photoflo (wetting agent).</td>
</tr>
<tr>
<td>2.</td>
<td>20°C.</td>
<td>4 hr.</td>
<td>Dry in rapidly circulating dry air.</td>
</tr>
<tr>
<td>3.</td>
<td></td>
<td></td>
<td>Expose at 514.5 nm.</td>
</tr>
<tr>
<td>4.</td>
<td>A°C.</td>
<td>2 min.</td>
<td>Wash off index matching fluid with methanol.</td>
</tr>
<tr>
<td>5.</td>
<td>A°C.</td>
<td>20 min.</td>
<td>0.5% Ammonium Dichromate solution.</td>
</tr>
<tr>
<td>6.</td>
<td>A°C.</td>
<td>20 min.</td>
<td>25% Agfa Structurix fixer + 1% Acid Hardener.</td>
</tr>
<tr>
<td>8.</td>
<td>B°C.</td>
<td>20 min.</td>
<td>50% Propanol.</td>
</tr>
<tr>
<td>9.</td>
<td>B°C.</td>
<td>20 min.</td>
<td>100% Propanol.</td>
</tr>
<tr>
<td>10.</td>
<td>40°C.</td>
<td>12 hr.</td>
<td>Dry in rapidly circulating hot air.</td>
</tr>
<tr>
<td>11.</td>
<td>20°C.</td>
<td></td>
<td>Coverplate using optical quality epoxy resin.</td>
</tr>
</tbody>
</table>
2.8.1 Adjusting bias hardness.

The first aim of processing is to remove excess chromium compounds and to adjust the final bias hardness. In this particular process this is done by using a hardening fixer on gelatin which contains a small concentration of ammonium dichromate. The fixer reduces the dichromate to the trivalent state which then increases the uniform bias hardness of the gelatin. After this stage the gelatin is no longer light sensitive and processing can proceed under normal lighting conditions.

2.8.2 Swelling.

The hologram is then washed to remove all chemicals from the gelatin before dehydration, which would otherwise crystallise and cause scatter. For transmission holograms it should now be possible to see considerable evidence of diffraction in the water bath due to the differential swelling of the gelatin in the exposed areas. In the case of reflection holograms it is usually not possible to see any diffraction because the swelling has a strong affect on the replay wavelength. The temperature of the first stages of the process (A°C.), which will be referred to as the initial processing temperature, is not critical and was typically 25°C. However, the temperature of the water bath and the propanol baths (B°C.) has a strong affect on the final modulation and can be adjusted to suit the thickness in use. This temperature will be referred to as the final processing temperature in following chapters.

The temperature and pH of the water bath control the degree of swelling of the gelatin and therefore also affect the replay Bragg condition. Increasing temperature and pH results in increased swelling and therefore a shift to longer wavelengths and increased modulation.

2.8.3 Dehydration.

The gelatin is then dried rapidly using propan-2-ol. This drying process must be done rapidly because if the water leaves the gelatin
slowly the gelatin will shrink to its initial, uniform thickness and refractive index. The propanol does this by replacing the volume occupied by the water in the gelatin matrix. However, as the propanol can only enter the gelatin from one surface the process takes place more rapidly on the emulsion side than it does on the glass side of the gelatin. As a result of this, the fringe spacing and modulation will not be uniform with depth and dichromated gelatin gratings are usually chirped. The effect of chirp on bandwidth is investigated in Chapter 4.

It is possible to control the level of chirp and therefore the bandwidth of the hologram by altering the overall drying rate [MCG80, CUL82]. If wide bandwidth is required a single hot 100% propanol bath is used. However, if narrow bandwidth is required then the drying is done at 20°C. in several stages with the concentration of propanol increasing steadily at each stage. This allows time for the propanol concentration to change uniformly throughout the gelatin and the grating produced is more uniform. Less modulation is achieved using the narrowband process but the level of noise or scatter is also reduced.

2.8.4 Final drying.

After removal from the propanol the drying method can have a strong affect on the modulation and uniformity. The hologram is very sensitive to humidity at this stage and the surrounding air must be as dry as possible to prevent loss of modulation, particularly since the evaporation of propanol may cause condensation.

Meyerhofer suggested that the hologram could be dried by raising it out of the propanol slowly while directing hot air along the interface between the plate and the liquid [MEY72]. Another method, which appears to produce high modulation, is to place the hologram on a hotplate, glass side downwards, at any temperature below the melting point of the gelatin. However, the method adopted for the holograms presented in this
thesis was to place the hologram in a cabinet of rapidly circulating dry air at 40°C. This was found to produce more reproducible and uniform results than other methods, although the level of modulation achieved may be smaller.

2.8.5 Anomalous effects.

In some cases a large level of scatter is found in the processed hologram. It is usually the result of an inadequate bias hardness or exposure, and high processing temperatures. As the processing temperature is increased the level of modulation and scatter rises evenly but sometimes a sudden transition to a milky, opaque state occurs. This is thought to occur because the stresses in the gelatin reach the limit of its physical strength. If sufficiently hardened this transition may not occur below the melting point of the gelatin.

Another similar related phenomenon has been seen by the author and by McGrew [MCG80]. When using low exposures, and therefore soft gelatin, the processed hologram can sometimes develop a patchy appearance. The hologram is divided into sharply defined areas which have two distinct characters. Some areas replay at a wavelength longer than the recording wavelength and have a low level of scatter. The other areas replay at a wavelength shorter than the recording wavelength and have a higher level of scatter (This could possibly be replay of a second harmonic or a sidelobe of the main response of the grating which has shifted into the infrared). Why this division occurs, and why the boundary between the areas is so sharp is not understood but it suggests that there is a sudden transition in the properties of the gelatin under particular conditions of temperature and stress.

In general the processing procedure results in a gelatin layer which is swollen relative to its original, unsensitised state and which has a lower refractive index. The final replay Bragg angle and
wavelength of the hologram will depend both on the changes due to sensitisation and the changes due to processing which are investigated in detail in Chapter 3.

2.9 POSTPROCESSING.

2.9.1 Baking.

The processed gelatin layer is usually swollen relative to the thickness of initial raw plates before sensitisation. If the hologram is baked at moderate temperatures it will shrink, with little loss of modulation, allowing the replay wavelength to be adjusted. Higher temperatures cause a loss of modulation, in addition to shrinkage. Chang and Leonard found that if there are strongly modulated fringes perpendicular to the substrate the gelatin may resist shrinkage and the baking time is extended [CHA79].

2.9.2 Reprocessing.

One useful property of dichromated gelatin holograms is the ability to adjust the hologram characteristics by repeating the swelling and dehydration processes until the desired efficiency, bandwidth and tuning are achieved [CHA76]. This is possible because the crosslinked structure of the gelatin is quite stable and is unaffected by the swelling and dehydration processes, unless excessive temperatures are used. The stability of the cross-linked structure is such that it is quite possible to re-sensitise and re-expose a fully developed hologram and reprocess it to produce a multiple grating.

To reduce the modulation of a hologram the bias hardness is increased before reprocessing. To increase modulation the temperature and the pH of the water bath are increased so that the gelatin is more swollen before dehydration. The bandwidth can also be adjusted during reprocessing by changing the dehydration rate. However, any change to
the processing procedure never has a unique effect and there always is some interaction between modulation, tuning and bandwidth.

Continual reprocessing of a hologram eventually causes some degradation of the gelatin which reduces the modulation that can be achieved and increases the level scatter and opacity.

2.9.3 Stability.

Unless dichromated gelatin holograms are dried fully and sealed against moisture they will deteriorate. This can be detected as a shift to shorter wavelengths, as the gelatin returns to its original thickness, accompanied by a loss in efficiency. To prevent this it is necessary to thoroughly dry the hologram either in a vacuum or by baking in dry air. It can then be sealed by cementing a glass coverplate over the emulsion with an epoxy resin. The resin used must be impervious to water and must be kept sealed during storage to prevent it absorbing water before use. If this is done carefully the holograms will be very stable, should stand temperatures up to 90°C. and will be unaffected by radiation, including ultraviolet light [SOS70].

2.10 SUMMARY.

The properties of dichromated gelatin have been the subject of considerable attention and the initial problems of reproducibility and scatter have been eliminated. It is common for diffraction efficiencies of 98% to be reported and a similar performance has been achieved in the holograms presented in this thesis. However, there are several features of the behaviour of dichromated gelatin which remain inadequately understood:

(1) It is common for authors to discuss thickness changes which have been calculated from the observed change in the Bragg angle during
processing. However, changes in the average refractive index, which will also affect the Bragg angle, are usually ignored. A thorough study of processing changes would make independent measurements of both changes as far as possible and correlate these with the observed change in the Bragg angle.

(2) The bandwidth of dichromated gelatin holograms is often found to be wider than would be expected for the physical thickness of the gelatin in use. Although some suggestions have been made as to why this disparity may occur, no accurate measurements or analysis of this behaviour has been reported and there has been no thorough investigation of the effect of different processing techniques.

(3) Finally, all references to real-time effects in the material have assumed that a small refractive modulation is responsible without presenting any evidence for this assumption, and no comprehensive study has been made of these effects.

The aim of the following three chapters is to investigate these particular features of dichromated gelatin.
CHAPTER 3.

THE RECORDING CHARACTERISTICS OF DICHROMATED GELATIN.

An ideal holographic recording material would be one which could accurately record and reproduce the spatial intensity variation of an interference pattern as a variation in refractive index and which would not perturb the incident light during recording.

The first of these criteria requires a linear recording characteristic, i.e. that refractive index changes should be proportional to exposure energy. For real recording materials the recording response will only be approximately linear and this will be over a limited exposure range. Real recording materials also suffer changes in thickness and average refractive index during processing which will have a strong affect on the replay Bragg condition of the final hologram. Before they can be utilised successfully detailed information about their recording properties and limitations must be known.

It is the aim of this chapter to determine the recording characteristics of dichromated gelatin and to examine some of the behaviour which results from the non-linearity that is found. The second criterion, the perturbation of the recording beams, will be investigated in Chapter 5.
3.1 MEASUREMENT OF THICKNESS AND REFRACTIVE INDEX.

In order to analyse the holograms recorded in this chapter it is necessary to establish the thickness and average refractive index of the gelatin at recording and at replay. It is possible to find the thickness and refractive index of thin silver halide emulsion layers by using the fact that at high angles of incidence these layers behave like an etalon [CO084, OWE82]. Modelling the periodic variation in the intensity of the transmitted and reflected beams with angle enables the thickness and refractive index to be found. This method relies on the fact that the refractive index of the silver halide emulsion (1.66 at 514.5 nm) is significantly higher than the glass substrate (1.515) or the index matching fluid (I.M.F.), di-n-butylphthalate (1.497). Therefore there is significant reflection at the glass/emulsion and emulsion/I.M.F. boundaries. However the technique was found to be unsatisfactory in most circumstances for processed dichromated gelatin layers because the refractive index (1.46 - 1.49) is too close to that of the glass and the I.M.F. The analysis of the interaction between the weak reflections becomes complicated because they are of the same order of intensity as a third reflection from the glass/I.M.F. boundary.

3.1.1 Thickness measurement.

Because of these problems a technique based on the Michelson Interferometer was developed (Fig. 3.1) to measure the physical thickness of the gelatin films. This uses the gelatin surface as one of the mirrors in the interferometer and an unsilvered optical flat for the other. Both have a similar reflectivity and therefore fringe contrast is high although the level of the reflected light is low. The interferometer is illuminated by a green filtered mercury lamp (\(\lambda = 546.1\) nm) and a white light source. A small slot is cut into the gelatin surface down to the glass substrate and the interferometer adjusted to
RECORDING CHARACTERISTICS OF DICHROMATED GELATIN.

Fig. 3.1. Thickness measurement method for gelatin films using a Michelson Interferometer.

Fig. 3.2. Thickness of sensitized gelatin, relative to the initial unsensitized gelatin thickness, against the concentration of the sensitizing solution.
give straight white light fringes across this slot. The thickness can then be determined by counting the number of green fringes between the white light maxima on the gelatin surface and that on the glass surface. This can be done to within ± 1/10th of a fringe and therefore measurements are accurate to about 0.03 µm.

Measurement of the thickness difference between exposed and unexposed areas does not require a slot to be cut in the hologram. The fringes are simply aligned across the boundary of the exposed region and an offset between the white light maxima is then clearly visible.

A similar technique was used by Case et al [CAS76] for measuring changes in optical thickness using light transmitted through the hologram rather than light reflected from the surface.

3.1.2 Refractive index measurement.

Refractive index measurements were made using an Abbe Refractometer. This instrument measures the refractive index of a material by determining the critical angle of total internal reflection. This is done by aligning cross-wires by sight on the borderline between light and dark regions in the field of view which occur at the critical angle. In ideal conditions it is possible to achieve an accuracy of 0.0002. When making these measurements it should be remembered that it is the surface refractive index that is being found and that refractive index may change with depth. Also the sharpness of the borderline, and therefore the accuracy, is determined by the uniformity of the refractive index.

During measurements on sensitized but unprocessed gelatin layers a sharp borderline was seen. However, the borderline is slightly blurred for processed layers which have been uniformly exposed and totally blurred for processed holograms. This is because for holograms the surface refractive index fluctuates rapidly. For this reason it is not
possible to use this method to measure the average refractive index of processed holograms. However, if we know the angles of the recording beams, the refractive index at recording and the thickness change between recording and replay it is possible to deduce the average refractive index at replay from the Bragg angle of the hologram.

All measurements of thickness and refractive index were made at 20°C (±1°C) and 40% (±2%) relative humidity. Refractive index measurements were made at 514.5 nm.

3.2 RESPONSE TO A UNIFORM EXPOSURE

The recording of a planar reflection hologram involves exposing the photosensitive material to light which has a sinusoidal spatial variation and a uniform average level. In this section the effect of a uniform exposure on the average refractive index and the thickness of the gelatin is investigated.

3.2.1 Measurements at recording.

All measurements in this section were made on plates cut from a single 20 cm by 25 cm gelatin plate for continuity. The thickness of the raw gelatin was measured at all positions exposures were to be made and was typically 18.2 µm. The average refractive index of the raw gelatin did not vary significantly and was 1.548.

The gelatin plates were sensitized following the procedure described in Chapter 2 using 2%, 5% or 10% solutions of ammonium dichromate (2% = 20 g/litre). Measurements were then made of thickness and refractive index, and the results are shown in Fig. 3.2 and Fig. 3.3 respectively. The points at 0% are values measured for the initial, unsensitized gelatin and are included for comparison.

Fig. 3.2 shows that the thickness of the gelatin layer is affected by the concentration of ammonium dichromate used in the sensitizing
Fig. 3.3. The refractive index of unexposed, unprocessed, sensitized gelatin against the concentration of the sensitizing solution. The point at 0% corresponds to the refractive index of the initial, unsensitized gelatin.

Fig. 3.4. The refractive index of processed gelatin films against exposure for 3 different concentrations of sensitizing solution.
solution. This implies that there is a considerable amount of ammonium dichromate within the gelatin. The fact that the thickness increase is small at low concentrations may mean that initially the ammonium dichromate fills some vacant space within the gelatin matrix.

Fig. 3.3 shows that the refractive index of the sensitized layer is also affected by the concentration of the sensitizing solution. This will mean that, for a particular external recording angle and wavelength, the recording angle and the propagation constant in the gelatin will depend on the concentration used.

The sensitized plates were then given a range of exposures to a uniform beam of light at 514.5 nm, incident at an external angle (in air) of 30°. Boundary reflections from the rear of the plate were eliminated by index matching to a neutral density filter. If this was not done a weak reflection hologram would be recorded. The plates were then processed by the procedure described Chapter 2.

3.2.3 Measurements after processing.

The refractive index of the gelatin after processing is plotted against exposure in Fig. 3.4, for the three different sensitizer concentrations. The refractive index changes with exposure are greatest for 10% sensitization and least for 2%. It is interesting to note that the refractive index for zero exposure appears to be almost independent of the sensitizer concentration and is much less than both the unsensitized and sensitized gelatin, but that differences appear with increasing exposure.

The thickness after processing relative to raw gelatin appeared to be very similar for the three sensitizer concentrations. However, we are most interested in the thickness after processing relative to the thickness after sensitization, i.e. the thickness change between recording and replay. This is shown in Fig. 3.5. The graph shows that,
RECORDING CHARACTERISTICS OF DICHROMATED GELATIN.

Fig. 3.5. Thickness of a processed gelatin film, relative to the thickness of the sensitized gelatin, against exposure for three different sensitizer concentrations.

Fig. 3.6. The refractive index of processed gelatin against relative thickness (relative to initial unsensitized gelatin).
for low sensitizer concentrations, processed holograms will be considerably swollen relative to recording. However for high sensitizer concentrations the thickness change will be smaller and there may even be shrinkage relative to exposure. For all sensitizer concentrations the thickness after processing appears to reduce with increasing exposure. This presumably results from the hardening of the gelatin during exposure.

In general, changes in average refractive index and thickness between recording and replay will affect the Bragg angle at replay or, for a fixed replay angle, the replay wavelength. The measurements in this section have shown that both these parameters change significantly with the concentration of the sensitizer used and also to a lesser degree with exposure used. This is in agreement with the observation of McGrew that the sensitizer concentration may be used to 'tune' dichromated gelatin holograms to a particular replay angle or wavelength [MCG80].

3.2.4 A model for the refractive index changes.

In Chapter 2 it was suggested that the refractive index modulation in dichromated gelatin holograms results from density changes induced during processing. It is not surprising therefore that a change in the refractive index is found to be accompanied by a change in thickness.

The processed gelatin is transparent and almost colourless and therefore it can be assumed that only small amounts of chromium compounds are incorporated in the gelatin by the exposure and processing. Because of the drying process and low humidity used it is unlikely that significantly more water is present in the gelatin after processing. Therefore it is likely that refractive index changes are predominantly due to density changes. A simple relationship between refractive index $n$ and density $D$ can be obtained from the relationships
\[ n = \left[ 1 + \chi_e \right]^{\frac{1}{2}} \]  
\[ \chi_e \propto N_0 \]  

where \( \chi_e \) is the susceptibility and \( N_0 \) is the density of dipole moments.

This suggests that

\[ n = \left[ 1 + aD \right]^{\frac{1}{2}} \]  

where \( D \) is the density of the gelatin and \( a \) is a constant. The gelatin used in dichromated gelatin holography is sufficiently hardened that very little dissolves during processing. This is confirmed by the fact that if processed gelatin is soaked in water and then dried it returns to its original thickness [MEY72]. Therefore density changes must be due to volume changes and not to a loss of gelatin. Since the gelatin is attached to a rigid glass plate and is only free to expand in one direction, density is inversely proportional to thickness. Hence

\[ n = \left[ 1 + \frac{k}{t} \right]^{\frac{1}{2}} \]  

where \( t \) is the thickness of the gelatin and \( k \) is a constant.

To test this hypothesis, refractive index is plotted against thickness in Fig. 3.6 from the measurements made on gelatin sensitized with 2\% ammonium dichromate. The theoretical curve was generated using (3.4) and \( k = 1.406 \). The simple model appears to be in reasonable agreement with the observed behaviour. In particular raw gelatin and unexposed, processed gelatin give very similar values of \( k \). However small differences appear as exposure is increased. This could be because the exposure does lead to the introduction of some ammonium dichromate or water through the crosslinking reaction.
The significance of these results is that similar behaviour will be seen in the average refractive index and thickness of processed holograms and a similar mechanism must be the source of the refractive index modulation.

3.2.5 Dispersion.

Measurements were also taken with the Abbe Refractometer at a number of wavelengths to investigate the dispersive properties of the recording material. Fig. 3.7 shows refractive index against wavelength for raw gelatin, sensitized gelatin and processed gelatin. The degree of dispersion appears to be similar in all three cases although the refractive index is not the same. We might expect the average refractive index and the refractive index modulation of processed holograms to have a similar degree of dispersion as these uniformly exposed layers. The refractive index of the index matching fluid, di-n-butyl phthalate, was also measured at different wavelengths as this will be required in later parts of this thesis.

3.3 RECORDING RESPONSE FOR REFLECTION HOLOGRAMS.

The recording properties of transmission holograms in dichromated gelatin have been investigated by several authors [FIL71, CHA79]. This study concerns reflection holograms which in general have higher spatial frequencies. Two gelatin thicknesses were used in the experiments: $\approx 5 \mu m$ gelatin films (sets A and C) and $\approx 20 \mu m$ films (sets B and D). These can both be processed effectively by the same procedure and therefore a comparison of the results for the two thicknesses is valid. Thicker gelatin layers require different processing techniques which are discussed in Chapter 4.
Fig. 3.7. Refractive index against wavelength for (a) unsensitised gelatin, (b) sensitised gelatin, and (c) processed gelatin.
3.3.1 Experimental method.

The approach of earlier authors has been to record planar gratings at different exposures in the material and from measurements of efficiency deduce the refractive index modulation obtained. In the case of transmission holograms the high modulation which can be achieved in dichromated gelatin does not present any experimental difficulties. As modulation is increased, the efficiency first rises to a maximum and then falls back to zero. This process is repeated cyclically. However, in reflection holograms the efficiency rises to a maximum but does not then fall with a further increase in modulation. The intensity of the transmitted 0th order falls to zero and remains there. The only effect of increasing the modulation further is to slowly increase the bandwidth of the hologram. This behaviour will be referred to as 'overmodulation'. As a result of this it is difficult to deduce accurately the modulation at these high values. However this information is important because much of the information related to the non-linearity of the material is to be found at high values of modulation and because we may wish to use this high modulation to record many efficient superimposed gratings.

To overcome this problem two different methods of investigating the recording properties have been used and the results of each are then compared. The first method is the conventional approach which is to record a series of single exposure planar reflection holograms at different exposures. The second method is to record a set of identical holograms each of which is given the same small exposure. These holograms are then exposed to a uniform single beam of light for a range of different exposures. From the efficiency of the weak holograms which result it is possible to find the rate of change of modulation with respect to exposure as a function of exposure. Then, by integration, the modulation against exposure characteristic can be found. Both of these
methods were applied to both gelatin thicknesses. The first method was used in sets A and B and the second method in sets C and D.

3.3.2 Recording.

The holograms were all recorded using the technique shown in Fig. 3.8. The sensitized plate is held in contact with a high reflectivity, front surface mirror by a thin layer of index matching fluid which reduces spurious reflections. The advantages of this method are that it is very stable and immune to the effects of air currents. It also has the advantage that it requires only one collimated beam and therefore uses the available light efficiently. One possible disadvantage is that the beam ratio cannot be altered, but this is irrelevant in these experiments as we require this to be constant for all the exposures. To expose the second set of holograms to a single beam the mirror is replaced by a neutral density filter. The external recording angle for all exposures was 30°.

One of the major problems associated with dichromated gelatin is that of reproducibility. To improve this as much as possible recordings were made in an environmentally controlled laboratory at 20°C and 40% relative humidity. The nine holograms in each set were recorded on one sensitized plate, 80 mm by 80 mm. Thus they were sensitized, exposed and processed in close proximity. This eliminated many of the problems which might otherwise have caused a lack of reproducibility within a set. The two plates required of each thickness were both cut from the centre region of a single 20 cm by 25 cm plate to ensure the quality and thickness of gelatin was consistent.

3.3.3 Processing.

The holograms were processed following the standard procedure described in Chapter 2 using an initial processing temperature, $A = 20^\circ C$ and a final processing temperature, $B = 25^\circ C$. Considerable effort was
Fig. 3.8. Recording arrangement for unslanted planar reflection holograms.

Fig. 3.9. Apparatus used to measure the angular response of the holograms at a selected wavelength (514.5 nm).
put into eliminating any differences in conditions and timescales during the sensitization, recording and processing of successive plates.

The thickness of the sensitized plate before exposure, and the relative thickness of each exposure after processing, was measured using the technique described in Sec. 3.1.

The absorption constant of the sensitized gelatin was found by measuring the transmission at 514.5 nm. This absorption will cause attenuation of the recording beams. The reflectivity of the front surface mirror was estimated by measuring the intensity of the light reflected back through the gelatin during recording. From this an average value for the intensity of the two recording beams and the beam ratio was calculated. The increase in absorption which occurs during exposure in dichromated gelatin is small at low exposures but may have had some effect in the high exposure holograms.

After thickness measurements were completed a glass plate was glued to the gelatin surface using 'Epotek' optical quality epoxy resin to protect the holograms from moisture.

3.3.4 Measurement of the angular characteristics of the holograms.

To characterize the holograms the variation in transmitted intensity with angle was measured at a wavelength of 514.5 nm using the apparatus in Fig. 3.9. The hologram was surrounded in a tank of the same index matching fluid used at recording. This apparatus was controlled by a microcomputer which was used to read and store all measurements. Measuring the power in the diffracted beams would not yield any extra significant information because the absorption is very low and the power in higher diffracted orders is small (see Fig. 6.5).

Several examples of these measurements for each set of holograms are shown in Figures 3.10 - 3.13.
TABLE 3.1. SUMMARY OF HOLOGRAMS RECORDED.

<table>
<thead>
<tr>
<th>SET</th>
<th>HOLOGRAM</th>
<th>THICKNESS (μm)</th>
<th>FIRST EXPOSURE ( \text{mJ/cm}^2 )</th>
<th>SECOND EXPOSURE ( \text{mJ/cm}^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>(two beam)</td>
<td>(single beam)</td>
</tr>
<tr>
<td>A</td>
<td>/1</td>
<td>5</td>
<td>50.0</td>
<td>-</td>
</tr>
<tr>
<td>A</td>
<td>/2</td>
<td>5</td>
<td>100.0</td>
<td>-</td>
</tr>
<tr>
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<td>5</td>
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<td>-</td>
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<td>400.0</td>
<td>-</td>
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<td>-</td>
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<td>-</td>
</tr>
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<td>A</td>
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<td>/9</td>
<td>20</td>
<td>25.0</td>
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</tr>
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Fig. 3.10. Examples of the angular response at 514.5 nm of 5 μm holograms recorded for method 1 (SET A) each with a theoretical fit (solid line) using coupled wave theory.
Fig. 3.11. Examples of the angular response at 514.5 nm of 20 μm holograms recorded for method 1 (SET B) each with a theoretical fit (solid line) using coupled wave theory.
Fig. 3.12. Examples of the angular response at 514.5 nm of 5 \( \mu \text{m} \) holograms recorded for method 2 (SET C) each with a theoretical fit (solid line) using coupled wave theory.
Fig. 3.13. Examples of the angular response at 514.5 nm of 20 μm holograms recorded for method 2 (SET D) each with a theoretical fit (solid line) using coupled wave theory.
3.4 THEORETICAL ANALYSIS.

3.4.1 Hologram characterisation.

The measured characteristic of each hologram was modelled theoretically using the analytical solutions to the two wave beta-value coupled-wave theory described in Chapter 1.

There are several parameters in these equations which can be defined by independent measurements made at recording and replay. The propagation vectors of the recording waves in the gelatin, $\rho$ and $\sigma$, can be found from the external recording angles, the recording wavelength, the external refractive index and the refractive index of the gelatin at recording. The $K$ vector of the grating at replay is defined in equations (1.2) and (1.4) using measured values of the thickness at recording and after processing.

All the other parameters can be found by matching the theoretical model to particular features of the experimental data. The average refractive index at replay, $n_0$, can be found from the position of the Bragg angle. As discussed in Sec. 3.1.2, $n_0$ cannot be measured independently using the Abbe Refractometer. However, since none of the other free parameters affect the position of the Bragg angle, $n_0$ can be found unambiguously.

The refractive index modulation, $n_I$, can be found from the efficiency of the hologram at the Bragg angle. The problem discussed in Sec. 3.3.1 is clearly seen in set B; Holograms B/3 to B/9 are all overmodulated. Exposures of $100 \text{ mJ/cm}^2$ and over lead to holograms which are almost 100% efficient but saturation does not appear until much higher exposures. It is possible to estimate the value of $n_I$ from the bandwidth of these holograms but it is difficult to obtain accurate results.
The average absorption constant, $\alpha_0$, can be found by matching the intensity of the transmitted light when the hologram is off-Bragg. Typically, the off-Bragg transmission of these holograms is about 97% after losses due to absorption and boundary reflections. Because the absorption constant is small the value of absorption modulation, $\alpha_1$, must also be small. The effect of adding $\alpha_1$ at these values was found to be negligible and therefore it was set to zero for all the calculations.

Several examples of theoretical curves and experimental data are shown in Figures 3.10 - 3.13. A consistent feature of all the graphs is that the bandwidth predicted by the theoretical model is always smaller than that found experimentally. The sidelobes of the holograms also differ considerably from those predicted by the theory. In particular they are asymmetric, the sidelobes at angles smaller than the Bragg angle always being stronger than those at angles larger than the Bragg angle. This behaviour is a particular characteristic of dichromated gelatin reflection holograms. Both of these differences are caused by the fact that refractive index modulation and fringe spacing are not constant but vary with depth in the gelatin. This will be investigated in greater detail in Chapter 4. As we are most interested in the relative value of $n_1$ as a function of exposure, the differences between the theoretical model and the experimental data can be tolerated.

3.4.2 Investigation of the non-linearity - Set A and B.

In both set A and B the modulation rises up to exposures of about $500 \text{ mJ/cm}^2$ and then falls slowly. The values of $n_1$ obtained in Sec. 3.4.1 are plotted against exposure for set A and set B in Fig 3.14 and 3.15 respectively. The exposure has been corrected for boundary losses and absorption. It can be seen that the saturation value of $n_1$ is slightly higher for the 20 $\mu \text{m}$ holograms (set B) than for 5 $\mu \text{m}$ holograms (set A) and that saturation occurs at similar exposures.
Fig. 3.14. Refractive index modulation against exposure for 20 \( \mu \text{m} \) holograms (SET B).

Fig. 3.15. Refractive index modulation against exposure for 5 \( \mu \text{m} \) holograms (SET A).
Chang and Leonard [CHA79] found that for dichromated gelatin transmission holograms the refractive index modulation could be modelled by the expression

\[ n_1 = M [1 - \exp(-E/D)] \]  \hspace{1cm} (3.5)

where \( E \) is the exposure, and \( M \) and \( D \) are constants for the particular gelatin, sensitizer concentration and processing used. This model may give reasonable agreement at low exposures but it cannot explain the observation that at high exposures the modulation, \( n_1 \), falls. The following modifications were made to improve the agreement above saturation.

First it was assumed that the change in refractive index as a function of position, \( n(\hat{p}) \), is given by

\[ n(\hat{p}) = N_0 \left[ 1 - \exp{\left( \frac{-E(\hat{p})}{E_0} \right)} \right] \]  \hspace{1cm} (3.6)

where \( E(\hat{p}) \) is the exposure as a function of position, \( \hat{p} \) is the position vector and \( N_0 \) and \( E_0 \) are constants. At exposure

\[ E(\hat{p}) = E_1 (1+r) + 2 E_1 r \cos(\hat{K} \cdot \hat{p}) \]  \hspace{1cm} (3.7)

where \( E_1 = I_1 t \), \( r = I_2/I_1 \), \( I_1 \) and \( I_2 \) are the intensities of the two recording beams, \( t \) is the exposure time and \( \hat{K} \) is the grating vector.

Substituting (3.7) into (3.6)

\[ n(\hat{p}) = N_0 \left[ 1 - v \exp{\left( z \cos(\hat{K} \cdot \hat{p}) \right)} \right] \]  \hspace{1cm} (3.8)

where \( v = \exp{\left( \frac{E_1 (1+r)}{E_0} \right)} \) and \( z = -2 r E_1 \).

Equation (3.8) can be expanded into a fourier cosine series

\[ n(\hat{p}) = n_0 + \sum_{m=1}^{\infty} n_m \cos(m\hat{K} \cdot \hat{p}) \]  \hspace{1cm} (3.9)

where the coefficients \( n_m \) are
\[ n_0 = A \left[ 1 - \exp \left( \frac{I_0(z)}{E_0} \left( \frac{E_1(1+r)}{E_0} \right) \right) \right] \quad (3.10) \]

\[ n_m = 2A \exp \left[ I_m(z) \left( \frac{E_1(1+r)}{E_0} \right) \right] \quad m = 1, 2, \ldots, \infty \quad (3.11) \]

where \( I_m(z) \) are Modified Bessel functions of order \( m \).

The model predicts that for a sinusoidal exposure an infinite series of harmonic gratings will be obtained. For this analysis we are most interested in the amplitude of the fundamental grating \( n_1 \). The free parameters we have in the model are \( N_0 \) and \( E_0 \). The best values of these parameters (listed in Table 3.2) were found by comparing the calculated values of \( n_1 \) with the experimental measurements, as shown in Fig. 3.14 and 3.15.

The model compares well with the experimental data including the behaviour at high exposures. The product of the two parameters \( N_0 \) and \( E_0 \) can be deduced unambiguously from the slope \( \frac{dn_1}{dE} \) at low exposures. However the individual values of \( N_0 \) and \( E_0 \) can be varied over a range of about 5% and still give reasonable agreement with the experimental data provided that their product remains the same.

3.4.3 Investigation of the non-linearity - Set C and D.

The principle of the second method is demonstrated in Fig. 3.16. A small two beam exposure, \( E_1 \) (in one beam), is made followed by a single beam exposure, \( E_2 \). From the modulation, \( n_1 \), which results we can find the rate of change of modulation with respect to exposure at an exposure of \( E_1 \)

\[ \frac{dn}{dE} = \frac{\Delta n}{\Delta E} \quad (3.12) \]

where

\[ \Delta n = 2 n_1 \quad (3.13) \]

\[ \Delta E = 2 E_2 \quad (3.14) \]
Fig. 3.16. The principle of the second method of determining the recording characteristic. A small two beam exposure is followed by a large single beam bias exposure.
\[ E_1 = E_s + E_2(l+r) \tag{3.15} \]

where \( r \) is the beam ratio of the two beam exposure.

The values of \( n_1 \) obtained in Sec. 3.4.1 are plotted against exposure for set C and set D in Figures 3.17 and 3.18 respectively. From Equation (3.12) and (3.13) \( \Delta n/\Delta E \) was found. Using numerical integration it is then possible to find \( n \) as a function of \( E \). This was done for both set C and D and the results are shown in Fig 3.19 and Fig 3.20.

We can now compare these results with the theoretical model used in Sec 3.4.2. Assuming the same characteristic (3.6) the best values of the constants \( N_0 \) and \( E_0 \) were found by comparing theoretical values of \( n_1 \) with the integrated experimental values. These are listed in Table 3.2 and the theoretical results are also shown in Figures 3.19 and 3.20.

<table>
<thead>
<tr>
<th>THICKNESS (( \mu m ))</th>
<th>METHOD</th>
<th>( N_0 )</th>
<th>( E_0 ) (mJ/cm(^2))</th>
</tr>
</thead>
<tbody>
<tr>
<td>SET A 5</td>
<td>1</td>
<td>0.068</td>
<td>( 7.1 \times 10^2 )</td>
</tr>
<tr>
<td>SET C 5</td>
<td>2</td>
<td>0.070</td>
<td>( 7.1 \times 10^2 )</td>
</tr>
<tr>
<td>SET B 20</td>
<td>1</td>
<td>0.085</td>
<td>( 5.6 \times 10^2 )</td>
</tr>
<tr>
<td>SET D 20</td>
<td>2</td>
<td>0.090</td>
<td>( 6.7 \times 10^2 )</td>
</tr>
</tbody>
</table>

3.4.4 Comparison of the two methods and the two thicknesses.

Comparison of the values of \( N_0 \) and \( E_0 \) in Table 3.2 shows that the two methods produce results that are in reasonable agreement and that the two gelatin thicknesses have quite similar recording properties.

It is interesting to note that \( N_0 \) (and therefore the maximum modulation that can be obtained) is slightly higher for 20 \( \mu m \) films than for 5 \( \mu m \) films. This difference must to be due to processing effects and
Fig. 3.17. Refractive index modulation against bias exposure for 5 \( \mu m \) holograms (SET C).

Fig. 3.18. Refractive index modulation against bias exposure for 20 \( \mu m \) holograms (SET D).
Fig. 3.19. Refractive index against exposure for 5 μm holograms (SET C) deduced by integrating values of dn/dE.

Fig. 3.20. Refractive index against exposure for 20 μm holograms (SET D) deduced by integrating values of dn/dE.
is contrary to the expectation that increased gelatin thickness would decrease the effectiveness of the processing solutions. It may be that there is loss of modulation at the gelatin/glass boundary or at the external gelatin surface; this would have a greater affect on the efficiency of thinner holograms. In the following Chapter the non-uniformity of the modulation will be explored in more detail.

The slightly lower value of $E_0$ in Set B is most likely due to errors resulting from the difficulties found in measuring $n_t$ for the highly efficient 20 μm holograms, which were described in Sec. 3.3.1.

An important feature of the results is that the maximum peak to peak modulation is considerably larger than the average refractive index change due to exposure which could be induced in the uniform single beam exposures measured in Sec. 3.2.3. This disparity can be explained by considering the mechanism by which modulation is obtained. In the uniform single beam exposures the exposure causes crosslinking which results in homogeneous forces during processing. However in the two beam exposures the rapidly varying hardness results in differential forces which causes a local transfer of gelatin and therefore a higher local refractive index change than can be induced in the bulk of the material.

In the following section it will be shown that the uniform component of the two beam exposures results in a change of thickness and average refractive index which is comparable to that found in the uniform single beam exposures.

3.5 OTHER FEATURES OF THE RECORDING MATERIAL.

3.5.1 Changes in replay angle with exposure.

All the holograms in this chapter were recorded with external angles of 30° which corresponds to 19.5° in the index matching fluid. Fig. 3.21 shows that the replay Bragg angle of the holograms is much
RECORDING CHARACTERISTICS OF DICHROMATED GELATIN.

Fig. 3.21. Replay angle against exposure for SET A and B measured in index matching fluid ($n_0 = 1.497$). The recording angle was 19.5° in this refractive index.

Fig. 3.22. Thickness change during processing against the uniform component of the exposure for 20 μm holograms (SET B). The thickness changes measured for uniform single beam exposures are included for comparison.
higher than the recording angle and that the angle falls with increasing exposure. This behaviour results from the different thickness and refractive index changes which have occurred for each hologram. These changes will be shown in the following sections to be similar to those seen earlier in the single beam exposures.

The replay angles of the 5 \( \mu \text{m} \) holograms (set A) are smaller than the replay angles of the 20 \( \mu \text{m} \) holograms (set B) because there is less swelling between recording and replay. One reason for this is that the concentration of ammonium dichromate within the gelatin is somewhat higher in the 5 \( \mu \text{m} \) films than in the 20 \( \mu \text{m} \) films (this is known because the absorption constant after sensitization was higher) but there may be other processing effects due to the thickness difference.

3.5.2 The variation in thickness change with exposure.

It is interesting to compare the thickness changes seen in the holograms in set B with those of the uniformly exposed gelatin layers in Sec. 3.2 which were from the same batch of 20 \( \mu \text{m} \) plates. For this purpose it is most appropriate to plot the thickness changes in set B against the uniform component of the spatially varying exposure, shown in (3.10) to be \( E_1(l+r) \). This has been done in Fig. 3.22. It can be seen that the thickness changes are of the same order but that after an initial rapid fall the thickness change in the holograms appears to saturate earlier than that in the uniformly exposed layers.

These observations are for the special case of an unslanted reflection hologram in which the gelatin is homogeneous in a plane parallel to the substrate and perpendicular to the direction of the thickness change. In all slanted holograms and in particular, unslanted transmission holograms, this is not the case and fringes of hardened gelatin (resistant to swelling) will tend to stiffen the layers and reduce these thickness changes [CHA79].
3.5.3 The variation in average refractive index with exposure.

In Fig. 3.23 the average refractive index \( n_0 \) of holograms in set B is compared with that of uniformly exposed gelatin. Again the data for set B has been plotted against the uniform component of the exposure. As in Sec. 3.5.2 the changes are of a similar order in each case but there are differences.

It should be noted that the values of \( n_0 \) for set B were deduced in Sec. 3.4.1 from measurements of initial and final thickness, initial refractive index and replay angle and that errors in these will have a cumulative affect on the accuracy of \( n_0 \), whereas the average refractive index of the uniform exposures was measured directly.

3.5.4 Scatter and absorption.

There are two sources of attenuation in the processed gelatin: absorption and scatter. However the contribution of each of these to the total loss cannot be seperated easily.

Absorption due to the presence of chromium in the layers can be seen as a slight green colouration in very thick gelatin layers but in the thicknesses used in this chapter colouration was negligible.

Scatter can often be seen as a milky opacity in holograms processed at high temperatures or where the bias hardness is inadequate.

The combined losses due to scatter and absorption are represented by the absorption constant \( a_0 \). The variation in this absorption constant with exposure for set B is plotted in Fig. 3.24. The absorption at high exposures is about double that at low exposures and losses vary between about 2.5% and 5.5%.

Noise gratings, which have a considerable effect in silver halide, have not been observed in these holograms because there is very little scatter at recording; this is fully discussed in Chapter 6.
RECORDING CHARACTERISTICS OF DICHROMATED GELATIN.

Fig. 3.23. The average refractive index against the uniform component of the exposure for 20 \( \mu \)m holograms (SET B) deduced from the replay Bragg angle.

Fig. 3.24. The absorption constant, \( \alpha_0 \), against exposure \( (E_1) \) for 20 \( \mu \)m holograms (SET B).
3.6 HARMONIC GRATINGS IN MULTIPLE EXPOSURE HOLOGRAMS.

The analysis in Sec. 3.4.2 predicts the existence of an infinite series of harmonic gratings (although the limiting effect of the spatial frequency response of the material has not been considered). Unfortunately it is not possible to replay any of these harmonics at optical wavelengths. For example, the second harmonic grating will not replay at wavelengths longer than 280 nm. It is usually possible to replay higher harmonics in transmission gratings [SLI85B] and in reflection holograms recorded at very high angles of incidence [HEA85]. However, in this Section it will be shown that harmonic gratings can also be found in multiple exposure reflection holograms.

3.6.1 Recording multiple exposure holograms.

Several gratings can be recorded in one hologram by making repeated exposures with different external recording angles using the apparatus shown in Fig. 3.8. Because there is an increase in the absorption of dichromated gelatin during exposure the effect of each successive exposure is reduced and the modulation achieved for each exposure falls as the number of previous exposures increases. However, if the sensitiser concentration and the total level of exposure is low this effect is small.

The angular response of a 16 \( \mu \text{m} \) reflection hologram, recorded with an exposure of 600 mJ/cm\(^2\) at 50° (in air) followed by an exposure of 300 mJ/cm\(^2\) at 25°, is shown in Fig. 3.25. This hologram was processed in a similar manner to the single exposure holograms but in order to increase the modulation available for the two gratings it was dried rapidly on a hotplate and not in warm air. Because of this the values of \( N_0 \) and \( E_0 \) will differ from those found earlier. The two large Bragg dips at the centre of the graph correspond to diffraction by the two expected fundamental gratings. However, there are also several other small but
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Fig. 3.25. The angular response of a sequential double exposure hologram at 514.5 nm.

Fig. 3.26. The theoretical modulation against exposure characteristic of the fundamental and intermodulation harmonic grating vectors. The vertical dashed line corresponds to the hologram in Fig. 3.25.
significant Bragg dips visible. At first the source of this additional diffraction was not understood but it was then found, from the angles of the incident and diffracted rays, that the Bragg dips correspond to diffraction by grating vectors which can all be expressed in the form \( \mathbf{k}_{n,m} = n\mathbf{k}_1 + m\mathbf{k}_2 \) where \( n \) and \( m \) are integers and \( \mathbf{k}_1 \) and \( \mathbf{k}_2 \) are the grating vectors of the two fundamental gratings. The spurious Bragg dips cannot be due to multiple diffraction by the two fundamental gratings, since the hologram is optically thick. The diffraction must therefore be due to harmonic intermodulation gratings which result from the non-linear recording characteristic of the recording material.

### 3.6.2 Theoretical analysis.

Assuming the same recording characteristic as before (Eqn. 3.6) it is possible to calculate the modulation which would be expected for both the fundamental gratings and the intermodulation harmonic gratings. This is shown in Appendix I. Using this analysis the theoretical modulation of all the gratings visible in Fig. 3.25 is plotted against the factor \( E_{50}/E_0 \) in Fig. 3.26, where \( E_{50} \) is the exposure at 50° (assuming the ratio of the two exposures is 2:1 and the beam ratio is 0.8). It can be seen that for each value of \( E_{50}/E_0 \) there is a unique ratio between the values of the modulation of the fundamental and harmonic gratings.

It is possible to determine the modulation of all the gratings which are visible in Fig. 3.25 using the single grating coupled wave theory used earlier. This is because the Bragg angles of the gratings are well separated, the angular bandwidth is small and hence, therefore, there is no interaction between the gratings. The value of the modulation for each grating is listed in Table 3.3. These values correspond closely to the vertical line drawn on Fig 3.26 and therefore the constants \( N_0 = 0.13 \) and \( E_0 = 7.0 \times 10^2 \) are obtained. Comparing these
values with those given in Table 3.2, the rapid hotplate drying process appears to increase the value of $N_0$ but has little affect on $E_0$.

**TABLE 3.3.** The modulation of the fundamental and harmonic gratings

<table>
<thead>
<tr>
<th>GRATING VECTOR</th>
<th>MODULATION, $n_1$</th>
<th>RATIO</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\hat{K}_1$</td>
<td>0.030</td>
<td>1.0</td>
</tr>
<tr>
<td>$\hat{K}_2$</td>
<td>0.017</td>
<td>0.57</td>
</tr>
<tr>
<td>$2\hat{K}_1 - \hat{K}_2$</td>
<td>0.004</td>
<td>0.13</td>
</tr>
<tr>
<td>$2\hat{K}_2 - \hat{K}_1$</td>
<td>0.0017</td>
<td>0.057</td>
</tr>
<tr>
<td>$3\hat{K}_1 - 2\hat{K}_2$</td>
<td>0.0009</td>
<td>0.03</td>
</tr>
</tbody>
</table>

3.7 SUMMARY.

The effect of a uniform exposure on the thickness and refractive index of the recording material has been investigated and a model has been proposed which relates the thickness change to the refractive index change. The uniform component present in two beam exposures has been shown to have a similar affect on thickness and refractive index, both of which affect the replay Bragg angle and wavelength of the hologram.

The non-linear recording characteristic of dichromated gelatin has been investigated by two different methods which have yielded similar results. A quantitative analysis has shown that the response of the recording material can be expressed in the form given in Equation (3.6) and the constants $N_0$ and $E_0$ have been determined. The recording characteristics of two different gelatin thicknesses have been compared and they have been found to be similar, with small differences.

Finally, the non-linear recording characteristic has been seen to lead to spurious harmonic intermodulation gratings in sequential double exposure holograms. The modulation of these gratings has been measured and compared with theoretical values obtained using the assumed form of the recording characteristic.
CHAPTER 4.

NON-UNIFORMITIES IN PLANAR REFLECTION GRATINGS.

In Chapter 3 it was seen that there are significant differences between the characteristics of planar dichromated gelatin reflection gratings and a theoretical model based on uniform coupled wave theory. These differences are thought to result from non-uniformities in the grating structure. The aim of this chapter is to investigate these non-uniformities and to extend the theoretical model to improve the agreement with the observed behaviour. The properties of gratings when replayed at wavelengths other than the recording wavelength are investigated and are compared with this extended model. The model is used to show that if narrow bandwidth is required a non-uniform refractive index modulation may be advantageous whilst non-uniform fringe spacing is undesirable. To demonstrate how these properties may be achieved the effect of different processing procedures on the grating non-uniformities are investigated.

4.1 INTRODUCTION.

The disagreement between uniform coupled wave theory and the properties of dichromated gelatin reflection gratings can be seen in Fig. 4.1 - 4.4. The angular response of several unslanted planar reflection gratings is compared with that predicted by uniform coupled-wave theory based on the measured gelatin thickness. It is clear that
Fig. 4.1. The angular response of a 5 \( \mu m \) planar reflection hologram at 514.5 nm compared with uniform coupled wave theory.

Fig. 4.2. The angular response of a 16 \( \mu m \) planar reflection hologram at 514.5 nm compared with uniform coupled wave theory.
Fig. 4.3. The angular response of a 50 μm planar reflection hologram at 514.5 nm compared with uniform coupled wave theory.

Fig. 4.4. The angular response of a 155 μm planar reflection hologram at 514.5 nm compared with uniform coupled wave theory.
the disagreement appears to be worse for thick gelatin layers but there are many common features. In particular the gratings have wider angular bandwidth than is predicted theoretically and there are also significant differences in the sidelobe structure: the sidelobes are asymmetric and are always larger at angles lower than the Bragg angle; the phase of the sidelobes is incorrect, and the 'zeroes' of the sidelobes have moved downwards. The asymmetry of the sidelobes is a common and recurring characteristic of dichromated gelatin holograms and has been observed by many authors [KUB79], [CUL82], [DUN85].

This behaviour occurs because the gratings are not uniform. The refractive index modulation, fringe spacing and average refractive index change with depth in the gelatin layer. There are several possible reasons why non-uniformities or chirp could occur. During exposure the recording beams may be attenuated due to absorption. Early evidence of the effect of absorption during recording was seen by Leith [LEI66] in experimental work on absorption gratings in silver halide emulsions. The effect can be quantified easily by measuring the absorption constant of the recording material. There may be more complex perturbations to the recording beams due to real time effects which are investigated in Chapter 5. Non-uniformities may also be introduced during the processing procedure. The gelatin layer is supported by an impervious glass substrate and processing solutions must diffuse throughout the gelatin layer from the exposed surface to reach all of the recording material. The most significant stage at which chirp could be introduced is during dehydration with propanol. The rate at which dehydration occurs has a significant affect on the efficiency and the replay Bragg angle of a hologram as a whole [MCG80]. It is therefore logical that if the dehydration rate is a function of depth there will be a localised variation in fringe spacing and modulation with depth. One author has
suggested that, because propanol can only penetrate gelatin in the presence of water, rapid drying of the exposed gelatin surface prevents propanol from reaching gelatin at greater depths and greatly reduces the rate of dehydration in these areas [MCG80]. The observation that different processing procedures have been found to result in different sidelobe structures and bandwidths is further evidence that processing is a major cause of chirp in dichromated gelatin holograms [MCG80], [CUL82].

The effect of chirp is usually an increase in bandwidth which can be advantageous in some applications. However, where narrow bandwidth is required, such as in reflection filters [DUN85] or head-up displays [CO077], chirp will be a problem. In Sec 4.2 it will be seen that particular variations in the refractive index modulation can reduce the bandwidth by reducing the strength of the sidelobes, if such a variation could be induced.

4.2 COUPLED WAVE THEORY FOR CHIRPED GRATINGS.

Previous theoretical analyses of non-uniform grating structures have concentrated on transmission holograms and on the effects of absorption during recording. This is because absorption has a strong affect on fringe amplitude in transmission holograms, for which recording materials are more readily available, and because the level of absorption or scatter in many of these materials is high. The effect of non-uniform modulation, but where fringe spacing and average refractive index are constant, was investigated theoretically for phase gratings by Kermisch [KER66] and Uchida [UCH73] and for both phase and absorption gratings by Kubota [KUB76B], [KUB78]. Owen and Solymar [OWE80] showed that absorption can also lead to a non-uniform average refractive index and
that although this may be ignored in transmission holograms, it can have a significant effect in reflection holograms.

As discussed in Sec. 4.1, attenuation of the recording beams is not the only source of grating non-uniformities. Kubota [KUB79B] found evidence of bending of interference fringes in planar silver halide transmission holograms which he ascribed to a prestress in the emulsion, formed during manufacture, which was relieved during processing. He derived coupled wave equations in which fringe spacing and refractive index modulation were a function of depth and solved these numerically with the appropriate boundary conditions for transmission holograms. Owen [OWE82] applied a similar analysis to non-uniformities in silver halide reflection gratings which, he suggested, were formed during processing. However, he followed the approach used by Kermisch [KER66] and Tomlinson [TOM75] whereby a non-uniform grating is modelled as being composed of a number of adjacent uniform gratings.

As yet there has not been a thorough investigation of the non-uniformities in dichromated gelatin holograms. This is despite the fact that considerable anomalies have been seen in the angular response of these gratings, particularly with large thicknesses [DUN85]. Kubota investigated transmission holograms recorded in dye sensitised dichromated gelatin [KUB78]. The considerable absorption during recording (due to the sensitising dye) led to a significant variation of the refractive index modulation and he obtained good agreement with his earlier theory [KUB76B]. However, there has been no analysis which includes the effect of non-uniform fringe spacing or average refractive index which might be expected to have strong effects in reflection holograms. There has also been no investigation of how the performance of dichromated gelatin holograms changes with wavelength due to dispersion.
4.2.1 Derivation of the coupled wave equations.

For this analysis we obtain coupled wave equations for the two wave case in which the refractive index modulation, the average refractive index and the fringe spacing are functions of depth. The absorption constant and the absorption modulation are not made functions of depth since in dichromated gelatin they are both small and have little effect. After defining the complex permittivity in terms of these spatially varying quantities it is possible to follow the approach taken in Chapter 1 to obtain the coupled wave equations. This is shown in Appendix II. An important result of this derivation is that it is found that it is not possible to distinguish the effects of a non-uniform fringe spacing from the effects of a non-uniform refractive index. This is because it is the optical path between the fringes which is most significant in determining the dephasing parameter. For an analysis of experimental results we therefore choose to specify $K_x$ as a function of depth and assume the average refractive index is constant. The coupled equations therefore become

$$
\frac{\rho_x}{\beta_a} \frac{\partial R}{\partial x} + \alpha_0 R + j\kappa(x)S \exp{j\gamma(x)} = 0 \quad (4.1)
$$

$$
\frac{\sigma_x}{\beta_a} \frac{\partial S}{\partial x} + \alpha_0 S + j\kappa(x)R \exp{-j\gamma(x)} = 0 \quad (4.2)
$$

where

$$
\gamma(x) = \int_0^x K_x(s) \, ds - \sigma_x x + \rho_x x \quad (4.3)
$$

4.2.2 Numerical solution of the coupled equations.

Because these coupled wave equations cannot be solved analytically they were solved by numerical integration using a Runge-Kutta method on a Vax 780 computer. A problem arises because the boundary conditions for reflection holograms are given on different sides of the hologram:
therefore the equations were solved by integrating from $x = d$ to $x = 0$ using the boundary conditions

$$R(d) = 1.0 \quad S(d) = 0.0 \quad (4.5)$$

yielding values of $R(0)$ and $S(0)$

$$R(0) = R_a \quad S(0) = S_a \quad (4.6)$$

From these we can obtain the values of $R(d)$ and $S(0)$ which we require, corresponding to the correct boundary conditions (4.4), as

$$R(d) = \frac{1}{R_a} \quad S(0) = \frac{S_a}{R_a} \quad (4.7)$$

4.2.3 Mathematical representation of the non-uniformities.

Although the solution of the coupled wave equations has been discussed the form of $K_x(x)$ and $\kappa(x)$ have not yet been specified. A suitable function for both was found to be a quadratic polynomial with its origin at the centre of the hologram. Following the convention of Kogelnik [KOG76]

$$K_x(x_n) = K_{x0} + \frac{2a_1x_n}{d} + \frac{12a_2x_n^2}{d} \quad (4.8)$$

$$\kappa(x_n) = \kappa_0 \left[ 1 + b_1x_n + \frac{b_2x_n^2}{12} - \frac{b_2}{12} \right] \quad (4.9)$$

where $x_n = -0.5$ and $x_n = 0.5$ correspond to the two boundaries of the hologram, $K_{x0}$ is the uniform component of $K_x(x)$, $\kappa_0$ is the average refractive index modulation and $a_1, a_2, b_1, b_2$ are constants.
The form of the polynomial for $\kappa(x)$ has been chosen so that the average modulation is constant and therefore the values of $b_1$ and $b_2$ do not affect the efficiency of the hologram.

### 4.3 Properties of the Model

The individual effect of each of the coefficients of the two polynomial variations are shown in Fig. 4.5 - 4.9. It is clear that each of these parameters has a particular affect on the sidelobe structure. Linear variations in $K_x(a_1)$ and modulation ($b_1$) produce similar results in that they both cause a blurring of the sidelobes. However, linear variations in modulation also affect the phase of the sidelobes and can broaden the main peak considerably. Quadratic variation in $K_x(a_2)$ is the only parameter which introduces asymmetry and affects the relative strength of the sidelobes either side of the Bragg angle. A quadratic variation in the modulation ($b_2$) varies the relative strength of the main peak and the sidelobes and it is possible use this to suppress the sidelobes to the extent that bandwidth is significantly reduced.

If the angle of incidence is set to zero and the replay wavelength is varied very similar results are obtained with this model to those of Kogelnik in his analysis of non-uniform almost-periodic structures [KOG76].

From the model it is possible to calculate the angular response of a specified non-uniform grating structure, but it is not possible to calculate the grating structure of a real hologram from its measured angular response. However, because the grating varies slowly and is continuous, the hypothesis is put forward that if only one set of parameters can be found that results in a theoretical response similar to the observed response then those parameters are an acceptable estimation.
Fig. 4.5. Linear variation of $K_x$ with depth: (a) the theoretical angular response and (b) the $K_x$ variation for different values of the parameter $a_1$.

Fig. 4.6. Quadratic variation of $K_x$ with depth: (a) the theoretical angular response and (b) the $K_x$ variation for different values of the parameter $a_2$. 
Fig. 4.7. Negative quadratic variation of $K_x$ with depth: (a) the theoretical angular response and (b) the $K_x$ variation for different negative values of the parameter $a_2$.

Fig. 4.8. Linear variation of modulation with depth: (a) the theoretical angular response and (b) the $\kappa$ variation for different values of the parameter $b_1$. 
Fig. 4.9. Quadratic variation of modulation with depth: (a) the theoretical angular response and (b) the $\kappa$ variation for different values of the parameter $b_2$.

Fig. 4.10. An intermediate stage in the fitting of the angular response of a 16 $\mu$m planar reflection grating (Hologram E) at 514.5 nm. ($a_1 = 0.0$, $a_2 = 1.54$, $b_1 = 0.0$, $b_2 = 0.0$)
of the hologram non-uniformities. This hypothesis is supported by observations made when using the theoretical model to fit experimental data. It is found that, although there is a small range of values for each parameter in which an acceptable match results, if a radical change is made to one parameter it is not possible to compensate for this by changes made to the others.

The fact that the parameters each have a characteristic effect makes it easier to determine suitable values to match experimental data and reduces the likelihood of ambiguity. However, when several parameters are combined they can often interact in a more complicated manner. This problem is worse with large levels of chirp and increases the effort required to find a reasonable comparison with experimental data.

4.4 EXPERIMENTAL DETAILS.

The angular response of holograms, chosen for comparison with the model, was measured at several wavelengths using the apparatus described in Sec 3.3.4. It is essential that the apparatus can resolve the rapidly varying sidelobes of the response if a valid comparison is to be made. For this reason the resolution of the spectrometer must be adequate and the angular size of the light source must be small. Very thick gelatin layers (>100μm) have very fast variations in the sidelobes and results from the spectrometer were compared with measurements made using an unexpanded laser beam to check that the apparatus had sufficient resolution.

Because the level of absorption is very low in most dichromated gelatin reflection gratings, the angular response of a non-uniform grating will be identical if it is rotated through 180°. This means that
if the grating non-uniformities deduced from the model are asymmetric it is not possible to determine which corresponds to the substrate side of the gelatin or to the external side. Because of this the angular response of holograms has only been measured from one orientation with the light incident on the glass substrate in all cases. The transmission is only shown for positive angles because, as the gratings are all unslanted, the angular response was symmetric.

There was no additional information to be gained by measuring the diffracted power because there is very little power in higher orders in these thick reflection holograms and because absorption losses are small. This means that the power in the first diffracted order is almost identical to the depletion of the transmitted power.

4.5 COMPARISON WITH EXPERIMENTAL RESULTS.

Two unslanted planar reflection gratings, recorded and processed using the procedure described in Chapter 3, were chosen for comparison with the model. The first of these (hologram E) is 16 µm thick while the other (hologram F) is much thinner and is 5 µm thick. The thicker hologram has a more detailed sidelobe structure and considerable time and effort was required to find a set of parameters which give agreement with the observed angular response.

The procedure used to obtain a fit of the experimental data is demonstrated in Fig 4.10 - 4.13 for hologram E. The first step is to obtain the best possible fit whilst assuming the grating is uniform (Fig. 4.2). The parameter $a_2$ is then set to give the correct degree of asymmetry in the sidelobe structure (Fig. 4.10). Parameters $a_1$ (Fig. 4.11) and then $b_1$ (Fig. 4.12) are introduced to give the correct level of 'blurring' of the sidelobes and to reproduce the features on the
Fig. 4.11. An intermediate stage in the fitting of the angular response of a 16 \( \mu \text{m} \) planar reflection grating (Hologram E) at 514.5 nm. 
\( (a_1 = 4.5, a_2 = 1.54, b_1 = 0.0, b_2 = 0.0) \)

Fig. 4.12. An intermediate stage in the fitting of the angular response of a 16 \( \mu \text{m} \) planar reflection grating (Hologram E) at 514.5 nm. 
\( (a_1 = 4.5, a_2 = 1.54, b_1 = -0.27, b_2 = 0.0) \)
right hand side of the Bragg dip. The choice of the relative sign of $a_1$ and $b_1$ is important and has a strong affect on the character of the angular response. Finally the relative strength of the sidelobes and Bragg dip is set with parameter $b_2$ (Fig. 4.13). This procedure is repeated several times until a reasonable match of both the features of the sidelobes and the shape of the main Bragg dip is obtained.

4.5.1 Replay with the recording wavelength.

A fit was obtained for holograms E and F when replayed at the recording wavelength (514.5 nm). The results are shown in Fig. 4.13 and 4.14 together with a graphical representation of the non-uniformities used to generate the theoretical results. Good agreement between the theory and experiment has been obtained and the theory can match many of the features of the angular response. The values of the parameters $a_1$, $a_2$, $b_1$ and $b_2$ are given and, to give some indication of the level of the ambiguity in these results, a tolerance has been put on the values of the parameters in each case. The variation in $K_x$ with depth, although small in absolute terms relative to the variation in modulation, has a comparable affect on the angular response.

There is some similarity between the non-uniformities in the two holograms, although it should be remembered that the scale of the thickness is different. One feature of both is that there appears to be some connection between $K_x$ and modulation changes. High modulation appears to be associated with low values of $K_x$ (large fringe spacing) and vice versa. It is not suprising that there is some relationship because the mechanism of modulation formation involves changes in thickness, density and average refractive index. It is interesting to see that maximum modulation does not occur at either surface but is offset from the centre of the hologram suggesting that there are two separate effects.
Fig. 4.13 (a) A theoretical fit of the angular response of a 16 µm planar reflection grating (Hologram E) at 514.5 nm, (b) the variation in $K_x$ and (c) modulation with depth. ($a_1 = 4.5 \pm 0.5$, $a_2 = 1.54 \pm 0.3$, $b_1 = -0.27 \pm 0.1$, $b_2 = -1.0 \pm 0.2$)
Fig. 4.14 (a) A theoretical fit of the angular response of a 5 μm planar reflection grating (Hologram F) at 514.5 nm, (b) the variation in $K_x$ and (c) modulation with depth. ($a_1 = 1.2 \pm 0.6$, $a_2 = 0.8 \pm 0.3$, $b_1 = -1.0 \pm 0.3$, $b_2 = -2.4 \pm 0.4$)
which result in loss of the modulation at each boundary. As discussed in Section 4.3.1, it is not possible to determine which side of these graphs correspond to a particular side of the gelatin but it is likely that the side with highest modulation (L.H.S.) is the external gelatin surface rather than the gelatin/glass interface, as this is most exposed to the processing chemicals during development.

It was found that the best fit for hologram E was obtained if the thickness was assumed to be 2 \( \mu m \) less than was measured after processing. This could be due to the fact that the simple polynomial used cannot model rapid changes which may occur at the edges of the grating. No disparity in the thickness was found for hologram F but this could be because there is less detail in the sidelobe structure. This lack of detail means that there is more ambiguity in the values of the parameters for hologram F which can be seen in the greater tolerances quoted in Fig. 4.13 and 4.14.

4.5.2 Replay at other wavelengths.

A reasonable agreement has been found between the theory and the behaviour of the hologram at one wavelength. It is important however, that the parameters which have been deduced predict the behaviour at other replay wavelengths correctly. Several quantities might be expected to change with wavelength: The average refractive index and the average refractive index modulation due to dispersion, and the absorption constant.

The angular response of holograms E and F are shown for a range of replay wavelengths in Fig. 4.15 a-c and Fig. 4.16 a-c together with theoretical responses which have been generated from the same values of \( a_1, a_2, b_1 \) and \( b_2 \) deduced in Sec. 4.5.1 but with new values for the average refractive index, modulation and absorption. It can be seen that
Fig. 4.15 (a) Theoretical fit of the angular response of a 16 μm planar reflection hologram (Hologram E) at 488.0 nm and 406.7 nm.

Fig. 4.15 (b) Theoretical fit of the angular response of a 16 μm planar reflection hologram (Hologram E) at 514.5 nm and 450.0 nm.
Fig. 4.15 (c) Theoretical fit of the angular response of a 16 \( \mu \text{m} \) planar reflection hologram (Hologram E) at 560.0 nm and 546.1 nm.

Fig. 4.16 (a) Theoretical fit of the angular response of a 5 \( \mu \text{m} \) planar reflection hologram (Hologram F) at 488.0 nm and 406.7 nm.
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Fig. 4.16 (b) Theoretical fit of the angular response of a 5 \( \mu \)m planar reflection hologram (Hologram F) at 546.1 nm and 450.0 nm.

Fig. 4.16 (c) Theoretical fit of the angular response of a 5 \( \mu \)m planar reflection hologram (Hologram F) at 560.0 nm and 514.5 nm.
the agreement between the theoretical and the experimental response is good over this range of wavelengths.

As was discussed in Sec. 4.2.1 we have, for convenience, chosen to model changes in fringe spacing and refractive index with depth as a variation in $K_x$ alone, due to the ambiguity which would otherwise be introduced. A variation of the fringe spacing with depth will not change with wavelength but a refractive index variation may change due to dispersion. To some extent this will be compensated for by changing the constant average refractive index in the model but we might expect to have to change the values of $a_1$ and $a_2$ slightly to optimise the fit at each wavelength. However, since only small differences can be seen in the quality of the agreement at the different wavelengths it must be concluded that it is not practicable to separate the effects of non-uniform fringe spacing and refractive index in this way and that modelling both these as a variation of $K_x$ is adequate for the purposes of these experiments.

From this analysis useful information about the dispersion of the parameters of the holograms can be determined. The change in the average refractive index with wavelength can be found quite accurately because the position of the Bragg angle is quite sensitive to this parameter and is shown in Fig. 4.17 for hologram E. The changes in the refractive index modulation and the absorption constant with wavelength can be found from the depth of the Bragg dip and the off-Bragg attenuation respectively and are shown in Fig. 4.18 and 4.19.

4.5.3 Effect of recording in an attenuating medium.

The effect of attenuation of the recording beams in dichromated gelatin is complicated by the fact that the absorption increases with exposure. However, when using a low sensitiser concentration of 2% and
Fig. 4.17. The variation of the average refractive index \( n_0 \) with wavelength (Hologram E).

Fig. 4.18. The variation of the refractive index modulation \( n_1 \) with wavelength (Hologram E).
Fig. 4.19. The variation of the absorption constant ($\alpha_0$) with wavelength (Hologram E).

Fig. 4.20. The variation in the uniform component of the exposure with depth for (a) 18 $\mu$m and (b) 155 $\mu$m dichromated gelatin.
at normal exposures levels this increase is small (< 15%) and is unlikely to have a significant affect on the non-uniformities introduced by the absorption. To estimate the non-uniformity of the exposure it is therefore assumed that the absorption is constant during the exposure.

The amplitudes of the two recording waves $A_1$ and $A_2$ for the recording configuration shown in Fig 3.8 are

$$A_1(x) = A_0 \exp \left[ -\frac{a x}{\cos \theta} \right]$$

$$A_2(x) = A_1(d) \cdot r \exp \left[ -\frac{a(d-x)}{\cos \theta} \right]$$

where $A_0$ is the initial amplitude, $a$ the absorption constant, $d$ the gelatin thickness, $\theta$ the recording angle and $r$ the reflectivity of the mirror behind the gelatin.

The intensity relative to the input intensity is therefore of the form

$$\frac{I(x)}{I_0} = R_0 + R_1 \cos(Kx)$$

where

$$R_0 = \exp \left[ -\frac{a x}{\cos \theta} \right] + r^2 \exp \left[ -\frac{2a(2d-x)}{\cos \theta} \right]$$

$$R_1 = 2 \exp \left[ -\frac{4ad}{\cos \theta} \right]$$

It can be seen that the fringe amplitude ($R_1$) is constant but that there is a variable uniform component of the exposure ($R_0$) through the hologram. This could lead to a variable average refractive index and, because the recording response is non-linear, to a small change in modulation with depth. The absorption coefficient of 18 $\mu$m and 155 $\mu$m dichromated gelatin and the reflectivity of the mirror were measured at 514.5 nm as
Using these values $R_0$ is plotted for the two thicknesses in Fig. 4.20. It can be seen that for 18 $\mu$m layers there is only a small variation of the uniform exposure level (<2%) through the emulsion. It can be concluded that processing must be responsible for the level non-uniformity seen in these holograms and not the attenuation of the recording beams. The variation is much higher (25%) in the 155 $\mu$m layer and therefore attenuation may be more significant in thick holograms.

In some situations, which are not encountered in this chapter, the absorption in dichromated gelatin may be more significant at the recording stage: Absorption is much higher when recording at shorter wavelengths [MEY72] or when using sensitising dyes such as methylene blue [KUB76].

4.6 THE EFFECT OF PROCESSING ON GRATING NON-UNIFORMITIES.

4.6.1 The effect of dehydration rate.

There are have been many variations in the processing procedures suggested for dichromated gelatin in published work. Changing the dehydration stage of the process produces the most significant differences and several authors have suggested alterations with the aim of increasing the efficiency or altering the bandwidth of the holograms [MCG80, CUL82]. It is not always clear if the changes in bandwidth are a result of a change in the grating non-uniformities or are a secondary result of a change in modulation or Bragg angle because an accurate angular response is not given. However, it is often suggested that
reducing the dehydration rate reduces bandwidth because it leads to a more uniform grating.

The rate of dehydration can be controlled by drying the hologram in several propanol/water mixtures of steadily increasing propanol concentration. The number of baths chosen can vary from 1 to 6. Additionally, the temperature of each of these baths [MCG80] and the time in each bath can be different. The most common dehydration procedure is to use 50% propanol followed by 100% propanol at 20°C - 40°C and this process has been used for most of the holograms in this thesis.

In general reducing the processing rate, by increasing the number of intermediate propanol baths or by reducing the temperature, results in a decrease in modulation, a decrease in scatter and a reduction in bandwidth. To investigate if there are any changes in the non-uniformities a number of planar gratings were recorded and then processed with a reduced dehydration rate. The two propanol baths of the standard process, shown in Table 2.1, were replaced by the following four propanol baths:

1) 25% Propanol. (All for 5 Mins at 25°C.)
2) 50% Propanol.
3) 75% Propanol.
4) 100% Propanol.

Temperatures A and B in Table 2.1 were both 25°C. It was found that, for the same exposure, the modulation achieved by this processing procedure was almost half that achieved using the standard process and that the replay angle was reduced by about 2.5° due to a reduction in swelling.

The angular response of an 18 μm hologram (Hologram G), processed in this manner, is shown in Fig. 4.21 together with a theoretical comparison. The sidelobe structure is significantly different to
Fig. 4.21 (a) A theoretical fit of the angular response of a 18 μm planar reflection grating (Hologram G - alternative processing) at 514.5 nm, (b) the variation in $K_x$ and (c) modulation with depth. ($a_1 = 2.0, a_2 = 0.8, b_1 = -1.8, b_2 = 0.4$)
holograms made with the standard process. The size of the sidelobes relative to the main Bragg dip is reduced and there is a greater degree of smoothing. Two plates of nine exposures were processed in this way and all had these characteristic features.

The results of the theoretical analysis are surprising because a simple result was expected - that slower processing would lead to a more uniform grating. However, it can be seen that, although there is some reduction in the variation in $K_x$ with depth compared to Hologram E, the variation in modulation is much higher. The smoothing of the sidelobes and the shape of the main Bragg dip could only be reproduced by using a large value of $b_1$, corresponding to a large linear modulation variation. It is not understood why this particular grating structure has been formed but it suggests that the processes occurring during the dehydration stage are complex. The simple proposition that slower processing leads to a more uniform grating is inaccurate and has probably developed because of a lack of understanding of the precise effects of grating non-uniformities. Because the strength of the sidelobes has been reduced the effective bandwidth of the hologram has been reduced by a useful amount.

4.6.2 The effect of reprocessing.

Reprocessing is often performed on dichromated gelatin holograms to adjust the modulation or Bragg angle to a desired level [CHA76]. Successive reprocessing can lead to a reduction in the modulation achieved (unless temperature is increased) and an increase in losses due to scatter. During some attempts at reprocessing it was found that it could also affect the non-uniformities in a hologram.

An 18 $\mu$m hologram (Hologram H) was processed using the standard processing procedure. It was not coverplated and suffered a loss of efficiency due to the effects of humidity. It was then reprocessed by
soaking in water and repeating the dehydration process using the four propanol baths described in Sec 4.4.1. The efficiency of the subsequent hologram was found to be lower than expected and losses are higher than normal at around 5%. The angular response of the hologram which resulted is shown in Fig. 4.22. It can be seen that the characteristic asymmetry found in most dichromated gelatin holograms has been reversed. The angular response of each of the nine exposures on this plate displayed this feature which has not been observed in any other holograms fabricated during research for this thesis.

A theoretical comparison suggests the reversal is due a change in the character of the \( K_x \) variation which is now at a maximum in the centre of the gelatin rather than at the edges. It is not understood why this has occurred but it suggests that the mechanism that causes degradation of the hologram during reprocessing also has a significant affect on the uniformity. This degradation, which results in a loss of efficiency and increased scatter, could be due to a partial destruction of the hardness differential in the gelatin.

These observations suggest that it may be possible to find a processing and reprocessing procedure that would result in a removal of the asymmetry of the sidelobes by inducing a more uniform or less quadratic variation of \( K_x \), rather than the complete reversal that has been observed.

4.7 NON-UNIFORMITIES IN THICK REFLECTION HOLOGRAMS.

4.7.1 Thick hologram processing.

A useful feature of dichromated gelatin is that holograms of a wide range of thickness can be fabricated. It is possible to use gelatin
Fig. 4.22 (a) A theoretical fit of the angular response of a reprocessed 18 \( \mu \text{m} \) planar reflection grating (Hologram H) at 514.5 nm, (b) the variation in \( K_x \) and (c) modulation with depth.

(\( a_1 = 2.0, a_2 = -1.8, b_1 = -2.0, b_2 = 1.0 \))
layers from 1 \( \mu m \) to >100 \( \mu m \) thick if a suitable processing procedure is chosen to suit the circumstances.

To achieve the same efficiency, the modulation required for thick holograms will be less than for thin holograms. Increasing the modulation above the minimum required for high efficiency results in an unwanted increase in bandwidth due to intermodulation. One aim of the process chosen for thick holograms is therefore to achieve much lower levels of modulation than is normally required.

It might be expected that thicker gelatin layers will be more difficult to process uniformly because the processing chemicals have to diffuse through more material. Another aim is therefore to reduce the level of non-uniformities that increase the strength of the sidelobes, or to increase the non-uniformities which tend to suppress the sidelobes.

Using a low level of exposure to achieve the reduction in modulation is not recommended. This is because the overall hardening effect of the exposure is lost and this can result in patchy and noisy holograms with high levels of chirp. A better method is to use a processing technique that controls the non-uniformities and reduces the modulation.

Whilst processing 50 \( \mu m \) gelatin layers it was seen that the colour change which accompanies Step 6 in Table 2.1 took over 10 mins. Because of this it was decided to increase all processing times to 20 minutes (instead of the 5 minutes used for thin holograms) to ensure that all steps reached completion throughout the gelatin. To reduce the dehydration rate the four propanol baths described in Section 4.6 were used and all processing temperatures were reduced to 20°C. The complete processing procedure is shown in Table 4.1 and has been used successfully to fabricate very thick reflection and transmission holograms.
TABLE 4.1 THICK HOLOGRAM PROCESSING PROCEDURE.

<table>
<thead>
<tr>
<th>STEP</th>
<th>TEMP.</th>
<th>TIME</th>
<th>PROCESSING SOLUTIONS (all 200 ml)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>20°C</td>
<td>20 min</td>
<td>2% Ammonium dichromate + wetting agent.</td>
</tr>
<tr>
<td>2.</td>
<td>25°C</td>
<td>4 hrs.</td>
<td>Dry in rapidly circulating dry air.</td>
</tr>
<tr>
<td>3.</td>
<td></td>
<td></td>
<td>Expose at 514.5 nm (≈ 600 mJ/cm²).</td>
</tr>
<tr>
<td>4.</td>
<td>20°C</td>
<td>2 min.</td>
<td>Wash off index matching fluid with methanol.</td>
</tr>
<tr>
<td>5.</td>
<td>20°C</td>
<td>20 min</td>
<td>0.5% Ammonium dichromate solution.</td>
</tr>
<tr>
<td>6.</td>
<td>20°C</td>
<td>20 min</td>
<td>25% Agfa Structurix fixer + 1% Acid Hardener.</td>
</tr>
<tr>
<td>7.</td>
<td>20°C</td>
<td>20 min</td>
<td>Distilled water.</td>
</tr>
<tr>
<td>8.</td>
<td>20°C</td>
<td>20 min</td>
<td>25% Propanol.</td>
</tr>
<tr>
<td>9.</td>
<td>20°C</td>
<td>20 min</td>
<td>50% Propanol.</td>
</tr>
<tr>
<td>10.</td>
<td>20°C</td>
<td>20 min</td>
<td>75% Propanol.</td>
</tr>
<tr>
<td>11.</td>
<td>20°C</td>
<td>20 min</td>
<td>100% Propanol.</td>
</tr>
<tr>
<td>12.</td>
<td>40°C</td>
<td>12 hrs.</td>
<td>Dry rapidly in circulating hot air.</td>
</tr>
<tr>
<td>13.</td>
<td>20°C</td>
<td></td>
<td>Coverplate using optical quality epoxy resin.</td>
</tr>
</tbody>
</table>

4.7.2 Thick hologram behaviour.

The angular responses of a 50 μm (Hologram I) and a 155 μm (Hologram J) reflection grating are shown in Fig. 4.23 and 4.24 respectively. It was found that no reasonable theoretical comparison could be generated with the quadratic polynomials defined in Eqn. 4.9 and 4.10, and it was necessary to define the \( K_x \) and modulation variation in terms of a quartic polynomial

\[
K_x(x_n) = K_{x0} + 2a_1x_n + 12a_2x_n^2 + 32a_3x_n^3 + 80a_4x_n^4
\]  \hspace{1cm} (4.15)

\[
\kappa(x_n) = \kappa_0 \left[ 1 + b_1x_n + b_2x_n^2 + b_3x_n^3 + b_4x_n^4 - \frac{b_2}{12} - \frac{b_4}{80} \right]
\]  \hspace{1cm} (4.16)
Fig. 4.23 (a) A theoretical fit of the angular response of a 50 µm planar reflection grating (Hologram I) at 514.5 nm, (b) the variation in $K_x$ and (c) modulation with depth. ($a_1 = 20.0$, $a_2 = -4.0$, $a_3 = -5.0$, $a_4 = 12.0$, $b_2 = -0.2$, $b_3 = -2.4$, $b_3 = 0.0$, $b_4 = -8.0$)
Fig. 4.24 (a) A theoretical fit of the angular response of a 155 μm planar reflection grating (Hologram J) at 514.5 nm, (b) the variation in $K_x$ and (c) modulation with depth. ($a_1 = 6.0$, $a_2 = -10.0$, $a_3 = 1.0$, $a_4 = 1.0$, $b_1 = -1.0$, $b_2 = 1.0$, $b_3 = 0.0$, $b_4 = -1.0$)
This necessity was not unexpected because a small level of chirp has a greater effect in thicker layers and because greater detail might occur with the larger scale of thick holograms.

However, using a quartic polynomial means that there are 8 free parameters in the model and this introduces several problems. The first problem is that cubic and quartic variations of $K_x$ and modulation do not have such a distinctive effect as the lower order variations although, in general, they tend to affect sidelobes which are further away from the Bragg angle. This means that matching has to be done by trial and error rather than in the logical progression described in Section 4.5. With 8 separate parameters finding a reasonable comparison is difficult and time consuming. The second problem is that there is now a greater degree of ambiguity between the parameters and it is difficult to make a quantitative estimate of the accuracy of the results which have been achieved.

For Hologram I it was found to be difficult to find why the sidelobes on the left hand side of the Bragg angle decay suddenly after the first two oscillations. This is very different to hologram J, where the lefthand sidelobes are sustained and decay slowly, which can be matched well using parameters $a_4$ and $a_2$. For both holograms it was found that a $K_x$ variation with a flat centre region and with rapid changes at each edges produced the best matching.

Although there are similarities between the grating structures suggested for the two thick holograms there are also significant differences. This could be because the 50 $\mu$m gelatin layers are produced by a spin coating method whilst the 155 $\mu$m layers are produced by casting.
4.8 COMPARISON WITH OTHER RECORDING MATERIALS.

It has been seen that there are common characteristics in the non-uniformities of many of the dichromated gelatin holograms seen in this chapter. It is therefore of interest to compare these characteristics with the behaviour of other recording materials.

4.8.1 Non-uniformities in silver halide emulsions.

The angular response of a 5 μm silver halide reflection hologram (Hologram K) is shown in Fig. 4.25. This was recorded in Agfa 8E56HD and processed for high efficiency as described by Cook [CO084]. Theoretical matching of this grating reveals high modulation occurs in regions of high $K_x$ or low fringe spacing. This is the opposite of the behaviour seen in dichromated gelatin. By considering the mechanism by which modulation is formed in the two materials it is possible to suggest reasons for this difference.

Refractive index changes in silver halide phase holograms result from the loss of high refractive index material (the silver halides) from a passive gelatin support. As a result the average refractive index falls and it is usual to see evidence of shrinkage after processing. Both these changes will lead to a reduction in the optical path between fringes. At depths in the gelatin where modulation is high the amount of material lost will, on average, be highest and therefore fringe spacing will be low. This argument does not explain why the modulation and fringe spacing are not uniform, but does explain why the variation in both quantities are connected if non-uniformities are introduced by non-uniform processing for example.

It is significant that swelling rather than shrinkage is normally seen after processing dichromated gelatin holograms. In Sec 4.6.1 it was found that reducing the dehydration rate resulted in less modulation and
Fig. 4.25 (a) A theoretical fit of the angular response of a 5 μm silver halide reflection grating (Hologram K) at 514.5 nm, (b) the variation in $K_x$ and (c) modulation with depth.

($a_1 = 1.0$, $a_2 = -0.6$, $b_1 = -0.1$, $b_2 = -0.5$)
Fig. 4.26 (a) A theoretical fit of the angular response of a 7 μm photopolymer reflection grating (Hologram L) at 406.7 nm, (b) the variation in Kx and (c) modulation with depth. 
(a₁ = 0.0, a₂ = 0.0, b₁ = -1.4, b₂ = -1.0)
less swelling (detected by the shift in replay angle). Low modulation is therefore associated small fringe spacing and high values of $K_x$. If the rate of dehydration is not uniform with depth in the gelatin, this relationship between modulation and fringe spacing will be seen in the non-uniformities which result.

Although these theories can explain the difference which has been detected by the theoretical model they do not explain all the features of the non-uniformities and it is likely that there are other complex processing mechanisms in both materials.

4.8.2 Non-uniformities in a photopolymer hologram.

Photopolymer recording materials are currently under development by Polaroid Corporation and others [MAT85], [IKE85]. The modulation in these holograms is very high, greater than 10% in some cases, but there are also considerable losses due to scatter and absorption.

The angular response of a 7 $\mu$m photopolymer hologram, Hologram L, at 406.7 nm is shown in Fig. 4.26. The comparison with the theoretical model suggests that there is a large variation in the modulation with depth but that the variation in $K_x$ is small. This results in weak sidelobes, despite the high modulation, and the angular response is almost square with sharp edges. This is a desirable filter response and these characteristics could be useful in some applications.

4.9 SUMMARY.

A coupled wave theory in which fringe spacing and modulation vary with depth has been developed. This has been shown to give better agreement with the angular response of dichromated gelatin holograms than uniform coupled wave theory. The angular response of two holograms has been measured over a wide range of wavelength and good agreement has been
obtained with the theory without changing any of the parameters except those which would be expected to be wavelength dependent. Thus the wavelength dependence of the refractive index, modulation and absorption constant has been found.

The model has been used to deduce the character of the non-uniformity of the gratings and mechanisms have been discussed which may cause the non-uniformities to occur. It has been shown that it is not possible to distinguish the effect of a non-uniform fringe spacing and a non-uniform refractive index, and it is not possible to determine the orientation of the non-uniformities relative to the substrate.

It has been found that processing has a strong affect on the non-uniformities although there may also be an effect due to the level of absorption at recording in some circumstances. The non-uniformities in holograms produced using different processing or reprocessing procedures have been investigated. In particular it has been found that increasing the number of intermediate propanol baths during the dehydration stage reduces the level of the sidelobes, although the width of the central Bragg minima may increase and the modulation achieved for a particular exposure is reduced. This may be useful in filter applications where sidelobes are a problem.

Finally, it has been found that different recording materials have different characteristic non-uniformities. In the case of dichromated gelatin the angular response is usually asymmetric, due to a quadratic variation in $K_x$, and the fringe spacing and modulation appear to be related.
CHAPTER 5.

REAL-TIME EFFECTS IN DICHROMATED GELATIN.

The dynamic properties of dichromated gelatin plates are investigated during exposure by illuminating them firstly by a single plane wave and secondly by two interfering plane waves produced from an argon ion laser at 514.5 nm. The grating recorded is shown to be a pure absorption grating. The experimental results obtained for the output beam intensities as a function of time are compared with the predictions of a theoretical model, and reasonable agreement is found. It is further shown that, if stored in a humid atmosphere, the recorded grating may self-develop into a phase grating of much higher efficiency.

5.1 INTRODUCTION.

Dichromated gelatin is most commonly used as a latent recording material which can be developed into high efficiency phase holograms. However, it has been known for some time that it can also be used as a real-time recording material and the small diffraction efficiency obtained (0.1-1.0%) has been attributed to phase modulation [SHA68, MEY71, MEY72]. In one instance, the change of refractive index during exposure was measured by Shankoff as \( \Delta n \approx 10^{-2} \) [SHA68]. However, a colour change is visible during exposure which would suggest that some level of absorption modulation is present.
There has recently been a renewal of interest in the real-time recording properties of dichromated gelatin due to the needs of the new techniques of real-time optical processing (e.g. phase conjugation, edge enhancement, etc.) [CAL84]. It has been shown that humidity has a strong affect on the efficiency and the sensitivity of the material which are optimised at 40% R.H. [CAL85].

The aim of this chapter is to investigate the mechanism behind the formation of these real-time gratings which has not yet been established.

5.2 THE CHANGE OF ABSORPTION WITH EXPOSURE TO A SINGLE BEAM.

During the exposure of dichromated gelatin a colour change is visible as a result of the photochemical reaction discussed in Sec. 2.2. The sensitised gelatin is initially bright orange but becomes dark brown during exposure, suggesting that there is an increase in absorption.

The experimental apparatus shown in Fig. 5.1 was used to monitor this change in absorption, during exposure to a single beam. Exposures were made at a wavelength of 514.5 nm, using an argon-ion laser. The experiment was first carried out using an unexpanded laser beam of about 1.8 mm diameter and a power of about 17.0 mW, incident in air at an angle of 45°. The small beam diameter and large angle were used to ensure that internal reflections were separated from the main beam, but this resulted in a non-uniform intensity. In order to achieve a uniform beam intensity the experiment was repeated using an expanded beam and a tank of index-matching liquid (di-n-butyl phthalate) to minimise the effects of boundary reflections. Unfortunately, the tank could not be used in the two beam experiments, described later in Sec. 5.3, due to instabilities in the liquid during the long exposures required.

The intensity of the transmitted light was measured by a photodiode and recorded by a storage oscilloscope for later analysis.
Fig. 5.1. Schematic diagram of single beam experiment.

Fig. 5.2. Output beam intensity against time for unexpanded single beam experiments.
The results of the measurements are presented as graphs of transmitted beam intensities versus time, in Fig. 5.2 for the unexpanded beam, and in Fig. 5.3 for the uniform expanded beam. In each case, results are given for plates sensitised in 5% and 10% solutions of Ammonium Dichromate. $I_0$ is the intensity of a beam transmitted through an unsensitised gelatin plate. The initial transmitted intensities, $I_5$ and $I_{10}$, clearly depend on the level of sensitisation.

The superimposed solid curves are calculated from the theory presented in the next section. The results show a steep decay in intensity at first, followed by a region in which saturation is reached. The decay is much slower for the expanded beam (Fig. 5.3) due to its lower intensity.

It is clear from the figures that the initial absorption constant is fairly high and that the exposure causes further increase in the absorption. Since the input light intensity decays with distance through the gelatin layer, its effect upon the absorption constant must also decay with distance. An appropriate model for this behaviour would have to allow the intensity, $I$, and the absorption constant, $a$, to vary with time and with the spatial coordinate, $x$.

In order to explain the measured results we shall postulate a model with two species of absorbing agents. At $t = 0$, when exposure starts, only species 1 is present, uniformly distributed with density $N_{10}$. During exposure the incident light causes conversion from species 1 into species 2. Denoting the densities of the two species by $N_1(x,t)$ and $N_2(x,t)$ respectively, we have the relationship

$$N_1(x,t) + N_2(x,t) = N_{10} \quad (5.1)$$

Introducing the notations $\sigma_1$ and $\sigma_2$ for the corresponding absorption cross-sections, we may write
Fig. 5.3. Output beam intensity against time for expanded single beam experiments.

Fig. 5.4. Schematic diagram of two beam experiments.
\[ a_i = gN_1 \sigma_1, \quad a_f = gN_1 \sigma_2 \]  

(5.2)

\[ a(x,t) = g[N_1(x,t) \sigma_1 + N_2(x,t) \sigma_2] \]

where \( g \) is a proportionality constant, \( a_i \) is the initial value of the absorption constant at \( t = 0 \) when the exposure starts, and \( a_f \) is the final value obtained when all of species 1 has been converted into species 2.

The temporal rate of change of \( N_2 \) will now be assumed to be proportional to the light intensity \( I(x,t) \) and to the amount still unconverted, \( N_1(x,t) = N_{10} - N_2(x,t) \). Hence we can write the differential equation

\[ \frac{\partial N_2}{\partial t} = -\gamma I(x,t)[N_{10} - N_2(x,t)] \]  

(5.3)

where \( \gamma \) is again a constant of proportionality.

It is convenient to express Eqn. (5.3) in terms of the absorption constant. With the aid of Eqns. (5.1) and (5.2) the differential equation becomes

\[ \frac{\partial a(x,t)}{\partial t} = \gamma I(x,t)[a_f - a(x,t)] \]  

(5.4)

A second differential equation may be deduced from the usual assumption that the spatial rate of change of intensity is proportional both to the absorption constant and to the intensity,

\[ \frac{\partial I(x,t)}{\partial x} = -\frac{2a(x,t)}{\cos \theta} I(x,t) \]  

(5.5)

where \( \theta \) is the angle of propagation measured in the photosensitive material.

Taking the input surface to correspond to the \( x = 0 \) plane, the initial boundary conditions may be written as

\[ a(x,0) = a_i \quad \text{and} \quad I(0,t) = I_0 \]  

(5.6)
where $I_0$ is the input light intensity. (Note that Eqns. (5.5) and (5.6) are similar to those of Tomlinson [TOM75], derived for the study of photochromic materials, in which both the absorption and the refractive index change in response to input light).

The above differential equations cannot be solved analytically, but a simple numerical method can be applied. At $t = 0$, the absorption constant is $\alpha_i$ everywhere in the material (i.e. for $0 < x < d$ where $d$ is the thickness of the material) and the intensity decays exponentially as

$$I(x,0) = I_0 \exp \left( -\frac{2\alpha_i x}{\cos \theta} \right) \quad (5.7)$$

Assuming now that $I(x,0)$, as given by Eqn. (5.7), is constant for the next interval $\Delta t$ we may work out the change of $\alpha$ in time $\Delta t$ from Eqn. (5.4) at every point in the material. Having determined $\alpha(x,\Delta t)$ we may then substitute it into Eqn. (5.2) and find $I(x,\Delta t)$ by numerical integration. The function $I(x,\Delta t)$ may then be used in Eqn. (5.4) to find $\alpha(x,2\Delta t)$, etc. Thus, effectively, we split up the material into a large number of thin slabs, and the total time of exposure into a large number of $\Delta t$ intervals, and assume that of the two variables $\alpha$ and $I$, one of them may always be regarded as being independent of time.

Values of $\alpha_i$, $\alpha_f$ and $\gamma$ obtained by matching experimental data with curves generated by this method are tabulated in Table 5.1. As might be expected $\alpha_i$ and $\alpha_f$ both vary with the concentration of the sensitising solution used, i.e. with the concentration of ammonium dichromate in the gelatin. The value of $\gamma$ and the ratio $\alpha_f/\alpha_i$ should, according to our model, be constants. Since the theoretical curves obtained are close to the experimental ones, and the variation in the above parameters is not excessive, we may conclude that our two-species model adequately describes the change induced in the absorption constant.
TABLE 5.1. PARAMETERS USED TO MODEL SINGLE BEAM EXPERIMENTS.

(a). Unexpanded single beam experiments.

<table>
<thead>
<tr>
<th>Sensitiser concentration (%)</th>
<th>Gelatin thickness (μm)</th>
<th>$\alpha_i (m^{-1})$</th>
<th>$\alpha_f (m^{-1})$</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>13</td>
<td>$7.37 \times 10^3$</td>
<td>$4.5 \times 10^4$</td>
<td>$1.2 \times 10^{-4}$</td>
</tr>
<tr>
<td>10</td>
<td>13</td>
<td>$1.39 \times 10^4$</td>
<td>$8.2 \times 10^4$</td>
<td>$1.1 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

(b). Expanded single beam experiments.

<table>
<thead>
<tr>
<th>Sensitiser concentration (%)</th>
<th>Gelatin thickness (μm)</th>
<th>$\alpha_i (m^{-1})$</th>
<th>$\alpha_f (m^{-1})$</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>10</td>
<td>$7.69 \times 10^3$</td>
<td>$4.6 \times 10^4$</td>
<td>$1.5 \times 10^{-4}$</td>
</tr>
<tr>
<td>10</td>
<td>10</td>
<td>$1.41 \times 10^4$</td>
<td>$9.0 \times 10^4$</td>
<td>$1.5 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

TABLE 5.2. PARAMETERS USED TO MODEL TWO BEAM EXPERIMENTS.

<table>
<thead>
<tr>
<th>Sensitiser concentration (%)</th>
<th>Gelatin thickness (μm)</th>
<th>$\alpha_i (m^{-1})$</th>
<th>$\alpha_f (m^{-1})$</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>14</td>
<td>$6.9 \times 10^3$</td>
<td>$3.9 \times 10^4$</td>
<td>$2.2 \times 10^{-4}$</td>
</tr>
<tr>
<td>10</td>
<td>14</td>
<td>$1.49 \times 10^4$</td>
<td>$7.3 \times 10^4$</td>
<td>$2.5 \times 10^{-4}$</td>
</tr>
</tbody>
</table>
The photochemical process responsible for the colour change is discussed in Sec. 2.2. One species we could associate with \( \text{Cr}_2\text{O}_7^{2-} \) ions and the other with some ion involving the trivalent state, \( \text{Cr}^{3+} \), which is formed in the photochemical reaction, but the precise species involved are not known.

5.3 THE RECORDING OF A REAL TIME GRATING BY TWO BEAMS.

The measurements described in the previous Section show clearly the change of absorption with exposure. It follows therefore that when two beams are incident symmetrically upon a sensitised plate a real-time unslanted transmission grating will be recorded. Using the apparatus shown in Fig. 5.4 gratings of this type were recorded in 14 \( \mu \text{m} \) gelatin sensitised in 5\% and 10\% ammonium dichromate. To minimise problems due to instability unexpanded laser beams were used of 1 mW total power.

The formation of the grating is monitored by periodically blocking one of the beams (beam 2 in Fig. 5.4) with a rotating shutter and by measuring the power in the diffracted beam: i.e. while beam 2 is cut off the diffraction of beam 1 by the grating is measured with photodiode 2. The geometry of the beams is chosen so that in the volume of the photosensitive material beam 1 is entirely within beam 2. It follows then that when beam 2 is off, the whole of beam 1 participates in the diffraction by the grating. The two part shutter was designed so that photodiode 2 saw only the weak diffracted beam and not the unattenuated strong transmitted beam during the recording periods which would otherwise degrade its ability to measure small changes in the weak beam accurately. The transmitted power of the uninterrupted beam (beam 1) was also monitored using photodiode 1.

The power in the diffracted beam as a function of time, measured by photodiode 2, is shown in Figs. 5.5a and 5.6a respectively. The time
Fig. 5.5. Diffracted and transmitted intensity against time for two beam experiments using 5% sensitisation.

Diffracted intensity:  (a) Experiment,  (b) Theory.
Transmitted intensity:  (c) Experiment,  (d) Theory.
Fig. 5.6. Diffracted and transmitted intensity against time for two beam experiments using 10% sensitisation.

Diffracted intensity: (a) Experiment, (b) Theory.
Transmitted intensity: (c) Experiment, (d) Theory.
scale is up to 100 seconds. The main features of the results are similar to the single beam experiments: diffraction efficiency first increases with the increase in absorption modulation and then gradually reaches saturation. It needs to be noted that if absorption modulation and total absorption increase together the diffraction efficiency is bound to decline beyond a certain stage because most of the diffracted beam is then absorbed. In the present case there is a further reason causing the saturation of diffraction efficiency, namely that the availability of absorption modulation decreases as the actual value of $\alpha$ approaches $\alpha_f$. The maximum achieved diffraction efficiencies were 0.4% and 0.7%, roughly in line with those reported earlier [SHA68, MEY71, CAL84].

The transmitted power, measured by photodiode 1, for the 5% and 10% sensitisations are shown in Figs. 5.5c and 5.6c respectively. It is interesting to see that the received power is consistently higher when beam 2 is off than when beam 2 is on. The received power appears to be less when both beams are incident than when only one of the beams is incident. At first sight this is rather surprising. When both beams are on one would expect some increase in power. However we are, in effect, measuring the superposition of two beams: a strong transmitted beam added to a weak diffracted beam. The measured intensity will obviously depend on the relative phases of the two beams.

We shall show later that the observed result follows directly from the coupled wave equations appropriate for diffraction by a pure absorption grating, but it is also possible to give a simple physical explanation. During recording there is a stationary sinusoidal intensity variation in the photosensitive material. The increase in absorption will be highest at the maxima of this interference pattern. High absorption in the areas of high intensity will then lead to a higher
overall absorption. Consequently, the absorption is higher when both beams are on than when only one beam is on.

A further experimental confirmation of the correctness of the above description is shown in Fig. 5.7 where again the power measured by detector 1 is plotted. At $t = 80 \text{ s}$ one of the mirrors was vibrated, introducing a rapid phase change in one of the beams. As may be seen, the rest position of the mirror gave the lowest received power. Any other position of the mirror led to an increase in received power because any movement of the interference maxima away from the absorption maxima reduced the effective absorption. Considered as a superposition of two beams, this illustrates that the diffracted beam is normally $\pi$ out of phase with the transmitted beam for a pure absorption grating.

This two beam absorption phenomenon may be regarded as the opposite of the Borrmann effect [BAT64, RUS80], well known in X-ray diffraction, which leads to a decrease of absorption when high intensity is associated with low absorption. There is also a relationship with Tomlinson's experiments in photochromic materials [TOM75] where modulation is obtained by bleaching the material, i.e. the absorption constant decreases with exposure. The absorption minima then occur at the intensity maxima and consequently the transmitted power is higher when both beams are present.

5.4 A TWO-BEAM MODEL FOR GRATING FORMATION.

We shall now extend the mathematical model of Sec. 5.2, which describes the output intensity and the temporal and spatial variation of the absorption coefficient in response to a single beam, to the case of two symmetrically incident beams.

Eqn. (5.4) for the temporal rate of change of the absorption constant is still valid provided we take $I(x,t)$ as the total intensity at
Fig. 5.7. Experimentally observed changes in transmitted intensity which occur after a grating has been recorded if a phase change is introduced which shifts the fringe pattern relative to the fixed grating.

Fig. 5.8. Transmission against angle at 514.5 nm for an undeveloped D.C.G. transmission hologram after 24 hours in increased humidity.
any given point. We shall assume the amplitudes of the two beams in the form

\[ R \exp j(\rho \cdot \tilde{r}) \text{ and } S \exp j(\sigma \cdot \tilde{r}) \]  

(5.8)

where \( \rho \) and \( \sigma \) are the wave vectors of the incident beams, \( \tilde{r} \) is the radius vector and \( \tilde{K} \) is the grating vector equal to \( \tilde{K} = \tilde{\rho} - \tilde{\sigma} \), and its modulus may be written as \( |\tilde{K}| = 2\beta \sin \theta \) where \( \beta = \frac{2\pi}{\lambda} \), \( \lambda \) is the wavelength in the medium and \( \theta \) is the angle of incidence of each beam. The total intensity at an arbitrary point in the hologram is then given by

\[ I = R^2 + S^2 + 2RS\cos(\tilde{K} \cdot \tilde{r}) \]  

(5.9)

where \( R \) and \( S \) may be assumed to be real since we shall restrict the analysis to a pure absorption grating.

To describe the interaction which will occur between the two beams as the grating forms we can use the coupled wave equations for the two wave case derived in Chapter 1. for a uniform grating. The coupled differential equations (1.16) (though not the analytical method of solution) are still valid when the parameters vary as a function of space [SOL81], so they are applicable to the present case. Because the replay beam is always identical to the recording beam the dephasing parameter \( \psi \) is always zero. For a pure absorption grating the coupled equations may therefore be written in the form

\[ \cos \theta \frac{\partial R}{\partial x} + \alpha_0 R = - \frac{\alpha_1}{2} S \]  

(5.10)

and

\[ \cos \theta \frac{\partial S}{\partial x} + \alpha_0 S = - \frac{\alpha_1}{2} R \]  

(5.11)
where $\alpha_0$ is the average absorption coefficient and $\alpha_1$ is the amplitude of the sinusoidal absorption modulation, i.e. the total absorption coefficient is of the form

$$\alpha = \alpha_0 + \alpha_1 \cos (\vec{K} \cdot \vec{r}) \quad (5.12)$$

It should be noted that although the recording process will give rise to higher harmonics but their effect is negligible if we are only concerned with diffraction in the vicinity of the Bragg angle.

At $t = 0$ both beams will be incident. The boundary and initial conditions are therefore

$$\alpha(x, y, 0) = \alpha_1, \quad R(0, t) = I_1, \quad S(0, t) = I_2 \quad (5.13)$$

where $y$ is the transverse coordinate, and $I_1$ and $I_2$ are the input intensities of beams 1 and 2 respectively.

It is now possible to apply the method described in Sec. 5.2 to the two beam case. At $t = 0$ the attenuation is uniform and thus the two beams propagate independently of each other in the lossy medium. Their amplitudes can therefore be written as

$$R = I_1 \exp - \frac{\alpha_1 x}{\cos \theta} \quad \text{and} \quad S = I_2 \exp - \frac{\alpha_1 x}{\cos \theta} \quad (5.14)$$

and the intensity is then given by Eqn. (5.9) (note that the grating vector $\vec{K}$ is defined by the interference pattern although there is no grating as yet). For the next $\Delta t$ interval we can now work out the increase in absorption at every point in the medium. The resulting $\alpha(x, y, \Delta t)$ is a periodic function in the $y$ direction since it is a response to the periodic variation of the interference pattern. It would then be possible to determine $\alpha_0$, the average value, and $\alpha_1$, the amplitude of the fundamental component of the absorption coefficient, both functions of $x$, by a Fourier expansion of $\alpha(x, y, \Delta t)$ in the $y$ direction but this would be very time consuming. However the values of
\( \alpha_0 \) and \( \alpha_1 \) obtained from the Fourier analysis would differ little from the values given by the approximate expressions

\[
\alpha_0 = \frac{\alpha_{\text{max}} + \alpha_{\text{min}}}{2} \quad \text{and} \quad \alpha_1 = \frac{\alpha_{\text{max}} - \alpha_{\text{min}}}{2}
\]  \hspace{1cm} (5.15)

where \( \alpha_{\text{max}} \) and \( \alpha_{\text{min}} \) are the values of \( \alpha(x,y,\Delta t) \) at the maxima and minima of the interference pattern and are functions of \( x \). It is therefore only necessary to calculate \( \alpha(x,y,\Delta t) \) for two values of \( y \), corresponding to the positions of the interference maxima and minima, to obtain \( \alpha_0 \) and \( \alpha_1 \) for each value of \( x \).

It is now possible to use Eqns. (5.10) and (5.11) to determine the \( x \) variation of \( R \) and \( S \) due to the absorption grating recorded during the period \( \Delta t \). For the next \( \Delta t \) interval the rate of increase of absorption depends on the intensity distribution obtained with the new values of \( R \) and \( S \). This process can be repeated to obtain the values of intensity and absorption at any position and time. However, there is a further complication to take into account because the experimental curves in Figs. 5.5 and 5.6 were obtained by chopping beam 2 periodically. Thus if our theory intends to describe the experimental results we must take into account that only one beam is present for a finite time period, which will actually lead to some erasure of the existing grating.

The mathematical problem appears in the form that when beam 2 is blocked (say at \( t = t_b \)) the boundary conditions suddenly change. The input to the differential equations is then given in the form of an initial condition (the spatial variation of \( \alpha \) at \( t_b \)) and the boundary conditions

\[
R(0,t_b) = \int I_1 \quad \text{and} \quad S(0,t_b) = 0
\]  \hspace{1cm} (5.16)

From these new conditions we may then calculate the spatial distribution of \( R \) and \( S \) from Eqns. (5.10) and (5.11) and then the increase in absorption constant from Eqns. (5.9) and (5.4), etc. When
beam 2 is again incident, (say at \( t = t_c \)), we return to the boundary condition for beam 2 as \( S(x, t_c) = \sqrt{I_2} \).

This model was implemented and was used to match the experimental results by varying the values of the free parameters \( \alpha_1, \alpha_f, \) and \( \gamma \). The theoretical curves produced by the model are shown in Figs. 5.5 (b,d) and 5.6 (b,d), and the parameters used in each case are tabulated in Table 5.2.

The agreement between theory and experiment is not as good as for the single beam case, but still, the theoretical curves show all the major features of the experimental results. The individual values of \( \alpha_f \) and \( \alpha_1 \) and their ratio may be seen to be reasonable close to those of the single beam results given in Table 5.1. Unfortunately, for \( \gamma \) there is a factor of discrepancy as large as 2. The reason may be partly that the unexpanded Gaussian beams were not well defined so their diameter could not be accurately determined, and also the intensity distribution in the beam was not uniform, in contrast to the assumption of the model. A further possibility is reciprocity failure reported earlier which means that \( \gamma \) depends on intensity [CHA79].

It is interesting to note that the exposures required to convert all the dichromate to \( \text{Cr}^{3+} \) are very high, about 25 J/cm\(^2\). This is about 20 times larger than the exposure needed to reach saturation when dichromated gelatin is used as a latent recording material and is developed in the usual way. This suggests that the saturation of developed gratings is not dependent on the amount of dichromate present, much of which remains unconverted during exposure, but that it is the number of active sites available to the \( \text{Cr}^{3+} \) or the physical properties of the gelatin which determine the maximum efficiency that can be achieved.
5.5 THE APPEARANCE OF PHASE MODULATION.

It has been shown in the previous sections that our experiments can be explained on the assumption of a pure absorption grating. Some of the results (that the detected power decreases when both beams are present and that vibration of a mirror always leads to increase in detected power) suggest quite unambiguously that the dominant mechanism of diffraction must be an absorption grating. If a phase grating is also present, it must be extremely weak.

There was no change in the properties of the grating if it was stored in an atmosphere with the same relative humidity (R.H. = 40%) as that in which the gelatin was sensitised, dried and exposed. However, when the recorded absorption gratings were stored in an atmosphere of higher relative humidity (R.H. = 50% - 60%) a phase grating with an efficiency of up to 15% appeared in the course of a few hours.

Using the apparatus shown in Fig. 3.9 measurements were made of the transmission of the gratings as a function of angle. An example is shown in Fig. 5.8, taken 24 hours after the exposure. The diffraction efficiency may be directly estimated from the depth of the Bragg dip (the loss of power in the transmitted beam when the Bragg conditions are satisfied) and comes to about 8%.

The theoretical curve was generated with the aid of a theoretical model based on a one-dimensional coupled wave theory for uniform transmission holograms [SYM83], implemented by Slinger [SLI85]. Although this model does not allow for a spatial variation of the grating parameters it should give a reasonable indication of the relative magnitude of the average phase and absorption modulations. The best agreement with the experimental results was obtained with the following values of the parameters:
\[ \alpha_0 = 1.8 \times 10^{-4}/m, \quad \alpha_1/\alpha_0 = 0.37 \]

and

\[ \epsilon_{r0} = 2.40, \quad \epsilon_{r1}/\epsilon_{r0} = 4.7 \times 10^{-3} \]

where \( \epsilon_{r0} \) is the average dielectric constant of the gelatin, \( \alpha_0 \) is the average absorption and \( \epsilon_{r1} \) and \( \alpha_1 \) are the respective modulations.

It is believed that the phase modulation obtained is produced by a mechanism similar to that found in dichromated gelatin holograms washed in water after exposure, but not fully developed by drying in propanol \( [MEY71] \). In the exposed regions the gelatin is hardened by \( Cr^{3+} \) ions formed during the photochemical reaction. When an exposed gelatin layer is placed in an atmosphere of increased relative humidity it will absorb water and swell. However, due to the hardening process described, it will swell less in exposed regions than in unexposed regions. It is thought that it is this differential swelling which produced the phase modulation. This hypothesis is supported by the theoretical results which show that the phase modulation is in phase with the absorption modulation, i.e. exposure results in increased absorption, but also hardening, reduced swelling, denser gelatin and thus a higher refractive index than in the less exposed regions.

These mixed phase and absorption gratings were still diffracting strongly after a year in an uncontrolled environment. There was some increase in absorption due a dark reaction, which was worsened by prolonged exposure to sunlight, but the gratings are surprisingly stable.
5.6 SUMMARY.

It has been shown that real-time gratings may be recorded in dichromated gelatin. The maximum efficiency obtained was 0.7%. They have been identified unambiguously as pure absorption gratings by comparing the experimental results with those of a theoretical model.

It may be possible to use these gratings in practical applications but the exposures required are very high. The real-time behaviour could be used as a diagnostic method to investigate the effect of different parameters on the rate of the photochemical reaction and on the rate of the dark reaction.

In addition, it has been shown that if the recorded absorption grating is stored in an atmosphere of higher relative humidity than that during preparation and recording, then a phase grating with an efficiency of up to 15% appears without any further development.
CHAPTER 6.

COPYING AND NOISE GRATINGS IN DICHROMATED GELATIN.

The aim of this chapter is to investigate the quality of dichromated gelatin holograms produced by copying silver halide holographic optical elements and display holograms. In particular the level of noise and scatter seen in the original and in the copy will be compared. It will be seen that a dichromated gelatin copy can be more efficient and produce less noise than the original hologram.

On the basis of some interesting behaviour seen in copies of silver halide planar reflection gratings, dichromated gelatin is used to investigate the behaviour of noise gratings, evidence of which has been seen in other recording materials and which are not yet fully understood.

6.1 MOTIVES FOR COPYING HOLOGRAMS.

The production of a copy of a hologram may be motivated by the fact that the original recording configuration or subject matter has become unobtainable. However copying techniques also give great flexibility: It is possible to copy into a different gelatin thickness or into a different recording material; the reference beam angle or the beam ratio can be altered and it may be possible to use a different wavelength for copying. This flexibility makes copying useful in a number of applications.
For example, copying can be used to increase the efficiency of weak holograms [PAL71]. Ruzek recorded low efficiency holographic portraits using only weak illumination of the subject to avoid eye damage and then produced a high efficiency hologram by copying [RUZ79].

If a hologram is replayed with the conjugate of its original reference beam, a real image can be produced. However, this real image is pseudoscopic, i.e. parallax and relief appear inverted. Holograms which can produce a non-pseudoscopic real image (or a pseudoscopic virtual image) can be obtained by copying this pseudoscopic real image [ROT66].

Computer generated holograms (CGH) can be made which produce arbitrary aspheric wavefronts. However, because they are optically thin, binary absorption holograms they are inefficient and produce many harmonic diffraction orders. An optically thick phase hologram of high efficiency can be made by copying the wavefront generated by a CGH [HAR66, FAI82].

Copying can be used to eliminate some of the problems caused by the insensitivity of dichromated gelatin in recording configurations where the intensity of the recording beams is low and the potential for instability is high [MEY72]. In these circumstances a master hologram can first be recorded in a more sensitive recording material such as silver halide. A copy can then be made in dichromated gelatin using one of the contact copying methods which are very stable and which make efficient use of the available light.

In Sec. 6.8 it will be seen that copying can also be used to generate display holograms of different colours from a green original using only green light for exposure.
6.2 COPYING TECHNIQUES.

Hologram copying techniques can be divided into two classes: Those in which the original and copy are in close contact and those in which they are spatially separated.

6.2.1 Contact copying methods.

The contact copying of holograms was first attempted for transmission holograms \[\text{HAR66, BRU66}\] and later for reflection holograms \[\text{BEL67, KUR68}\]. The arrangements used for transmission and reflection holograms are very similar and are shown in Fig. 6.1 and Fig. 6.2 respectively. The master hologram and photosensitive copy plate are clamped together, separated by a thin layer of index matching liquid to eliminate spurious reflections and fringeing. In general, copies produced with this method are very similar to the original hologram except that the exposure and reference beam angle can be changed and the beam ratio may be different. In both reflection and transmission copying, the plates are illuminated with a single reference beam.

In the reflection case, the incident light passes through the copying emulsion and is the reference beam for the copy as well as the replay beam for the master hologram. Because of this, best results are obtained if the absorption in the recording material used for the copy hologram is low and if the efficiency of the master is high. In the transmission case, the undiffracted zeroth order of the master hologram is the reference beam for the copy. If the intensity of this undiffracted beam is not uniform, the efficiency of the copy will be modulated and therefore best results are obtained if the efficiency of the master hologram is deliberately kept low \[\text{MEY72}\].

Because the separation of reference and signal beam takes place in the emulsion of the master hologram, which is in close contact with the copy plate, a low coherence illuminating source can be used. In the case
Fig. 6.1 Contact copying method for transmission holograms.

Fig. 6.2 Contact copying method for reflection holograms.
of thin transmission holograms, copying with white light is possible (a process analogous to contact printing in photography) but minimising the separation of the two emulsions is critical and a vacuum printing press is necessary [HAR66].

Because only the stability of the relative position of the master and copy plate is critical, these methods are relatively immune to air currents and the stability of the illuminating optics. This means that very long exposures become feasible. Additionally, because only a single collimated beam is required, contact techniques are very efficient in their use of light. These features of contact copying allow the use of a wide range of illuminating sources (such as filtered discharge lamps [OLI82]) and the use of low sensitivity recording materials. Mass production of holograms could be simplified by recording multiple copies of a single master hologram by contact techniques rather than recording a series of originals with a more complicated recording arrangement and an expensive light source.

6.2.2 Non-contact copying methods.

Non-contact methods do not have the advantages of simplicity or stability described for contact methods and require separate reference beams for master hologram and copy plate. However, because this allows the beam ratio to be altered freely, no restrictions are placed on the efficiency of the master hologram or the absorption in either master or copy.

It is possible to place the copy plate in any position with respect to the wavefront produced by the master hologram and therefore the copy is not a replica of the master in the same sense as in a contact copy. It becomes possible to isolate the copy from higher orders generated by a thin master hologram (such as a C.G.H) and it is possible to record a reflection hologram copy from a transmission master hologram.
In the case of display holograms the field of view of the image may be altered and it is possible to position the copy plate within the image itself.

6.2.3. The quality of hologram copying.

The quality of a copy may be below that of the original master hologram due to distortion of the desired wavefronts, changes in the spatial frequency spectrum or due to the presence of additional noise. The quality of copy holograms was first investigated by Landry [LAN67] who examined the effect of changing the spacing between master and copy and the effect of changing the reference beam angle on the resolution, noise and loss of information in the copy.

Distortion can be minimised by replaying the master hologram with the exact reference beam that was used to record it. However if there has been a thickness change during processing and the master is a thick, selective hologram it may be necessary to change the replay angle since the Bragg angle will have changed. This will introduce distortions or aberrations and any further thickness change during the processing of the copy will increase the problem. It is also possible that a thickness change may make it impossible to replay all gratings in the master hologram simultaneously, using monochromatic light, leading to a loss of information. It is therefore desirable to use a master hologram with a low angular selectivity and to minimise any thickness change.

The modulation transfer function of the copying process has been investigated theoretically by Kaspar [KAS74] and both theoretically and experimentally by Suhara et al [SUH75]. In general, if the Bragg condition can be satisfied simultaneously for all the gratings in the hologram, the modulation transfer function will be limited only by the response of the recording material used for the copy hologram.
To discuss the effects of noise in copying it is necessary to describe noise in holography in general. The major sources of noise in holograms are: scattered light due to inhomogeneities in the recording material \cite{Goo67, Pen70, KoZ68}; intermodulation due to a non-linear recording response \cite{Fri67, Goo68, KoZ70} and speckle \cite{Mck75}. The level of scatter in dichromated gelatin is very low (unless the temperature used during processing is too high or the gelatin is insufficiently prehardened \cite{Bra69}) but in silver halide and other recording materials, scatter can be the predominant source of noise. The effects of 'primary' scatter due to inhomogeneities in a processed hologram at replay are obvious. However, it is not generally realised that scatter at the recording stage can also have a considerable affect on the behaviour of a hologram due to the formation of spurious gratings called noise or scatter gratings \cite{Sym82b}. The scattered light field present during the recording of a hologram will result in the recording of a large number of weak noise gratings which may then reconstruct the scattered light field at replay. These noise gratings will exhibit Bragg like selectivity and will therefore be termed 'Bragg scatter'. Fortunately, the level of scatter in dichromated gelatin at the recording stage is very low and therefore very little Bragg scatter might be expected after processing.

When a copy is recorded, primary and Bragg scatter from the master hologram may be recorded in addition to any scatter in the copy recording material. At replay of the copy hologram, Bragg scatter of all these contributions will be seen in addition to primary scatter from the processed copy recording material. The content of the final noise field of a copy hologram is therefore complex and will be affected by the properties of the recording materials used for the master and for the copy.
6.3 COPYING SILVER HALIDE PLANAR REFLECTION GRATINGS.

Dichromated gelatin is an ideal recording material for contact copying due to its low absorption and scatter. Previous work which has involved copying silver halide holograms with dichromated gelatin has concentrated on display holograms. Meyerhofer increased the efficiency of transmission display holograms by copying [MEY72] and Olivia et al found that copying with a relatively low coherence light source could reduce speckle noise [OLI82]. However, for this study we consider a dichromated gelatin copy of a silver halide planar reflection grating.

6.3.1 Recording procedure.

A silver halide reflection hologram for use as the master was recorded in Agfa 8E56HD emulsion and processed by the method described by Cooke and Ward [CO084]. The recording angles were 44° and 46°, measured in air, so that the grating was slightly slanted. A diffraction efficiency of 69.3% was obtained. A contact copy was then made using the arrangement shown in Fig. 6.2. The copy plate was an 20 μm gelatin layer sensitized in 2% ammonium dichromate. The use of a low sensitizer concentration is preferable to reduce absorption - the transmission of this plate at the recording wavelength, 514.5 nm, was 86.8%. The two plates were clamped together and placed in a tank of index matching fluid to eliminate all possible spurious reflections from the external surfaces. The angle of the plates was set so that the silver halide master hologram would be exactly at the Bragg angle during the exposure. An exposure of 800 mJ/cm² was chosen and the beam ratio, set by the diffraction efficiency of the master hologram, was 1 : 0.69.

After carefully separating the two plates, the dichromated gelatin copy was processed with the standard procedure described in Sec. 2.8 using an initial processing temperature of A = 25°C and a final processing temperature of B = 40°C to maximise modulation and to amplify
any interesting spurious signals from the silver halide master hologram. An interesting comparison can be made between the copy hologram and hologram B/8 (seen in Chapter 3) because one is a copy and the other an 'original', recorded in the same thickness gelatin, with the same exposure and similar processing.

6.3.2 Replay at the recording wavelength.

The angular response of the master and copy holograms was measured using the apparatus shown in Fig. 6.3. This is similar to the spectrometer apparatus used for measurements in earlier Chapters (Fig. 3.9) except that the use of an unexpanded laser beam allows the power in both the transmitted and the diffracted beam to be measured. The data obtained was processed to correct for the Fresnel losses at the glass tank walls and is shown in Fig. 6.4 and Fig. 6.5. The spectrometer apparatus was used to measure the transmission of the two holograms over a wider range of angle and the results can be seen in Fig. 6.6.

The bandwidth of the copy is smaller due to the greater gelatin thickness and the replay angles differ because of a thickness change in the copy during processing. It can be seen that the diffraction efficiency of the copy (94.9%) is higher than the master hologram and the grating is overmodulated. The attenuation of the transmitted light, due to absorption and scatter, is much lower in the copy than in the master. However the diffraction efficiency is slightly lower than that of hologram B/8 and the losses are slightly higher. The performance of the master, copy and hologram B/8 is summarised in Table 6.1.

It is difficult to make a quantitative comparison of the level of scatter in the three holograms because, using monochromatic light, it can be seen that the scatter in the copy is directional and the angular distribution of scatter is different for each case (Sec. 6.6). Using white light illumination, a more uniform scatter field is seen and a
Fig. 6.3 Measurement of the angular response using an unexpanded laser beam. Photodiode 1 measures the power in the transmitted beam and photodiode 2 measures the power in the diffracted (-1) order.

Fig. 6.4 The angular response at 514.5 nm of the master hologram for the planar grating copying experiments - a 5 μm silver halide planar reflection grating.
Fig. 6.5 The angular response at 514.5 nm of the 20 μm dichromated gelatin planar reflection grating copy.

Fig. 6.6 Comparison of the angular response at 514.5 nm of the master and copy planar reflection gratings.
simple measurement of the relative level of scatter was made using the arrangement shown in Fig. 6.7. The results are given in Table 6.1. It was found that the scatter from the copy was much lower than that from the master but that it was higher than that from the original hologram B/8.

The reason why the copy produces less scatter than the original hologram is because the level of primary scatter in dichromated gelatin is very low. The scatter in the DCG copy will therefore be primarily due to Bragg scatter (recorded scatter from the silver halide master) with a small level of primary scatter (similar to that seen B/8), whilst the scatter in the silver halide master hologram will consist of considerable primary scatter as well as Bragg scatter. However primary scatter is a stronger source of noise under white light illumination since it will scatter light from any angle and at any wavelength whereas Bragg scatter will only replay at a particular angle for each wavelength because it displays Bragg like selectivity. Because of this, the overall scatter from the master is much higher than from the copy hologram.

It is relevant to note that the scatter from unexposed regions of the silver halide hologram was found to be about 70% of the level measured in the exposed region, indicating that the level of primary scatter is high.

6.3.3 Replay at other wavelengths.

The performance of the copy was then examined at other wavelengths using the spectrometer and white light source (Fig. 3.9). Weak coloured scatter could be seen emanating from the region of the exposure. The colour of this scatter appeared to change if viewing angle, or the angle of the incident light, was varied. However, coloured scatter of this nature could not be seen in hologram B/8 or in areas of the copy which
Fig. 6.7 Simple arrangement for measuring the level of scatter of the master and copy planar reflection gratings when replayed with white light using a large area photodiode (1cm²).

Fig. 6.8 Angular response of the planar reflection grating copy at 455 nm showing the spurious diffraction due to noise gratings at ≈ 20°.
### TABLE 6.1

**COMPARISON OF THE PROPERTIES OF PLANAR REFLECTION GRATINGS: SILVER HALIDE MASTER, DCG COPY AND HOLOGRAM B/8.**

<table>
<thead>
<tr>
<th></th>
<th>HALIDE MASTER</th>
<th>DICHROMATED GELATIN COPY</th>
<th>HOLOGRAM B/8. (DCG ORIGINAL)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>THICKNESS (μm)</strong></td>
<td>5</td>
<td>20</td>
<td>21</td>
</tr>
<tr>
<td><strong>MODULATION (n₁)</strong></td>
<td>0.062</td>
<td>0.045</td>
<td>0.033</td>
</tr>
<tr>
<td><strong>DIFFRACTION EFFICIENCY.</strong></td>
<td>69.3%</td>
<td>94.9%</td>
<td>98.2%</td>
</tr>
<tr>
<td><strong>DEPLETION OF TRANSMITTED BEAM ON BRAGG.</strong></td>
<td>96.2%</td>
<td>100%</td>
<td>100%</td>
</tr>
<tr>
<td><strong>OFF-BRAGG TRANSMISSION. (NORMAL INCIDENCE)</strong></td>
<td>58.1%</td>
<td>91.5%</td>
<td>93%</td>
</tr>
<tr>
<td><strong>RELATIVE SCATTER FOR WHITE LIGHT REPLAY.</strong></td>
<td>1.0</td>
<td>0.172</td>
<td>0.066</td>
</tr>
</tbody>
</table>
where not exposed at recording and therefore it must be the result of the scatter recorded in the copy.

At a wavelength of 455 nm a single very sharp dip resulting in loss of almost 20% of the incident light can be seen at an angle of +21.5° (Fig. 6.8) but a similar dip cannot be found at negative angles. This angle corresponds to the angle of the diffracted wave from the silver halide master hologram at recording. At wavelengths longer or shorter than 455 nm this single dip splits into two weaker dips either side of 21.5° (Fig. 6.9 a-d). No spurious dips could be seen at 514.5 nm because of the position of the main Bragg dip due to the planar grating. At 595 nm where the plane grating replays as a single dip at normal incidence there is again some evidence of spurious dips at positive angles. There is no evidence of similar behaviour in hologram B/8.

The shape of the strong sharp dip seen at 455 nm is very different to the characteristic shape of the angular response of a planar grating. Additionally, the manner in which this dip splits into two weaker dips and the positions at which these dips are found as the replay wavelength is changed cannot be explained in terms of any single reflection or transmission grating. However, similar behaviour has been observed in single beam exposures in silver halide emulsions [SYM82B, SYM83B, WAR84] and has been ascribed to diffraction by noise gratings which are recorded because of interference between the single incident reference beam and light scattered in the recording material. Similar noise gratings must therefore be present in the copy hologram. The strong dip at 455 nm corresponds to the simultaneous replay of a large number of these gratings, thereby reconstructing some of the scattered light field from the master hologram, present at the time of recording. The theoretical model developed later in this Chapter will show that thickness changes
Fig. 6.9 Angular response of the planar grating copy at other wavelengths showing the behaviour of the spurious noise gratings.
COPYING AND NOISE GRATINGS IN DICHROMATED GELATIN. 6-11

and refractive index changes in the recording material determine the wavelength and angle at which this strong diffraction is found.

It is difficult to see if there is similar behaviour in the master hologram because of the wide bandwidth of the main Bragg dip, which covers the area of interest for a wider range of wavelength. However, some evidence of the effects of noise gratings in silver halide planar gratings has been reported [HEA85, SLI85].

6.4 SINGLE BEAM RECORDINGS.

The question arises that if a single beam exposure in silver halide results in strong diffraction due to noise gratings, can similar behaviour be seen in dichromated gelatin? To investigate this, sensitized dichromated gelatin plates were placed in an index matching tank and exposed to a single beam of light (514.5 nm at 30°) for different exposures. Care was taken to ensure no reflected beams from the tank walls, or scattered light from optical components, reached the plate.

After processing, the plates were analysed at several wavelengths using the spectrometer apparatus, but no sign of strong diffraction could be detected. An example of the angular response is shown in Fig. 6.10. The small dips at ± 27° are due to a weak plane grating which has been recorded because there are weak reflections at the substrate, gelatin and index-matching fluid boundaries. This explanation can be confirmed by analysing the change in position of these weak dips at different wavelengths. In contrast the angular response of a silver halide single beam exposure is also shown (provided by A. A. Ward). This was recorded at 514.5 nm and is shown replayed at 518.5 nm. The radical difference between the results obtained from the single beam exposures is due to the fact that the level of scatter in sensitized dichromated gelatin is very
Fig. 6.10 Angular response of a single beam exposure in dichromated gelatin (514.5 nm) and in silver halide (518.5 nm).

Fig. 6.11 (a) Recording arrangement and Ewald sphere representation for noise gratings in silver halide. All possible $\mathbf{K}$ vectors lie between point $P$ and the surface of the Ewald sphere.
Fig. 6.11 (b) Recording arrangement and Ewald sphere representation for transmission noise gratings in dichromated gelatin. All possible $K$ vectors lie between point $P$ and the surface of one half of the Ewald sphere.

Fig. 6.11 (c) Recording arrangement and Ewald sphere representation for reflection noise gratings in dichromated gelatin. All possible $K$ vectors lie between point $P$ and the surface of one half of the Ewald sphere.
low during recording. All the losses in the material are due to absorption and not scatter, and therefore a single beam exposure results in a uniform refractive index change, rather than the formation of noise gratings. The level of scatter in silver halide emulsions is very much higher and an antihalation dye is often incorporated to increase absorption and reduce the effects of scatter during recording.

The 'failure' of the single beam recordings in dichromated gelatin suggest that the noise gratings detected in the planar grating copy holograms must result from the recording of scattered light from the silver halide master hologram.

6.5 SYNTHESISED NOISE GRATINGS IN DICHROMATED GELATIN.

The noise gratings detected in the planar grating copy holograms are of particular interest because the effects of Bragg scatter have been isolated from the original source of the scatter, which would otherwise result in the addition of primary scatter at replay. We can assume that all the visible scattered light in the copy is due to Bragg scatter by noise gratings because the level of primary scatter in the processed dichromated gelatin is very low.

Examining the characteristics of the reconstructed noise field from the copy under different replay conditions may yield information which will help explain the complex behaviour of noise gratings and why strong diffraction is seen at a unique angle and wavelength. However, the planar reflection grating copy contains, primarily, a planar grating which is an unnecessary complication. It would be simpler to examine the behaviour of a hologram which is a copy of the noise field alone. A suitable name to describe this would be a synthesised noise grating.
6.5.1 Recording synthesised noise gratings.

Suitable arrangements for recording a synthesised noise grating in dichromated gelatin by contact copying are shown in Figure 6.11. Scatter is obtained from an unexposed, unprocessed Agfa 8E56HD silver halide plate, complete with sensitizing and antihalation dyes, and thus the recorded scatter is very similar to that present in a silver halide hologram during exposure.

The amount of scatter at high angles in the dichromated gelatin during recording is limited by the antihalation dye in the 8E56 emulsion and because the refractive index of the thin layer of index matching fluid between the plates is slightly lower than that of the dichromated gelatin.

In a single beam recording in silver halide all the possible $K$-vectors of the recorded noise gratings lie between on the surface of the Ewald sphere and the point $P$ at the end of the recording wavevector (Fig. 6.11a). The character of the noise gratings will therefore range from optically thin transmission gratings of low selectivity to optically thick reflection gratings of high selectivity, assuming that light is scattered in all directions.

When a synthesised noise grating is recorded it is possible to record a population of transmission noise gratings or a population of reflection noise gratings depending on the relative position of the silver halide and dichromated gelatin plates. The locus of permitted $K$-vectors will therefore be half the surface of the Ewald sphere in each case (Fig. 6.11b,c) and thus copying allows the affect of the two components to be studied separately.

If a non-contact method was used, it would be possible to select a particular section of the noise field to be recorded in the copy and therefore the individual contribution of each section to the total level
of diffraction could be found. However this study will be limited to contact copying.

6.5.2 Transmission noise gratings.

A set of transmission noise gratings were recorded in 22 μm gelatin at 19.5° in the index matching tank and were processed with the standard recording procedure using an initial processing temperature, $A = 25°C$ and a final processing temperature, $B = 30°C$. When the gratings were examined in a beam of white light, strong forward scatter could be seen but very little backscatter. The colour of the scattered light changed if replay angle or viewing angle was varied in a similar manner to the behaviour seen in the planar copy hologram.

The angular response of the transmission noise gratings was measured at several wavelengths using the spectrometer and some examples of the results are shown in Fig. 6.12 & 6.13. This particular noise grating was recorded in gelatin sensitized in 6% ammonium dichromate, with an exposure of 1200 mJ/cm$^2$. The observed behaviour is very similar to that seen in the planar copy hologram and in the single beam exposures in silver halide, despite the fact that the reflection noise gratings are absent. The small dips which can be seen in some graphs are due to weak planar reflection gratings which result from boundary reflections.

At the wavelength at which a single dip occurs (462 nm ± 3 nm), which I term $\lambda_s$, there is now almost total depletion of the incident light. The angular bandwidth of this single dip is much narrower than would be expected for a planar grating recorded in the same thickness gelatin with recording angles of ±30°. At longer or shorter wavelengths the angular response shows two dips which increase in separation and decrease in efficiency as the difference between replay wavelength and $\lambda_s$ increases. There is still considerable diffraction at 370.0 nm and 600 nm, but the angular bandwidth is very much larger. The angles at
Fig. 6.12 The angular response of a transmission noise grating in dichromated gelatin at different wavelengths up to $\lambda_s$ (462 nm).
Fig. 6.13 The angular response of a transmission noise grating in dichromated gelatin at wavelengths longer than \( \lambda_s \).
which minima occur in the transmitted intensity are plotted against wavelength in Fig. 6.14.

The reduction in the intensity of the transmitted light seen in the angular response is a measure of the total amount of light diffracted by the population of noise gratings within the hologram. The total amount of diffraction will depend on the amplitude of the modulation of the gratings but will also depend strongly on what proportion of the total number of gratings are on Bragg (or close to their Bragg angle) at each incidence angle and wavelength. If there was no thickness or refractive index change in the recording material during processing it would be possible to replay all the gratings at the original recording angle and wavelength. However, if a thickness change does occur it will no longer be possible to replay all the gratings simultaneously under any replay conditions (see Sec. 6.6.2).

To investigate the effects of thickness and refractive index changes during processing, six sets of noise gratings were recorded in gelatin sensitized in different concentrations of ammonium dichromate. Changing the sensitizer concentration was shown to have a strong affect on the level of thickness change in Chapter 3. The angular response at $\lambda_s$ for three of these sets are shown in Fig. 6.15 - 6.17. It can be seen that the diffraction efficiency for a particular exposure rises rapidly as the sensitizer concentration is increased. The maximum depletion of the transmitted beam is plotted against sensitizer concentration in Fig. 6.18. The amplitude of the modulation of the noise gratings might be expected to increase by a factor of up to 2.5 if the sensitizer concentration is increased from 2% to 12%, due to increased sensitivity [CHA79]. However, this is not sufficient to explain the rapid increase in efficiency which is observed. An increase in exposure by the same factor, which is analogous to increasing sensitivity, does
Fig. 6.14 The angles at which minima occur in the angular response of a transmission noise grating against wavelength. A single minimum is found at 462 nm.

Fig. 6.15 The angular response at $\lambda_0$ (= 460 nm) of transmission noise gratings sensitized in 2% ammonium dichromate.
Fig. 6.16 The angular response at $\lambda_0 (\approx 460 \text{ nm})$ of transmission noise gratings sensitized in 6% ammonium dichromate.

Fig. 6.17 The angular response at $\lambda_0 (\approx 460 \text{ nm})$ of transmission noise gratings sensitized in 10% ammonium dichromate.
not lead to such a rapid rise in efficiency. The rapid rise is probably due to the fact that the thickness change during processing is large for gelatin sensitized in low concentrations of ammonium dichromate but is small for the higher concentrations. A higher fraction of the noise gratings can therefore be replayed simultaneously when a high sensitizer concentration is used.

The shape of the high exposures in 10% sensitized gelatin is particularly interesting (Fig. 6.17). It is thought that the bottom of these curves may correspond to total diffraction of the incident light but a small offset is seen because there is strong scatter in the direction of the spectrometer entrance slit. Diffraction is seen over a wide angular range but unlike overmodulated planar transmission gratings, the efficiency does not appear to fall at high exposures and unlike planar reflection holograms, there is no flat bottom to the angular response at high exposures.

The wavelength required for a single dip did not vary significantly with exposure or with sensitizer concentration and it was not possible to see any conclusive trend because it is difficult to determine \( \lambda_s \) exactly. It is essential to mount the holograms in a rotation stage with precise adjustment on all axes to obtain maximum diffraction due to the high angular selectivity. This is not usually necessary for planar gratings because the Bragg angle describes a cone of possible replay directions about the grating normal. However, in the case of a noise grating there is only one possible replay vector direction at \( \lambda_s \).

6.5.3 Reflection noise gratings.

Several attempts were made to record reflection noise gratings but no strong diffraction was found when these recordings were replayed and there is very little evidence of the effects of these gratings. When
the holograms were examined in white light it was possible to see some backscattered light but this was much weaker than the forward scatter observed in the transmission noise gratings. No forward scatter was visible and the backscatter had the familiar property that the colour changed as replay angle or viewing angle was altered.

The angular response of a typical example of a reflection noise recording is shown in Fig. 6.19. This hologram was recorded at 30° in the index matching tank using 20 μm gelatin sensitized in 2% ammonium dichromate. At all wavelengths only weak dips can be seen and these are in the positions which would be expected if they were caused by a weak unslanted planar reflection grating, recorded due to boundary reflections. However a recurring characteristic of these measurements is that the profile of the angular response is asymmetric. This asymmetry was reproducible and all other possible experimental causes were eliminated. It is perhaps significant that there is always a greater reduction of the transmitted intensity at positive angles because the recording angle was positive.

The level of back scattered light from the 8E56 emulsion might be expected to be lower than the level of forward scatter but this is not sufficient to explain the radical difference between the behaviour of the transmission and reflection noise gratings. A more likely reason is the much higher selectivity of the reflection noise gratings. This will make it difficult to replay many of the gratings simultaneously if thickness changes have occurred. The scatter replayed in the transmission noise gratings is predominantly in the direction of the undiffracted transmitted light and therefore the gratings which cause the scatter are mostly optically ‘thin’ and unselective. However in the reflection case the noise gratings are all highly selective volume gratings.
Fig. 6.18 The maximum depletion of the transmitted beam against sensitiser concentration for the set of transmission noise gratings.

Fig. 6.19 The angular response at 514.5 nm of a reflection noise grating. Note the vertical scale.
From these observations it can be concluded that the behaviour seen in single beam recordings in silver halide emulsions is predominantly due to transmission noise gratings. It is also possible to explain why diffraction due to noise gratings was visible in the planar copy hologram when it was replayed in the direction of the diffracted beam from the silver halide emulsion rather than the direction of the reference beam - interference between the scattered light and the diffracted beam would result in the recording of transmission noise gratings whilst the reference beam and scattered light would result in reflection noise gratings.

6.6 ANGULAR DISTRIBUTION OF SCATTERED LIGHT.

6.6.1 Measurement of distribution.

In order to investigate the angular distribution of the scattered light from the transmission noise gratings they were illuminated with an unexpanded laser beam at 514.5 nm (Fig. 6.20). The scattered light was projected on to a translucent screen (position A) and could be photographed. At the replay angle corresponding to the minima in the angular response, a bright uniform circle of scattered light could be seen (Plate 6.1a). However as the replay angle was altered the distribution of the scattered light changed radically. Sharp ring patterns corresponding to cones of scattered light could be seen, particularly at negative angles (Plate 6.1 b-d, 6.2 a-b).

The angular diameter, thickness and intensity of these rings changed if replay angle or wavelength was varied. When replayed in white light, there must be a superimposition of rings corresponding to different wavelengths in the incident spectrum. This would explain the behaviour of the coloured scatter observed in noise gratings and planar copy holograms.
Fig. 6.20 Investigation of the angular distribution of scattered light from noise gratings. Screen A was used for the transmission noise grating and position B for the reflection noise grating.

Fig. 6.21 Recording geometry for noise gratings in two dimensions. $\hat{K}_i$ can be any vector between point P and the circumference of the Ewald circle.
A similar investigation was also made of reflection noise gratings. In order to see the small amount of scatter that was detected it was necessary to reposition the screen to position B. A weak ring of scatter travelling at a high angle of incidence was seen (Plate 6.2c). The dark line corresponds to the edge of the plate. No similar rings could be seen on the other side of the hologram.

Scattering rings due to noise gratings have been previously reported in holograms recorded in polymethyl methacrylate [MOR73], lithium niobate [MAG74] and swollen silver halide emulsions [RAG78] which are all recording materials in which there is scatter during exposure.

6.6.2 Interpretation of scattering rings.

The scattered light is found at angles where the Bragg condition for the noise gratings is satisfied. This occurs at the intersection between the locus of possible K vectors and the replay Ewald sphere. Previous analyses of scattering rings have assumed that this locus is the surface of a sphere [FOR74, MAG74, RAG77]. When the replay angle and wavelength correspond to the recording conditions the Ewald sphere and locus of K vectors coincide exactly and all the noise gratings are replayed simultaneously, leading to strong diffraction. However, if the replay conditions are altered the Bragg condition is only satisfied where the two spheres intersect i.e. on the circumference of a circle. It is this which leads to the cones of scattered light.

If there is a thickness change during processing the locus of K vectors will become an ellipsoid. This requires a more complicated analysis and it is now impossible to replay all the noise gratings simultaneously as it is obviously impossible for the surfaces of a sphere and an ellipsoid to coincide exactly.

The K vectors, \( \hat{k}_1 \), of the possible scatter gratings are defined by
PLATE 6.2

(a) -20°

(b) -30°

(d) Planar grating copy 30°

(c) Reflection type 30°

Planar grating copy
\[ \mathbf{K}_1 = \hat{\mathbf{p}}_1 - \hat{\mathbf{q}}_1 \]  

(6.1)

where \( \hat{\mathbf{p}}_1 \) is the wavevector of the reference beam and \( \hat{\mathbf{q}}_1 \) is the wavevector corresponding to the scattered light which may be in any direction. Considering the recording configuration in two dimensions in Fig. 6.21, \( \mathbf{K}_1 \) will be given by

\[ \mathbf{K}_1 = \beta_1 \left[ \cos \theta_1 - \cos \phi_1 \right] \mathbf{i}_x + \beta_1 \left[ \sin \theta_1 - \sin \phi_1 \right] \mathbf{i}_y \]  

(6.2)

where \( \beta_1 \) is the propagation constant in the recording material, \( \theta_1 \) is the angle of the reference beam and \( \phi_1 \) is the angle of scattered light. For single beam recordings \( \phi_1 \) can be any angle but for transmission type synthesised noise gratings \(-90^\circ < \phi_1 < 90^\circ\). If the thickness changes by a factor

\[ t = \frac{T_1}{T_2} \]  

(6.3)

where \( T_1 \) is the initial thickness and \( T_2 \) is the processed thickness, the \( \mathbf{K} \) vectors of the scatter gratings at replay, \( \mathbf{K}_2 \), are

\[ \mathbf{K}_2 = t \beta_1 \left[ \cos \theta_1 - \cos \phi_1 \right] \mathbf{i}_x + \beta_1 \left[ \sin \theta_1 - \sin \phi_1 \right] \mathbf{i}_y \]  

(6.4)

At replay the Bragg condition will be satisfied if

\[ | \hat{\mathbf{p}}_2 + n \mathbf{K}_2 | = \beta_2 \]  

(6.5)

where \( \hat{\mathbf{p}}_2 \) is the replay wavevector, \( \beta_2 \) the propagation constant in the processed hologram and \( n \) the order of diffraction. Substituting for \( \mathbf{K}_2 \) in (6.5)

\[ \left[ \beta_2 \cos \theta_2 - t \beta_1 \left( \cos \theta_1 - \cos \phi_1 \right) \right]^2 + \left[ \beta_2 \sin \theta_2 - \beta_1 \left( \sin \theta_1 - \sin \phi_1 \right) \right]^2 = \beta_2^2 \]  

(6.7)
where $\beta_2$ is the propagation constant of the replay wavevector. This equation can be solved to find the values of $\phi_1$ which will lead to scattered light at replay. Using the relationship

$$\sigma_{2x} = \rho_{2x} - n K_{2x}$$  \hspace{1cm} (6.8)

which leads to the result

$$\beta_2 \cos \phi_2 = \beta_2 \cos \theta_2 - t_2 \beta_1 \left[ \cos \theta_1 - \cos \phi_1 \right]$$  \hspace{1cm} (6.9)

the angles of the scattered rays at replay, $\phi_2$, corresponding to the intersection of the Ewald circle and ellipse of $K$ vectors can then be found.

The angular diameter of the scattering rings from a transmission noise grating, recorded in gelatin sensitized in 10% ammonium dichromate, were measured at 514.5 and 632.8 nm for a range of replay angles. These angles are plotted in Fig. 6.22 together with theoretical values obtained from the theoretical model using the following values:

- $T_2 = 1.01 \times T_1$
- $T_1 = 1.62$
- $n_1 = 1.62$
- $n_2 = 1.477$
- $n_2 = 1.469$

(514.5nm)
(632.8nm)

Because it was not possible to measure $n_2$ experimentally, it was necessary to use the values from the single beam measurements in Chapter 3. Despite this, reasonable agreement was found. However, it was not possible to explain the angles at which weak scattered light was found in the reflection noise gratings using this theoretical model.

Fig. 6.23 corresponds to the replay configuration of the transmission noise grating in Plate 6.2b. Visualised in 3 dimensions it can be seen that the $+1^{\text{th}}$ order is responsible for the small complete ring and the $-1^{\text{th}}$ order for the segment of large radius. The width of the rings will depend on the angular bandwidth of the individual scatter
Fig. 6.22 Experimental and theoretical vertex angle of noise cones against replay angle for a transmission noise grating in dichromated gelatin.

Fig. 6.23 Replay of a transmission noise grating corresponding to the scattering rings seen in Plate 6.1b. Light is scattered at angles where the broken lines cross the Ewald sphere.
gratings and the slope at which the replay Ewald sphere and ellipsoid of K vectors intersect. If this slope is large, only a small range of gratings will be close enough to the Ewald sphere for significant diffraction and a narrow scatter ring will result. Because the angular response of the individual scatter gratings is a sinc function, weak concentric rings corresponding to sidelobes can be seen surrounding some of the principle scattering rings in Plate 6.1 & 6.2.

Using this simple model it is also possible to see why strong diffraction occurs at only one specific replay wavelength and angle. Fig. 6.24 and 6.25 show the Ewald circle and ellipse of K vectors corresponding to the angle and wavelength, \( \lambda_s \) at which strong diffraction is seen, for the 2% and 10% sensitized transmission noise gratings. In both cases there is a wide angular range in which the circle and ellipse almost coincide, which will mean that a large number of the gratings will be replayed simultaneously and therefore there will be significant depletion of the incident beam. If the replay wavelength is changed to a significant degree it is not possible to obtain such a close match between the circle and ellipse at any replay angle.

A more quantitative analysis of noise gratings has recently been developed in the Oxford Holography group by Riddy and Solymar [RID86] for the low efficiency case, where the loss of power from the incident beam is assumed to be negligible. A quantitative analysis of high efficiency noise gratings is being developed but this is difficult because it is necessary to consider a very large number of coupled wave equations, the solution of which will require a large amount of computation.

6.6.3 Scattering rings in planar grating copy holograms.

Scattering rings were also found in the planar grating copy holograms. These were relatively weak compared to the synthesised noise gratings but they were more complicated in structure. Both the behaviour
Fig. 6.24  Replay conditions corresponding to strong diffraction at $\lambda$ for a synthesised transmission noise grating in 2% sensitized dichromated gelatin ($n_i = 1.567$, $n_f = 1.48$, $t = 0.89$, $\lambda_s = 462$ nm).

Fig. 6.25  Replay conditions corresponding to strong diffraction at $\lambda$ for a synthesised transmission noise grating in 10% sensitized dichromated gelatin ($n_i = 1.62$, $n_f = 1.49$, $t = 0.99$, $\lambda_s = 465$ nm).
seen in transmission type and reflection type noise gratings was observed, due to the fact that there were two strong plane waves present during recording. Because there are also two plane waves present at replay (the reference beam and the diffracted beam) there are two sets of replayed scatter rings.

It was also possible to see a dark ring against the background of scattered light which led to 'missing' sections in some of the scatter rings (Plate 6.2d). The cause of this dark ring was understood when it was seen that there was strong diffraction by the planar grating when this dark ring crossed the position of the incident beam. This suggests that the dark ring corresponds to the Bragg condition of the planar grating. If the Bragg angle is $\theta_b$, any scattered light travelling at $\theta_b$ to the grating normal will be diffracted by the planar grating and therefore there is a cone of missing light.

A possible use for synthesised noise gratings would be to produce very efficient diffuse illumination since transmission type noise gratings can convert almost 100% of the incident light into uniform forward scatter, with no loss into backscatter. The sharp angular response might also be useful in filter applications.

6.7 COPYING DIFFUSE OBJECT HOLOGRAMS.

To make a quantitative study of the copying of display holograms it is most appropriate to copy a hologram of a simple, uniformly illuminated diffuse object. For this study a reflection hologram of a small diffusing screen, recorded in silver halide, was used for the master hologram. At replay it was possible to measure the brightness of the image of the diffusing screen and the level of scattered light in the dark background seen in the holograms. Using these measurements the performance of the master and the copy could be compared.
6.7.1 Recording the master and copy.

The master hologram was originally recorded as part of a study of AGfA 8E56 HD recording material by A. A. Ward. The recording configuration is shown in Fig. 6.26. The object was a 15 mm by 30 mm diffusing screen surrounded by a black mask. A polariser was included to increase the fringe contrast at the recording plate. Some shrinkage occurred during the processing of this hologram and therefore it was swollen by soaking in a 5% sucrose solution so that it would replay at the original recording angle when illuminated at 514.5 nm.

A set of dichromated gelatin copy holograms of differing exposure was recorded using the contact copying method, using 20 μm thick gelatin sensitized in 2% ammonium dichromate and processed with the standard processing procedure (Processing temperatures A = B = 25°C).

6.7.2 Measurement of master and copy performance.

A quantitative comparison of the performance of the holograms using monochromatic illumination is complicated by the effect of thickness changes. Because thickness change during processing is a function of exposure, the set of dichromated gelatin holograms (and the master hologram) have slightly different replay angles. Furthermore, using monochromatic illumination the Bragg scatter from the copies varies rapidly with direction in a similar manner to the ring structures seen previously.

To avoid these problems the performance of the holograms was compared using white light illumination at the original recording angle. The data obtained is therefore relevant to the applications in which diffuse object reflection holograms are likely to be used, i.e. white light replay display holograms. The replay configuration is shown in Fig. 6.27. The white light source was a 50W tungsten halogen light bulb
Fig. 6.26 Recording configuration of the silver halide master hologram for the diffuse object copying experiments.

Fig. 6.27 Apparatus for image brightness and background noise measurements of master and copy diffuse object holograms.
which has a reasonably uniform spectral output (± 5%, 480 - 600 nm) across the wavelength range in which significant diffraction occurs.

The brightness of the image and the level of scatter were measured using a Macam radiometer/spotmeter with a radiometric filter which has a uniform response across the visible spectrum (± 7%, 450 - 950 nm). This instrument was used to measure the intensity of light over a 5mm diameter circular area of the image in three different positions and the intensity of the scattered light in the surrounding background at four positions.

6.7.3 Relative performance of copy and master holograms.

From these measurements the average intensity of the image and the average noise level was calculated for the complete set of copy holograms and the values obtained are plotted in Fig. 6.28, relative to the values measured in the silver halide master. It can be seen that maximum efficiency in the copies is obtained at a similar exposure level as that required for maximum modulation in planar gratings, found in Chapter 3.

The maximum brightness of the image, obtained at an exposure of $2800 \text{ mJ/cm}^2$, is $1.44 \times$ the brightness of the master hologram, despite the fact that the thicker gelatin of the copy holograms will result in greater wavelength selectivity and therefore a narrower reflected spectrum. This apparent anomaly is investigated further in Sec. 6.8.3.

The intensity of the scattered light in the dark background surrounding the diffusing screen reaches a maximum of $0.7 \times$ the level found in the master hologram but does not decline significantly at high exposures. The intensity of the scatter is not zero at zero exposure due to a small level of primary scatter in unexposed gelatin.

It is possible to define the signal to noise ratio of each copy
Fig. 6.28 Image brightness and background noise in the diffuse object copy holograms (relative to the brightness and noise of the master hologram) against exposure for replay with white light.

Fig. 6.29 Signal to noise ratio against exposure for the diffuse object copy holograms for replay with white light. The S/N ratio of the master hologram was 5.18.
hologram as the brightness of the image divided by the brightness of the black background. Calculated values are plotted in Fig. 6.29. A maximum signal to noise ratio of 11.0 is obtained at 800 mJ/cm² compared to a value of 5.18 for the master hologram. At higher exposures the signal to noise ratio decreases due to the fact that the brightness of the image declines whilst the brightness of the scatter does not. This is thought to be due to the non-linearity of the recording characteristic which will distort the amplitude distribution of the gratings recorded in the hologram. This would reduce the amplitude of the modulation of strong gratings relative to weak gratings at high exposures. Another possible mechanism for the reduction in signal to noise ratio at high exposures could be the generation of intermodulation noise due to the non-linear recording characteristic.

The higher performance of the diffuse copy holograms relative to the master hologram can be explained with the same reasoning used in Sec. 6.3.2. The increase in efficiency results from the higher values of $\kappa d$ (the product of thickness and modulation) that can be obtained in dichromated gelatin whereas the increase in the signal to noise ratio results from the absence of the high level of primary scatter found in processed silver halide emulsions.

6.8 DISPLAY HOLOGRAM COPYING.

When a display hologram is recorded it is important that the subject is uniformly illuminated from several directions to eliminate shadows and to ensure uniform brightness of the image at replay. To achieve this, much of the available light is wasted and the amount of light which finally reaches the recording plate is small. Recording a dichromated gelatin display hologram directly may therefore be difficult due to its low sensitivity. However, dichromated gelatin display
holograms can be obtained easily by contact copying holograms recorded in more sensitive materials.

6.8.1 Copying procedure.

Several dichromated gelatin copies were made of silver halide display holograms by the contact method. These were reflection holograms which can be replayed in white light due to their high wavelength selectivity.

Many of the silver halide master holograms had undergone shrinkage during processing. To replay the holograms at 514.5 nm it was therefore necessary to use a higher angle of reference beam than was used in the original recording. However, this change did not introduce any perceptible distortion in the copy holograms.

There were some problems due to small flaws in some of the original holograms. Small dust particles which become embedded in the gelatin surface during processing cause small 'bumps' to form when the original holograms dry. These bumps change the fringe slant or spacing (and therefore the Bragg angle) in a small area around them. In monochromatic illumination, dark spots can be seen at the position of these bumps due to the change in Bragg angle. However, these spots are not visible under white light illumination because, in this case, the change in Bragg angle causes a small change in replay colour, but no change in brightness. When the hologram is copied using monochromatic light, the black spots are recorded permanently into the copy hologram as a reduction in modulation rather than a small change in fringe slant. As a result the black spots are visible in the copy holograms in both white light or monochromatic illumination.

6.8.2 Comparison of master and copy.

If a master hologram without these surface defects was used, very good copies could be made. The copy holograms appeared brighter than the
original holograms and there was a significant improvement in contrast due to a reduction in scatter. The colour of the scattered light in the copies varied if the replay angle or viewing angle was changed in a similar manner to that seen in the planar copy holograms and synthesised noise gratings. Intermodulation noise, visible as a 'halo' around bright areas of the image in the original holograms, was also seen in the copies but did not appear to be increased. The use of high exposures and high processing temperatures led to a visible decrease in the signal to noise ratio in the copies, presumably due to the effects of non-linearity and saturation discussed in 6.7.

An example of master and copy can be seen in Plate 6.3. The master hologram was recorded in 5 μm AGFA 8E56 HD emulsion and the copy recorded in 20 μm gelatin, sensitized in 2% ammonium dichromate. If a white light source of small angular diameter is used to replay the holograms (to minimize blurring due to finite source size) the image of the copy hologram appeared to be sharper than that of the master because the thicker gelatin results in higher wavelength selectivity and therefore less blurring due to dispersion. Despite this higher wavelength selectivity, the copy appeared brighter than the original when replayed with white light.

6.8.3 Wavelength response of master and copy.

To investigate these observations, the transmission of these two holograms as a function of wavelength was measured at the normal replay angle of 30° and is shown in Fig. 6.30. The brightness of the image depends on the total amount of light diffracted from the incident spectrum (the area of the Bragg dip) whilst the sharpness of the image is related to the bandwidth of the hologram (the width of the Bragg dip) \[\text{WAR85}\]. It can be seen that the response of the copy is narrower than that of the master hologram but that the total amount of diffracted light
PLATE 6.3

SILVER HALIDE MASTER HOLOGRAM

DICHROMATED GELATIN COPY
Fig. 6.30 A comparison of the wavelength response of a 5 μm silver halide master display hologram and a 20 μm dichromated gelatin copy.
is slightly greater. The shape of the wavelength response of the copy is more suitable for display holograms and explains the increase in sharpness and brightness.

6.8.4 Colour control.

The maxima of the light diffracted by the copy occurs at 563 nm rather than the recording wavelength, 514.5 nm, due to swelling which occurred during processing. The image in the copy is therefore yellow. Green and blue copies of the green master were produced by using gelatin sensitized with 5% and 10% ammonium dichromate respectively. The dependence of replay colour on the concentration of the sensitizer is a result of the different thickness and refractive index changes which occur during processing, which were measured in Chapter 3. No perceptible distortion could be seen as a result of these thickness and refractive index changes. It may be possible to produce red display holograms in this way by increasing the degree of swelling using the method suggested by Coleman and Magarinos [COL81].

A two colour copy was produced by recording separate green and blue exposures using a two colour silver halide master hologram. However blue and green are not the ideal combination of colours for a two colour display hologram: Green and red would be preferable.

6.9 SUMMARY.

The motives for copying holograms have been discussed and dichromated gelatin has been found to be have ideal properties for use as a copying medium.

The properties of a dichromated gelatin copy of a silver halide planar reflection grating have been examined. It has been shown that the copy is more efficient than the original and also that the copy produces less noise or scatter under white light illumination. It has
been found that noise is recorded in addition to the desired wavefront and that, at replay, the copy displays some of the behaviour associated with noise gratings recorded in silver halide emulsions.

It has been shown that a single beam exposure in dichromated gelatin does not normally result in noise gratings because the level of scatter at the recording stage is very low. This is in contrast to the behaviour of single beam exposures in silver halide.

'Synthesised noise gratings' have been recorded in dichromated gelatin which are recordings of the scattered light field from a silver halide emulsion. Transmission type synthesised noise gratings have been found to have very similar properties to silver halide noise gratings but reflection type synthesised noise gratings display only weak diffraction. This implies that the strong diffraction seen in silver halide noise gratings is predominantly due to transmission gratings. An interesting feature of the synthesised noise gratings is that they do not display any of the behaviour of overmodulated transmission or reflection planar gratings at high exposures.

Noise rings or cones have been seen in the synthesised noise gratings and in the planar grating copy. A theoretical model has been developed which includes the effects of thickness changes and average refractive index changes and which successfully predicts the apex angle of the cones of scattered light.

Dichromated gelatin copies of silver halide diffuse object and display holograms have been fabricated. It has been found that brighter and sharper images can be obtained and lower levels of noise. The relationship between noise, brightness and exposure has been explored and it has been found that the image colour can be controlled easily by varying the concentration of the ammonium dichromate sensitising solution.
In this short chapter a method for modelling holographic optical elements is presented. Initially a ray tracing approach is developed which includes the effects of planar glass substrates. This is then extended to model the angular and wavelength selectivity of the hologram using a vectorial form of coupled wave theory which is valid for any incidence angle and polarisation.

7.1 Introduction.

The behaviour of holographic optical elements has been analysed using a ray tracing approach by many authors [OFF66, ABR69, LAT71B, HOL81]. When designing systems which use holograms for imaging, such as Head-up displays, the important information required is the spotsize of an image point and the distortion of the image across the field of view. Ray tracing methods are well suited to determining these quantities. Alternative methods, such as wavefront matching, may have advantages in some circumstances [LAT71A] but can involve lengthy algebra.

It is difficult to know the present state of the art in ray tracing methods because the (reputedly) more sophisticated implementations are available only as a commercial package at a price beyond the resources of this research group.
Ray tracing methods are however, unaware of the angular and wavelength selectivity of thick volume gratings and cannot yield any information about the amplitude of the diffracted light. This selectivity will limit the field of view when imaging extended objects [SYM83] and will reduce the overall efficiency when the replay wavelength is changed [ST085]. A suitable method for modelling this selectivity would be to combine the ray tracing approach with a suitable form of coupled wave theory.

A theoretical model using this approach has already been developed within the group [FER80, OWE82]. The purpose of this Chapter is to extend this model to include the effects of the glass substrate and coverplate and to include the vectorial nature and polarisation of the replay wave.

This work is part of a continuing development program of theoretical models for holographic optical elements within the Oxford Holography group, the eventual aim of which is to model arbitrary systems containing multiple holographic optical elements and conventional elements.

7.2 3-DIMENSIONAL RAY TRACING THROUGH A HOLOGRAPHIC LENS.

In the references given for previous work on ray tracing the effects of hologram substrates are usually ignored. For dichromated gelatin holograms both a substrate and a coverplate are required and the total thickness of the hologram is typically 5 mm. For this reason it was felt that it was important to include their effects.

7.2.1 Hologram recording.

The first stage of the ray tracing procedure is to calculate the recording wavevectors at each position on a grid of equally spaced points
across the aperture of the holographic lens, which may be elliptical or rectangular.

All holographic lenses recorded with spherical wavefronts can be described in terms of the position of two points from, or to which the two recording waves are diverging or converging. For ease of comparison with experimental recording configurations these point sources are initially specified in spherical polar coordinates but are immediately converted into cartesian coordinates centred on the hologram emulsion (Fig. 7.1).

Fig. 7.2 shows the geometry of one of the recording beams. For a specified position on the hologram, \((0, y, z)\), and a specified source position, \((x_b, y_b, z_b)\), it is necessary to find the position, \((x_d, y_d, z_d)\), where the ray meets the surface of the substrate. This was initially done by a Fermat minimalisation method, similar to that used recently by Verboven and Lagasse [VER85] in a ray tracing analysis of holographic mirrors. However the following method was found to be more useful since it is applicable to converging beams without modification, simply by specifying a negative value of \(x_b\).

From Fig. 7.2 the following relationships can be written directly

\[
\begin{align*}
 r &= \left[ \left( y - y_b \right)^2 + \left( z - z_b \right)^2 \right]^{1/2} \quad (7.1) \\
\frac{y_d - y_b}{y - y_b} &= \frac{z_d - z_b}{z - z_b} = \frac{r_d}{r} \quad (7.2) \\
\tan \theta &= \frac{r_d}{(x_b - x_d)} \quad (7.3) \\
\tan \alpha &= \frac{r - r_d}{x_d} \quad (7.4)
\end{align*}
\]
Fig. 7.1 The coordinate system used to describe hologram recording. The two spherical wavefronts are defined by the positions of two point sources S1 and S2.

Fig. 7.2 The geometry of one of the recording rays (in the plane of incidence) showing refraction at the surface of the glass substrate.
Using Snell's law

\[ n_1 \sin \alpha = \sin \theta \]  

(7.5)

where \( n_1 \) is the refractive index of the glass substrate. From (7.3), (7.4) and the relationship

\[ \sin \alpha = \frac{\tan \alpha}{\sqrt{1 + \tan^2 \alpha}} \]  

(7.6)

a quartic is obtained:

\[ a_4 r_d^4 + a_3 r_d^3 + a_2 r_d^2 + a_1 r_d + a_0 = 0 \]  

(7.7)

where

\[ a_4 = n_1^2 - 1 \]  

(7.8)

\[ a_3 = -2r(n_1^2 - 1) \]  

(7.9)

\[ a_2 = n_1(x_b - x_d)^2 + n_1 r - x_d^2 - r^2 \]  

(7.10)

\[ a_1 = -2r n_1(x_b - x_d)^2 \]  

(7.11)

\[ a_0 = n_1(x_b - x_d)^2 r^2 \]  

(7.12)

This can be solved for \( r_d \) numerically by Newton's method using the initial approximation

\[ r_d = \frac{r}{x_b}(x_b - x_d) \]  

(7.13)

and hence \( x_d \) and \( y_d \) can be found from (7.2).

From \( (x_d, y_d, z_d) \) and \( (0, y, z) \) the wavevectors of the two waves in the substrate can be found and hence, using Snell's law, the wave vectors \( \rho \) and \( \sigma \) in the emulsion. If there was no intention to develop the model beyond the ray tracing stage it would now be sufficient to use the vector formulation described by Welford [WEL75]. However, because of the way
the model will be extended in Sec. 7.3, the grating vector $\mathbf{K}$ at $(0, y, z)$ is now determined

$$\mathbf{K} = \mathbf{p} - \mathbf{a}$$  \hfill (7.14)

7.2.2 Hologram replay.

At replay the hologram is replayed by a spherical wavefront specified by the source position $R_1$ and the aim is to find the position $I$, where diffracted rays from each point on the hologram cross the image plane (Fig. 7.3).

Using the method described in the previous section the wavevector, $\mathbf{p}_0$, of the replay beam in the emulsion can be found. For the purposes of ray tracing the thickness of the emulsion is assumed to be negligible. Using the beta value construction shown in Fig. 1.2b the wavevectors of the diffracted beams $\mathbf{p}_n$ are then determined from the relationships

$$\mathbf{p}_{ny} = \mathbf{p}_{0y} - n\mathbf{K}_y$$
$$\mathbf{p}_{nz} = \mathbf{p}_{0z} - n\mathbf{K}_z$$
$$|\mathbf{p}_n| = \beta$$ \hfill (7.15)

where $\mathbf{K}_y$ and $\mathbf{K}_z$ are the $y$ and $z$ components of the grating vector, $n$ is the order of diffraction and $\beta$ is the propagation constant in the emulsion. If the perpendicular line drawn from end of the $\mathbf{K}$ vector does not intersect with the circle the diffracted wave will be evanescent and will not propagate.

It can be seen from this diagram that the directions of the diffracted rays are independent of the component of the $\mathbf{K}$ vector and the replay wavevector in the direction of the surface normal. This leads to the result that the directions of the diffracted wavevectors are unaffected by thickness changes and are independent of the initial and final refractive index of the hologram. The Bragg condition will be
Fig. 7.3 The geometry of hologram replay. The replay reference beam is an arbitrary spherical wave defined by the position of the source R1.

Fig. 7.4 Recording configuration for the double focus lenses.
affected by such changes but this will not be apparent in the ray tracing model.

It is then a simple matter to propagate the rays to the outer surface of the second substrate (if present) on the other side of the hologram and then to the intersection with the image plane at $I$. Some rays, diffracted at high angles, may be unable to propagate out of the substrate due to total internal reflection.

7.3 AN INVESTIGATION OF SPURIOUS FOCI IN A DOUBLE EXPOSURE LENS USING RAY TRACING.

Holographic optical elements containing multiple gratings have several useful applications. Holographic beam splitters and combiners [CAS75] and multiple imaging lenses [LIA83] have been demonstrated using multiple exposure holograms recorded in dichromated gelatin.

Holograms containing several planar grating have been analysed using coupled wave theory [CAS75, SLI85] and the thin grating decomposition approach [ALF75]. The existence of spurious diffraction orders has been observed experimentally, which have been ascribed to multiple grating interactions [SLI85]. It is not obvious how these spurious waves might affect holographic optical elements containing several gratings. If they form a focus, or an approximate focus, their effects may become apparent, even if the diffraction efficiency into the spurious waves is low.

The aim of this section is to demonstrate that spurious foci can be formed by multiple diffraction and that ray tracing can be used to identify the grating interactions responsible.

7.3.1 Recording and replay of a double focus off-axis holographic lens.

Double focus holographic lenses were recorded in $7 \mu m$ dichromated gelatin by making two sequential exposures (Fig. 7.4): The first
exposure with beams A and B and the second exposure with beams A and C. The lenses were not recorded by simultaneous exposure to three beams as this would result in the recording of three gratings, one associated with each pair of recording waves. One hologram was recorded with $\theta = 30^\circ$ and another with $\theta = 10^\circ$. The hologram recorded with the large interbeam angle will be optically thick whereas the hologram recorded with the small interbeam angle will be optically thin and higher diffraction orders may be present.

The lenses were then replayed as shown in Fig. 7.5. Ideally, all the incident light would be diffracted into the two principal foci but in the optically thin case, higher diffraction orders will occur and for both cases there may be some power remaining in the transmitted replay beam. However, several other diffracted beams were clearly visible in both holograms which could not be explained in terms of higher order diffraction from a single grating. To enable these beams to be seen a screen was placed at the two principal foci and photographs were taken which can be seen in Plate 7.1.

In Plate 7.1a. from the optically thick hologram, the two principle foci and the undiffracted transmitted beam can each be seen to be associated with a number of much weaker, collinear beams. There is no evidence of any light diffracted at higher angles. In Plate 7.1b from the optically thin hologram, it can be seen that there are many diffracted beams of significant power and that these are also ordered in concentric groups. The spurious beams are either diverging waves with a virtual source on the opposite side of the hologram or waves which come to a focus in between the hologram and the screen. It was decided to modify the ray tracing analysis to investigate whether multiple diffraction was responsible.
Fig. 7.5 Replay configuration for the double focus lenses used for Plate 7.1.

Fig. 7.6 The geometry of the extended model. The four rays at the corners of each small square element are traced to the image plane and the diffraction efficiency at the centre of the element is calculated.
PLATE 7.1

TWO FOCUS LENS $\Theta = 30^\circ$

TWO FOCUS LENS $\Theta = 10^\circ$
7.4.3 Ray tracing of higher order and multiple grating diffraction.

The propagation constant of a ray \( \hat{\rho}_{n,m} \) following multiple diffraction can be found from the beta value construction, which was used earlier, which can be expressed as

\[
\hat{\rho}_{x_{n,m}} = \hat{\rho}_0 - n\hat{K}_1 - m\hat{K}_2 \quad (7.16)
\]

\[
\hat{\rho}_{y_{n,m}} = \hat{\rho}_0 - n\hat{K}_1 - m\hat{K}_2 \quad (7.17)
\]

\[
|\hat{\rho}_{n,m}| = \beta \quad (7.18)
\]

where \( \hat{\rho}_0 \) is the replay wavevector, \( \hat{K}_1 \) and \( \hat{K}_2 \) are the two grating vectors, \( n \) and \( m \) are the orders of diffraction from each of the two gratings, and \( \beta \) is the propagation constant in the emulsion. These equations were incorporated into the ray tracing method so that it was possible to determine the effects of all the possible multiple diffraction orders.

It should be noted that there may be harmonic and intermodulation harmonic gratings present in the hologram, due to the non-linearity of the recording characteristic, which can be expressed in terms of the two fundamental gratings as

\[
\hat{K}_{p,q} = p\hat{K}_1 + q\hat{K}_2 \quad (7.19)
\]

However, these harmonic gratings will lead to diffracted rays in exactly the same direction as those due to multiple diffraction for \( p = n \) and \( q = m \) and it is not possible to distinguish their effects.

At the centre of each lens the symmetry of the recording configuration means that \( K_{1x} = -K_{2x} \) and \( K_{1y} = K_{2y} = 0 \). From (7.16) - (7.18) it follows that the all \( \rho_{n,m} \) where the difference \( n - m \) is a constant will have the same direction. This explains why the concentric grouping of the diffraction orders is seen.

Using the extended ray tracing program, the form of the diffracted beams for all possible values of \( n \) and \( m \) between \( \pm 3 \) was determined and
are summarised in Table 7.1 and 7.2. For the 30° grating it is only possible to replay orders where \((n-m) \leq 1\) but for the 10° grating it was possible to replay orders where \((n-m) \leq 3\). All other orders are evanescent in the hologram or cannot leave the substrate due to total internal reflection. The angle quoted is the angle of the primary or central ray of each beam, which is common to each grouping. If the focal length specified is negative, the beam is diverging from an approximate virtual focus on the opposite side of the hologram. If a positive focal length is given, the beam comes to an approximate focus at that distance from the hologram. The quality of the focus for each beam is specified by the quoted spotsize which is the diameter of the circle of least confusion. Apart from the two primary foci, most of the foci are highly astigmatic and come to a spatially separate vertical and horizontal line foci either side of the quoted focal length.

It is also possible to use the ray tracing program to find the size of the halo's seen on the screen in the two plates. Using this technique it was found that the large outer elliptical halo around the central spot in Plate 7.1a is due to the \(-3K_1 - 3K_2\) order. Inside this is a small ellipse which corresponds to the \(3K_1 + 3K_2\) order. There were two smaller halo's due to the \(K_1 + K_2\) and \(-K_1 - K_2\) orders present but these are not clearly discernable in the plates due to the brightness of the undiffracted 0th order.

Around the left hand principal focus there is a large halo due to the \(3K_1 - 2K_2\) order which is too weak to be seen clearly in the plate. The small bright circular halo corresponds to the \(3K_1 + 2K_2\) and \(-2K_1 - K_2\) orders. A smaller bright halo due to the \(2K_1 + K_2\) order was present but again is not discernable due to the brightness of the principal focus.
TABLE 7.1

SUMMARY OF THE DIFFRACTION ORDERS OF THE 30° DOUBLE FOCUS LENS.

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<th>DIFFRACTION ORDER</th>
<th>n</th>
<th>m</th>
<th>BEAM ANGLE (Deg)</th>
<th>FOCAL LENGTH (cm)</th>
<th>MINIMUM SPOTSIZE (mm)</th>
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<td>FOCAL LENGTH (CM)</td>
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It is interesting to note that no spurious foci with a focal length greater than the principal foci were predicted by the model and none could be found experimentally.

7.4 EXTENSION OF THE RAY TRACING MODEL.

The logical extension to the simple ray tracing approach is to find the power in the individual rays reaching the image plane, in addition to their positions of intersection. To do this the aperture of the hologram can be considered as being composed of a large number of small elements in which the local \( K \) vector and modulation are taken as being constant.

Using the ray tracing method, the four rays which pass through the corners of each elemental area can be found and their eventual position on the image plane of the rays can be determined (Fig. 7.6) for each diffracted order. The intensity at the image plane of light from each elemental area can then be found by calculating the power within the 'tube' bounded by the four rays at each stage as they propagate through the hologram and onto the image plane. This will require the area of the elements at the hologram and on the image plane to be found and the Fresnel losses at each surface boundary to be calculated. Finally, it is necessary to determine what fraction of the power will be diffracted into each significant diffraction order. Because the thickness of the hologram is small compared to the focal length of the lenses to be considered, the \( K \) vector will vary slowly across the hologram and the localised use of uniform grating coupled wave theory is permissible.

7.4.1 Vector coupled wave theory.

It is not possible to apply the coupled wave theory derived in Chapter 1 to this problem because of the restrictions which were imposed on the geometry of the replay configuration. It was assumed that the fringe planes and the polarisation of the incident light were
perpendicular to the plane of incidence. For work on planar gratings it is possible to manipulate the replay configuration to ensure that these replay conditions are satisfied but in general it will not be possible to do this for a holographic lens. It is therefore necessary to use a coupled wave theory valid for arbitrary incidence angle and polarisation which has recently been described by Syms [SYM85].

It is possible to simplify his derivation somewhat since a non-symmetric grating profile would not be expected and because the absorption modulation in dichromated gelatin is negligible. The dielectric constant within the grating can therefore be written as

$$\epsilon(\hat{r}) = \epsilon'_r - j\epsilon''_r + \sum_{i=1}^{\infty} \epsilon'_r \cos(iK \cdot \hat{r})$$

(7.20)

where $\epsilon'_r$ is the average dielectric constant, $\epsilon''_r$ the absorption constant, and $\epsilon'_r$ are the amplitudes of $i$th harmonic of the grating profile. $\hat{K}$ is the grating vector and $\hat{r}$ the radius vector. The time-independent vector wave equation inside the grating is therefore

$$\nabla \times (\nabla \times \mathbf{E}) - \beta^2 \left[ 1 - \frac{j\epsilon''_r}{\epsilon'_r} + \sum_{i=1}^{\infty} \frac{\epsilon'_r}{\epsilon'_r} \cos(iK \cdot \hat{r}) \right] \mathbf{E} = 0$$

(7.21)

An infinite plane wave incident on the grating from a surrounding medium of the same average refractive index can be expressed as

$$\mathbf{E} = E_0 \hat{p} \exp(-j\hat{\rho}_0 \cdot \hat{r})$$

(7.22)

where $\hat{\rho}_0$ is the propagation vector and $\hat{p}$ the polarisation vector such that

$$|\hat{\rho}_0| = \beta, \quad |\hat{p}| = 1, \quad \hat{p} \cdot \hat{\rho}_0 = 0$$

(7.23)

Suitable solutions of equation (7.21) which may have any of three possible orthogonal polarisations are
\[ E = E_0 \sum_{n=-\infty}^{\infty} \sum_{m=1}^{3} A_{n,m} \hat{p}_{n,m} \exp(-j\rho_n \cdot r) \] (7.24)

where
\[ \hat{p}_{n,1} \cdot \rho_n = 0, \quad \hat{p}_{n,2} = \rho_0 / |\rho_0|, \quad \hat{p}_{n,3} = \hat{p}_{n,1} \times \hat{p}_{n,2} \] (7.25)

and where the diffracted waves are chosen to be defined by \( K \)-vector closure and are therefore \( \hat{\rho}_n = \hat{\rho}_0 - n\hat{K} \). The derivation now follows the standard coupled wave procedure followed in Chapter 2 substituting (7.24) into (7.21) and equating the coefficients of the exponentials to zero and neglecting second derivatives as before. In the limit \( \beta \to \infty \) the amplitudes \( A_{n,2} \) are all zero and therefore the diffracted orders are transverse waves. The vector equations which result can be simplified to a set of scalar equations by taking the scalar product with \( \hat{p}_{n,1} \) and with \( \hat{p}_{n,3} \). If it is chosen that \( \hat{p}_{n,1} = \hat{p}_0,1 \) the orthogonal polarisations are not coupled together and two independent sets of coupled wave equations are obtained for each polarisation

\[ c_n \frac{dA_{n,1}}{dx} + (\alpha + j\nu_n)A_{n,1} + \sum_{i=1}^{\infty} j\kappa'_i \left[ A_{n+i,1} + A_{n-i,1} \right] = 0 \] (7.26)

and

\[ c_n \frac{dA_{n,3}}{dx} + (\alpha + j\nu_n)A_{n,3} + \sum_{i=1}^{\infty} j\kappa'_i \left[ (\hat{p}_{n+i,3} \cdot \hat{p}_{n,3})A_{n+i,3} + (\hat{p}_{n-i,3} \cdot \hat{p}_{n,3})A_{n-i,3} \right] = 0 \] (7.27)

where
\[ c_n = \rho_{nx} / \beta, \quad \nu_n = \frac{\beta^2 - \rho_n^2}{2\beta}, \quad \alpha = \frac{2\epsilon_r' \epsilon_r}{r_0^2}, \quad \kappa'_i = \frac{\beta \epsilon_{ri}}{4\epsilon_r' r_0} \] (7.28)

To find the diffracted rays in the surrounding medium it is necessary to match the tangential components of the wavevectors and the polarisation vectors at the boundary of hologram.
It can be seen that for polarisation parallel to the plane containing the incident and diffracted rays (and hence also $\mathbf{k}$) the coupling is reduced by a factor equal to the cosine of the angle between the incident and diffracted waves. When the waves are perpendicular to each other the coupling is reduced to zero and diffraction does not occur. This is analogous to the absence of reflection at Brewster's angle at the boundary between two materials with different dielectric constants.

In the implementation of this vector coupled wave theory the higher harmonics of the grating profile were ignored and only five diffraction orders were considered $-2, 0, +2$ to reduce the computation requirements.

7.4.2 Implementation.

The first step in the model is to specify the input polarisation of the replay wave in the medium surrounding the hologram. The components of the replay wave with polarisation perpendicular and parallel to the plane of incidence must then be found so that the Fresnel coefficients at the input face of the hologram can be determined. From these coefficients the amplitude and polarisation vector inside the hologram can be found.

It is then necessary to find the components of the replay wave with polarisation perpendicular and parallel to the plane containing the replay wave and $\mathbf{K}$ vector so that the boundary values of $A_{n,1}$ and $A_{n,2}$ can be determined. The two sets of coupled wave equations are then solved numerically using a Runge-Kutta method to find the amplitudes of the diffracted waves at the exit boundary. Because the coupling for the two components is different, the polarisation vectors of the diffracted waves will change with depth in the hologram.
After determining the polarisation vector at the exit of the hologram it is necessary to find the components of the diffracted waves with polarisation perpendicular and parallel to the plane of incidence at the exit boundary, so that the Fresnel coefficients can be used to find the final amplitudes and polarisation vectors of the diffracted waves in the external medium.

The model provides the crucial information about the efficiency of the Bragg diffraction process (areas of the lens which are significantly off-Bragg will contribute little to the output beam). It is then a simple matter to calculate the overall efficiency of the lens by summing the total power diffracted into the desired order over the complete aperture of the lens. Using this approach the model will be used to optimise the design of a lens to suit a particular application in the Chapter 8.

The model may also be used to examine aberrations but will not yield the exact intensity profile near the foci since it is not valid when the tubes begin to overlap. A two step process is however possible. The model may be used to determine the electric field distribution at the exit boundary of the hologram which may then be substituted into a Kirchhoff type diffraction integral to yield the intensity at any desired point.

7.5 POLARISATION EFFECTS IN OFF-AXIS AND ON-AXIS LENSES.

The model can be applied to any replay configuration of any transmission holographic optical element recorded with spherical or planar wavefronts. It is applied here to demonstrate the effects of the polarisation of the replay wave on the efficiency of an off-axis and an on-axis holographic lens, since polarisation is specifically included in the model.
The recording configuration for the two lenses which will be considered theoretically is shown in Fig. 7.7. The thickness of the both holograms was set at 10 μm. Note that the on-axis lens is identical to the lens which will be used in Chapter 9. When the two lenses are replayed by a point source at the position of the recording point source both lenses will collimate the light into a parallel beam (Fig. 7.8). The model was used to find the efficiency variation ($P_{\text{out}} / P_{\text{in}}$) across the aperture of each lens for replay by horizontally polarised (in the plane of the diagram) and vertically polarised light.

It was assumed that the modulation was uniform across the aperture of the hologram although in general the uniformity would depend on the amplitude variation across the recording beams. The coupling constant, $\kappa$, was $1.55 \times 10^5 \text{ (m}^{-1}\text{)}$ for the on-axis lens and $1.43 \times 10^5 \text{ (m}^{-1}\text{)}$ for the off-axis lens which were the highest values which could be used without any areas of either hologram being overmodulated. The variation in the amplitude of the replay beam is not included for clarity and it is the efficiency of the lens that is calculated.

7.5.1 The off-axis lens.

The variation of efficiency across the off-axis lens for replay with horizontal and vertical polarised light is shown in the form of a contourplot in Fig. 7.9 and Fig. 7.10 respectively. The replay point source lies to the left of the aperture. Fig. 7.11 (a) and (b) are plots of the efficiency along the central horizontal and vertical axes respectively. The horizontal axis lies in the plane of the diagram in Fig. 7.8.

Along the central horizontal axis the $K$ vector has no vertical component. The plane containing the replay wavevector and the $K$ vector is therefore in the horizontal plane. For replay with light polarised in the horizontal plane the coupling is therefore reduced and the efficiency
Fig. 7.7 Recording configuration of the two lenses selected for analysis using the extended theoretical model, (a) the off-axis lens, (b) the on-axis lens.

Fig. 7.8 Replay configuration of the two lenses selected for analysis using the extended theoretical model, (a) the off-axis lens, (b) the on-axis lens.
Fig. 7.9 Contourplot of efficiency across the aperture of the off-axis lens for light polarised along the horizontal (y) axis.

Fig. 7.10 Contourplot of efficiency across the aperture of the off-axis lens for light polarised along the vertical (z) axis.
Fig. 7.11 Efficiency of the off-axis lens (a) along the horizontal y axis and (b) along the vertical z axis for —— horizontally polarised and —— vertically polarised light.
A MODEL FOR HOLOGRAPHIC OPTICAL ELEMENTS.

Fig. 7.12 Contourplot of efficiency across the aperture of the on-axis lens for vertically polarised light.

Fig. 7.13 Efficiency against radius for the on-axis lens for horizontally polarised light (parallel to radius) and vertically polarised light (perpendicular to radius).
is considerably lower than that for vertical polarisation, particularly in the areas of the lens where the angle of diffraction is high.

The effects of this decrease in diffraction efficiency is reduced slightly by the fact that the Fresnel losses are lower for the horizontal polarisation.

7.5.2 The on-axis lens

The efficiency of the on-axis lens when replayed with vertically polarised light is shown in the form of a contourplot in Fig. 7.12 and along the horizontal and vertical axis in Fig. 7.13. It is not necessary to calculate the efficiency for horizontal polarisation since the hologram is radially symmetric and this corresponds to rotating Fig. 7.12 through 90°.

The difference in the efficiency for the two polarisations is minimal since the largest interbeam angle is only 13.98° and the coupling is reduced by a maximum of \( \cos(13.98°) = 0.97 \) at the edges of the lens. This corresponds to a loss of diffraction efficiency of less than 1% and the efficiency at the edges is actually higher for horizontal polarisation due to the lower Fresnel losses at the input boundary of the hologram.

7.6 SUMMARY.

A ray tracing analysis of holographic optical elements has been developed which includes the effects of planar substrates. This has been extended by combining the ray tracing approach with vector coupled wave theory. The extended model is capable of modelling the wavelength and angular selectivity of volume holographic elements.

The model has many applications and can be used to determine the field of view of an imaging system using holographic lenses or can be
used to investigate the effects of changing the replay wavelength, hologram thickness and refractive index.

The model has been used to compare the effect of the polarisation of the replay wave in an off-axis and an on-axis lens and it has been seen that in general the off-axis lens is more sensitive to the replay polarisation. Larger polarisation effects would only be seen for high angles of diffraction which would not normally be encountered in holographic lenses since this would correspond to very high angles outside the hologram and total internal reflection would occur unless replay was in an index matching tank.
CHAPTER 8.

OPTICAL MEASUREMENT OF POSITION USING AN ON-AXIS HOLOGRAPHIC LENS.

A position sensor based on the dispersive properties of an on-axis holographic lens is described. The optimisation of the parameters of the lens required for the application is discussed and examples are fabricated. The performance of the sensor is investigated and applications are suggested. The position sensor is also shown to be capable of measuring the orientation of planar surfaces and of operating remotely, using an optical fibre.

8.1 INTRODUCTION.

The use of optical, non-contacting techniques for the measurement of distance, rotation and velocity have many advantages which have resulted in their growing use in many applications. In general, optical methods have no moving parts and therefore do not exert contact pressure on the test object or suffer from the hysteresis or backlash associated with mechanical systems of measurement. The lack of moving parts also increases their reliability and, because they have no inertia, they can respond rapidly. Optical methods are also useful in hostile environments because measurements can be made remotely either through an optical window or by using optical fibres.

One simple optical technique for measuring distance is to project a spot of light onto a surface and then image this spot onto a detector,
such as a linear photodiode array. The distance of the surface can then be found by triangulation [DAV84]. Another simple technique, with many applications, is to examine the retro-reflected light from the focus of an astigmatic lens [COH84]. If very high accuracy is required there are many interferometric methods which can be used [BEN83].

Recently, the development of more sophisticated optical techniques, which can measure position, shape and orientation, has been stimulated by the requirements of robotics and by the rapid growth in microcomputers. These techniques typically project a dot array [DUF84] or a grid of lines [SLI85] onto a surface which is viewed by a video camera connected to a microcomputer. After a considerable amount of image processing and computation the topography or orientation of the surface can be determined from the position of particular features of the projected pattern.

Several optical systems which utilise the dispersive properties of diffraction gratings for the measurement of distance or angular rotation have been described by Hutley [HUT84]. The distance measurement systems rely on the fact that the focal length of a Fresnel zone plate is, to a first approximation, inversely proportional to wavelength. These zone plates, which are surface relief gratings, have similar optical properties to an on-axis volume holographic lens.

The most versatile configuration for measuring distance by this method is shown in Fig. 8.1. White light passing through pinhole A is collimated with a conventional lens and then focused onto the test surface by the zone plate. Because the focal length varies rapidly with wavelength only one colour is brought to a sharp focus on the test surface. Light of this colour which is specularly or diffusely reflected from the test surface forms an image of the light source at pinhole B. If light passing through pinhole B is analysed using a spectrometer it is
Fig. 8.1 Optical measurement of distance using a zone plate or an on-axis holographic lens.

Fig. 8.2 Theoretical power into the $-1^{\text{th}}$ diffracted order against radius for on-axis holographic lenses of different thickness, recorded and replayed at 514.5 nm (Polarisation is normal to radius).
Fig. 8.3 Theoretical power into the $-1^{\text{th}}$ diffracted order against radius for on-axis holographic lenses of different thickness, recorded at 514.5 nm and replayed at 632.8 nm (Polarisation is normal to radius).

Fig. 8.4 Theoretical minimum spotsize at the focus of the on-axis lens against replay wavelength.
found that there is a bright peak in the spectrum at one wavelength, corresponding to the colour of light which comes to a focus on the test surface. By measuring this wavelength it is therefore possible to deduce the distance of the surface from the zone plate. The advantages of this system are that the peak wavelength is unaffected by the slant or reflectivity of the test surface or by fluctuations in the brightness of the light source.

One problem encountered with this system is keeping the overall losses to an acceptable level. At present, the maximum efficiency of suitable zone plates is about 30% into the first diffracted order. After two passes through the zone plate and the 50% beamsplitter the overall efficiency of the system therefore only 2.5%. If the test surface is a diffusely reflecting surface, rather than a mirror, the efficiency is also limited by the numerical aperture of the zone plate. However, it is difficult to fabricate efficient zone plates of a high numerical aperture.

In contrast to these surface relief gratings it is possible to record high numerical aperture volume holographic optical elements in dichromated gelatin, due to its high spatial frequency response. A volume grating could be recorded which will have identical dispersive properties to a Fresnel zone plate. If it can be made optically thick it could reach almost 100% efficiency into the first diffracted order. The purpose of this chapter is to determine whether the properties of volume holographic optical elements make them suitable for this particular application and to investigate the performance of a completed system.
8.2 THEORETICAL PERFORMANCE OF AN ON-AXIS LENS.

8.2.1 Theoretical efficiency.

The recording configuration for a suitable on-axis holographic lens is shown in Fig. 7.7b. It can be seen that in the central region of the lens the angle between the recording beams will be low. This will result in an optically thin grating in these areas and at replay there will be significant diffraction into higher diffraction orders. The existence of these higher orders will reduce the power in the desired first diffracted order and will result in additional foci. The focal length of the 2nd order focus at a wavelength of $\lambda$ will be the same as the focal length of the 1st order focus at a wavelength of $2\lambda$. It is therefore necessary to restrict the range of input wavelengths into the distance sensor to slightly less than one octave, to avoid the ambiguity which would be introduced by the overlapping of the spectral orders. The spectral output range of most practical light sources for the distance sensor is usually within this limit and the spurious foci do not cause any problems. However they do result in a loss of efficiency.

At larger distances from the centre of the lens the grating will be optically thick and the power in the higher orders will decline. This means that as the numerical aperture of the lens is increased, the efficiency of the lens will increase. However, the grating will also become more selective and the efficiency at wavelengths other than the recording wavelength will decline. Because it is necessary for the lens to operate with white light at fixed, normal incidence, this selectivity may become a problem.

8.2.2 Selection of hologram thickness.

The problems caused by higher orders and by the wavelength selectivity will be affected by the thickness of the hologram. If thick gelatin is used, the effect of higher orders will be reduced and the
average efficiency across the aperture of the lens will increase. However, the wavelength range over which the lens will have a useful efficiency will be reduced due to increased wavelength selectivity. If thin gelatin is used the hologram will replay over a wider range of wavelength, but only the edges of the lens will be efficient due to the power in higher orders at lower radii. It is therefore important to choose a thickness such that there is a compromise between these two problems for the particular numerical aperture that is required.

Due to the size of the optical components available to record the lenses, an aperture of 70 mm and a focal length of 9 cm was chosen. This corresponds to a numerical aperture of about 0.4, but higher numerical apertures could be fabricated if required.

Using the model developed in Chapter 7, the expected variation in power of the first diffracted order across the width of the aperture was calculated for 5, 10 and 20 μm thick holograms and is shown in Fig. 8.2 and Fig. 8.3, for replay at 514.5 nm (the recording wavelength) and 632.8 nm respectively. These wavelengths lie either side of the peak output wavelength of the quartz halogen light bulbs used in the final system.

Because an on-axis lens is radially symmetric the efficiency is plotted against radius. As expected, the overall efficiency of the lenses increases with radius and with thickness, when replayed at the recording wavelength. However, when replayed at 632.8 nm the efficiency can also be seen to fall at higher radii as thickness is increased.

The results suggest that the optimum gelatin thickness for the distance sensor is about 10 μm. From the theoretical calculations, the maximum overall efficiency that can be obtained for this thickness is 64.9% at 514.5 nm and 63.8% at 632.8 nm, but this could be higher if anti-reflection coatings were added to the external surfaces of the
hologram. However, it may not be possible to achieve this performance in practice due to the effects of thickness changes and losses in the processed gelatin.

8.2.3 Aberrations in the on-axis lens.

If the hologram in Fig. 8.2 is replayed with the conjugate of the plane recording beam at the original recording wavelength and angle there should be no aberrations in the replayed wavefront, despite the presence of the substrate and coverplate (assuming this is flat), and a sharp focus will be obtained. However, when the hologram is replayed at other wavelengths, aberrations will occur.

Using the raytracing program developed in Chapter 7, the effect of changing the replay wavelength on the spot size at the focus was investigated. The hologram was assumed to have a 3 mm substrate and a 2 mm coverplate, and was replayed at normal incidence for over the range of wavelengths that will be used in the experimental measurements. The minimum spotsize which was obtained at each wavelength is shown in Fig. 8.4.

It can be seen that the performance of the lens falls progressively as the difference between the replay wavelength and the recording wavelength increases and that above 700 nm the spot size is greater than 1.0 mm.

The effect of aberrations and the poor quality of the focus will be to reduce the maximum resolution which can be achieved in the final measurement system. It will be shown in Sec. 8.6.1 that reducing the source pinhole size increases resolution. However, no increase in resolution will be obtained if the pinhole is made smaller than the minimum spot size at the wavelength in use.

An off-axis replay configuration is used in Sec. 8.8 and therefore the theoretical performance of the lens was also investigated as the
angle of the incident plane beam was changed. The minimum spot size, obtained at 514.5 nm, against off-axis replay angle is shown in Fig. 8.5. Again the performance of the lens is poor away from the original recording angle.

8.3 FABRICATION AND PERFORMANCE OF AN ON-AXIS LENS.

8.3.1 Recording procedure.

The recording of an on-axis holographic lens is more complicated than off-axis types because the diverging and collimated beams must be collinear. A way in which this may be achieved is shown in Fig. 8.6. A mirror with a 1 cm hole drilled in the centre is used to deflect the plane beam onto the axis of the hologram.

Because uniform illumination over a wide angular range is required to obtain the high numerical aperture, it is necessary to use two microscope objectives to produce the diverging spherical beam. The first objective expands the laser beam to fill the aperture of the second. Because the second objective must be very close to the mirror it is not possible to place a pinhole at this point to act as a spatial filter, but it is possible to place one between the two objectives. This lack of spatial filtering after the final objective results in some small variations in intensity in the diverging beam and this causes some small local variations in efficiency in the final hologram. However, in this application, these variations have little effect.

10 μm gelatin, sensitised in 2% ammonium dichromate was used with an exposure of 600 mJ/cm². This was processed with the standard procedure using an initial processing temperature A = 25°C, and a final processing temperature B = 35°C, in order to obtain the required modulation. Because the efficiency of a transmission hologram falls when it is overmodulated it is important to obtain the correct modulation to
Fig. 8.5 Theoretical minimum spot size at the focus of the on-axis lens against off-axis replay angle, for replay at 514.5 nm.

Fig. 8.6 The technique used for recording the 0.4 N.A., 9 cm focal length, on-axis holographic lens (at 514.5 nm).
achieve maximum efficiency. The exposure and processing temperature must therefore be carefully chosen to suit the gelatin thickness.

A small area at the centre of the plate is exposed only to the diverging beam at exposure because of the hole at the centre of the mirror. This area was masked out to prevent the incident light passing through at replay and therefore the completed lens has an annular aperture.

8.3.2 Efficiency measurements.

The efficiency with which the lens diffracts light at normal incidence into the first diffracted order was measured at different radial positions across the hologram using the apparatus in Fig. 8.7. The hologram was moved using a stepper motor driven translation stage whilst the photodiode was aligned with the diffracted beam by hand. Intensity measurements from the photodiode were read directly into a 380Z microcomputer which also controlled the translation stage.

The results are plotted in Fig. 8.8 for replay at 514.5 nm and 632.8 nm. The behaviour of the lens is close to that predicted by the theoretical model but the efficiency is less, reaching a maximum of 74%. The overall efficiency of the lens can be calculated by averaging these measurements over the total area of the hologram. Because the area of the outer region is much higher than the inner region the low efficiency found at low radii does not reduce the average efficiency as much as might be expected. The overall efficiency was found to be 54.5% at 514.5 nm and 44.1% at 632.8 nm. This is about 16% less than the maximum predicted theoretically either because the modulation achieved was not exactly the optimum value or due to the processing limitations discussed earlier.

An exact agreement with the theoretical model would not be expected because the intensity of the diverging spherical recording beam
OPTICAL MEASUREMENT OF POSITION USING AN ON-AXIS LENS.

Fig. 8.7 Apparatus used for measuring the local efficiency of the lens. The photodiode must be moved in conjunction with the hologram on the translation stage.

Fig. 8.8 Experimental measurements of the local efficiency of the on-axis lens against radius, for replay at 514.5 nm and 632.8 nm.
is not uniform at the recording plate due to the limitations of the recording geometry, the lack of a final spatial filtering, and because expanding the recording beams too much wastes the available light and results in undesirably long exposure times.

The efficiency of this holographic on-axis lens is a considerable improvement on the performance a Fresnel zone plate. Because there are two passes through the lens, the system efficiency is improved by a factor of more than 3.3 and the numerical aperture is much larger. The lack of efficiency at low radii is not particularly important because the light source could be biased to illuminate the outer annular area. The high efficiency at larger radii is important because the increasing numerical aperture of the lens increases the resolution and the collecting efficiency of the measuring system.

8.4 CONSTRUCTION OF THE MEASUREMENT SYSTEM.

There are several possible configurations of the position measurement system. In particular it is possible to replace the pinholes in Fig. 8.1 with the ends of optical fibres so that the sensor can be operated remotely from the light source and detector, which might be useful in some applications.

It would be possible to eliminate the necessity of a conventional lens in the sensor if a holographic lens was recorded with collinear diverging and converging spherical waves. However, recording such a lens would difficult. In order to assess the effect of the aberrations of the holographic lens on the performance of the system, the conventional lens chosen for the measurements in this study was a high quality achromatic doublet, but in normal use a simpler lens would be adequate.

The light source for the sensor must be limited in bandwidth to eliminate secondary peaks from higher order foci, as discussed earlier.
It must also be capable of focusing enough light through the initial pinhole so that sufficient light reaches the detector after the system losses. The spectral output of the light source over the required bandwidth does not need to be uniform but the sharp peaks seen in arc lamps might cause problems. For this study, a 100 Watt quartz halogen slide projector was used with a pinhole positioned just forward of the front lens.

Two types of detector were used for the system: a spectrometer and a colour sensing photodiode detector provided by the National Physical Laboratory. The high resolution of the spectrometer allowed the limits of the system performance to be determined but in practical applications this expense and complexity would not required and the colour sensing photodiode system described in Sec. 8.9 would be more practical.

8.5 OUTPUT RESPONSE OF THE SENSOR.

The configuration shown in Fig. 8.1 was constructed using a 10 μm thick, 0.4 N.A, 9 cm focal length, holographic lens. A 0.6 mm pinhole was placed at the source and a 0.2 mm pinhole at the output of the system, which was also the input pinhole for the spectrometer.

The behaviour of the system was investigated by placing a mirror normal to the optical axis, 9 cm from the holographic lens. If a white screen was placed in the position of the output slit a bright coloured image of the source pinhole could be seen. If the screen was moved back and forth along the optical axis containing the output pinhole it was possible to see images of the source of varying colour. If the mirror was replaced with a white diffuse surface the behaviour of the system changed. A sharp image of the focused spot on the diffuse surface could be seen at only one position along the optical axis. If this position was
changed the image became blurred. This particular position is equivalent to the position of the source pinhole i.e. it is the focal point of the conventional lens.

The output pinhole was placed at this position using the image of the focus from the diffuse surface for alignment. To avoid spurious foci due to surface reflections falling on the output slit, it was necessary to introduce a small offset angle to the holographic lens. With a mirror positioned normal to the optical axis and 9 cm from the holographic lens, the spectrum of the light passing through the output pinhole was investigated with the spectrometer and is shown in Fig. 8.9.

A sharp peak is seen close to the original recording wavelength for both test surfaces. The peak is particularly sharp when the test surface is a mirror and has a half power bandwidth of 2.3 nm. The peak seen for the diffuse surface is broader and is slightly asymmetric. This asymmetry was found to change if the output pinhole position was changed and it is thought to be the result of any misalignment of this pinhole. The peak intensity of the light reaching the detector is about a factor of 15 smaller for the diffuse surface than for the mirror.

When the position of the test surface was moved the peak wavelength of the output varied, as expected. The peak output wavelength of the sensor, as a function of the position of the mirror, is shown in Fig. 8.10. The working range of the sensor, set by the usable spectral range from the light source, was between 6.0 and 12.0 cm.

The spectrum of the output signal from the sensor was measured with the mirror placed at several distances from the holographic lens (Fig. 8.11). The bandwidth of the output increases significantly at long wavelengths (short distances) due to the changing geometry of the dispersed focus on the test surface and due to the increasing aberrations in the holographic lens. Some slight broadening can be seen at short
OPTICAL MEASUREMENT OF POSITION USING AN ON-AXIS LENS.

Fig. 8.9 Spectral output of the system, measured by a spectrometer, using a mirror or a diffuse test surface at normal incidence, 9 cm from the H.O.E.

Fig. 8.10 Peak output wavelength of the system against the distance of the test surface.
Fig. 8.11 Spectral output of the system for different distances of a mirror test surface.

Fig. 8.12 The effect of the source pinhole size on the spectral output of the system, for a mirror test surface, 9 cm from the H.O.E. (The variation in overall signal level has been removed for clarity)
OPTICAL MEASUREMENT OF POSITION USING AN ON-AXIS LENS.

8-12

wavelengths which is also thought to be due to aberrations of the holographic lens.

8.6 THE RESOLUTION OF THE SYSTEM.

Assuming that the resolution of the detector is adequate, the resolution of the system is dependent on the bandwidth of the peak seen in the output spectrum. This bandwidth will be determined by the geometry of the system including the size of the pinholes and apertures, and the rate at which the focal length of the holographic lens changes with distance. This will determine how rapidly light either side of peak wavelength is dispersed around the focus on the test surface and therefore the contrast of the peak wavelength with respect to the rest of the spectrum. Aberrations in either of the lenses will blur the focus and will therefore affect the output spectrum bandwidth and hence the resolution.

It is possible to detect movements of about 40 μm in the middle of the measurement range (9 cm) when using the 0.6 mm source pinhole. This corresponds to a shift in wavelength of 0.25 nm. However at the limits of the measurement range the increase in bandwidth and the change in the rate of change of wavelength with distance, reduce the resolution to about 150 μm at a distance of 12 cm and to about 400 μm at 6 cm.

8.6.1 The effect of source size.

In order to obtain a measurable output signal the source pinhole must be large enough to let sufficient light into the system. However the effective source size also affects the resolution of the system. The effect of pinhole size on the bandwidth of the output peak is shown in Fig. 8.12. For these measurements the mirror was placed 9 cm from the holographic lens and the spectrometer pinhole was always 0.2 mm.
It can be seen that there is a progressive deterioration of the resolution, due to increasing bandwidth, as the aperture is increased. There is also a rapid rise in the average level of the output signal (which is not shown for clarity). In order to obtain a measurable signal at the limits of the working range of the sensor, where the efficiency is low, it was not feasible to reduce the source pinhole size below 0.6 mm using the simple light source described. However, if a more efficient illuminating system was used or if the measurement range required was smaller the results suggest that resolution could be increased by reducing the pinhole size further.

8.6.2 The effect of aperture size.

Using the same configuration and a 0.6 mm source pinhole the spectrum of the output signal was measured for three different apertures at the holographic lens (Fig. 8.13). Reducing the aperture resulted in a reduction in the level of the output signal and an increase in bandwidth leading to a loss of resolution. There was a large increase in resolution between 30 mm and 50 mm aperture, but less of an increase between 50 mm and 70 mm aperture. This could be because the resolution of the system is then limited by some other factor, such as the source pinhole size.

8.6.3 The effect of a tilt in the test surface.

In many situations it is likely that the test surface will not be exactly normal to the optical axis. It will be seen in Sec. 8.7 that the efficiency of the system is not particularly affected by tilt in diffuse test surfaces. However, because a tilted surface will cut through the dispersed focus of the holographic lens at an angle it might be expected that some change may occur in the spectrum of the output signal.

The spectrum of the output signal for a diffuse surface at normal incidence and one at 30° to the optical axis is compared in Fig. 8.14.
Fig. 8.13 The effect of the numerical aperture of the H.O.E. on the spectral output of the system, for a mirror test surface, 9 cm from the H.O.E.

Fig. 8.14 The effect of a 30° slant in the test surface on the spectral output of the system, for a diffuse test surface 9 cm from the H.O.E.
The surface was 9 cm from the holographic lens and a 0.6 mm source pinhole was used. It can be seen that the tilt increases the bandwidth by about 17% but the loss of resolution this causes is quite small.

8.7 SYSTEM EFFICIENCY.

Although it is not shown in Fig. 8.11, the intensity of the output signal varies widely over the working range. The peak output intensity as a function of test surface position is shown in Fig. 8.15. This variation is due to the variation in the output of the light source with wavelength and due to the wavelength selectivity of the holographic lens (which is compounded because there are two passes through it). As a result of this it is important that the wavelength detecting system is insensitive to large changes in signal intensity if the full working range of the system is to be employed. It may be possible to reduce the variation output intensity by filtering but only at the expense of the overall efficiency of the system.

It has already been seen that a slant in a diffuse test surface does not significantly affect the resolution of the system. However, the affect on the efficiency of the system has not been explored. The peak intensity of the output signal was therefore measured at a number of slant angles, for the mirror and for the diffuse surface, located 9 cm from the holographic lens. The results are shown in Fig. 8.16. When the angle of the mirror exceeds half the apex angle of the focus (20°) the incident light is deflected completely away from the holographic lens and the efficiency of the system falls to zero. However, for the diffuse surface, a slant of 45° is required before the efficiency falls by 50% and operation of the sensor with even larger tilt angles is possible. In situations where a large degree of tilt is likely to occur it is therefore necessary to use a diffuse test surface. However, small
Fig. 8.15 The variation in the peak output signal intensity against test surface distance, for a mirror or a diffuse test surface.

Fig. 8.16 The variation in the peak output signal intensity against test surface slant angle, for a mirror or a diffuse test surface, 9 cm from the H.O.E.
deviations from the optical axis have little effect for either of the test surfaces and exact alignment is unnecessary.

8.8 OFF-AXIS CONFIGURATION FOR ORIENTATION MEASUREMENTS.

The measurement system described so far measures the distance of the test surface along the optical axis. If it was possible to measure the distance along other axes projected out from the lens it would be possible to measure the position of a surface in several places simultaneously. If the surface was flat it would be possible to determine its distance and orientation with respect to the optical axis unambiguously from three distance measurements along three known axes.

This would be useful in a number of applications. For example, a robot may be required to locate a bolt in a drilled hole. If it tries to do so at anything other than normal incidence it will be unsuccessful. It may possible to detect the position of the hole by analysing the image from a video camera but it would be difficult to determine the orientation of the surface in this way.

A possible configuration for measuring the distance along three axes is shown in Fig. 8.17. Three optical fibres provide illumination and collection of the reflected light and three sets of the source/detector optics are required. The three axes are defined by the position of the ends of the fibres with respect to the optical axes of the lenses and, because the configuration is confocal, the fibres are automatically aligned with the reflected signal.

For this system to work there are several requirements. The first is that the system continues to work when there is a considerable tilt in the test surface. The second requirement is that the system continues to work when the source is located at an adequate distance from the optical axis.
Fig. 8.17 Possible configuration for measuring distance along three different (off-axis) axes using an on-axis lens. If the axes are not coplanar, it is possible to deduce the orientation of a flat surface from these distances.

Fig. 8.18 The variation in the peak output signal intensity against off-axis replay angle, for a mirror test surface, 9 cm from the H.O.E.
The first requirement would be satisfied if a diffuse test surface is used. To test the second requirement, an optical fibre version of the sensor was constructed. The output pinhole was replaced by the end of a 1 mm polymer optical fibre which was then connected to the spectrometer. This optical fibre has a high acceptance angle (N.A. = 0.58) and therefore collects most of the incident light. The source was not altered except that a 1 mm pinhole was used. The use of the fibre allowed the source and detector to be moved off axis easily without any alignment problems with the spectrometer.

As mentioned in Sec. 8.5 it has already been necessary to position the source slightly off axis relative to the holographic lens to avoid spurious foci, due to surface reflections, falling on the output pinhole. This offset is typically 0.2°. If the separation of three separate test points on the test surface is to be adequate for accurate triangulation to take place, the off-axis angle of each source will have to be much larger.

The intensity of the output signal was measured for a range of off-axis angles, adjusting the position of the source pinhole and output fibre for each case. The results can be seen in Fig. 8.18 The efficiency falls quite rapidly with angle and has fallen to 20% at 3°. These losses might be acceptable on their own but coupled with the losses due to the use of a diffuse surface rather than a mirror and with the loss of efficiency at the limits of the measuring range, they may not be a sufficient output intensity. However, if the off-axis angle is reasonably small and the measurement range is limited to the region of high efficiency, the output intensity of the sensor will be adequate. It may be possible to reduce the loss of efficiency with off-axis angle by using a thinner hologram, thus reducing the angular selectivity.
Placing the source off-axis also has an affect on the spectrum and bandwidth of the output signal. The spectrum of a diffuse surface at 9 cm for the system 3° off-axis is compared with the system on-axis in Fig. 8.19. The bandwidth of the output peak is considerably increased and the resolution of the system has fallen to about 200 μm.

An off-axis angle of 3° would result in a separation of the three measuring points of over 8 mm at 9 cm from the lens. A resolution of distance of 200 μm at this separation would allow the angle of a surface to be determined to within 1.4°.

A possible solution to the problems which occur due to off-axis replay would be to separate the lens into three sections, exposing each individually to one of three pairs of recording beams arranged along different axes. The system would be then be equivalent to placing a third of three normal on-axis systems in close proximity and would not suffer the disadvantages of changing the replay angle but the aperture for each of the axes would be reduced.

8.9 USE WITH A COLOUR SENSING PHOTODIODE DETECTOR

Although a spectrometer has been used for all previous measurements in this chapter, it would not in practice be the best method of decoding the signal wavelength since it is bulky and expensive. It also does not present the final data in a very useful form and it is not possible to make real time measurements.

A more appropriate detector, based on a Sharp semiconductor colour sensing photodiode, was lent by the National Physical Laboratory. The photodiode in this device is in fact two superimposed photodiodes which have different spectral responses, as one acts as a spectral filter for the other. When illuminated with reasonably monochromatic light the ratio of the output from the two detectors varies with wavelength.
Fig. 8.19 The spectral output of the system when operated 3° off-axis compared with that when operated on-axis, for a diffuse test surface, 9 cm from the H.O.E.

Fig. 8.20 Output voltage from the colour sensing photodiode detector against test surface distance.
The advantage of using such a system is that the output signal can be read directly as a voltage and the response time to rapid movements is limited only by the response of the photodiodes and photodiode amplifier/divider which can be made very fast. The system also has the advantage that it is cheap and compact.

8.9.1 Calibration.

The photodiode sensor was designed to operate with wavelengths between 600 nm and 1 μm. Unfortunately this does not match the optimum measurement range of the dichromated gelatin lens. Fabricating a lens to work optimally at the centre of this range would be more difficult due to the lack sensitivity when recording at these wavelengths. However, satisfactory results were obtained with a test surface placed between 6 and 9 cm from the lens. The output from the photodiode sensor is relatively insensitive to variations in the light level and changing the intensity by a factor of 30 causes the output to change by only 1% of the full output range.

The photodiode and the light source were both connected to the optical measurement device using 1 mm optical fibres. It was found that a movement of 40 μm caused a voltage change of about 1.8 mV and the sensitivity and stability of the device exceeded the resolution of the distance measurement system, despite its simplicity.

A calibration curve of voltage against distance was obtained using a mirror test surface and is shown in Fig. 8.20. The fall in output voltage as the distance is reduced below 5.4 cm is thought to be due to the presence of a second order focus which would confuse the sensor by adding a second spectral component of considerable strength.

8.9.2 Real-time measurements.

To demonstrate that combining the distance measurement system with the colour sensing photodiode allows real time measurements to be made, a
Fig. 8.21 Square wave output voltage from the colour sensing photodiode detector when the system is used to examine the distance of a two blade shutter rotating at 500 r.p.m (a) for a fixed surface 2 cm behind the blades and (b) for a fixed surface 1 cm behind the blades.

Fig. 8.22 Real-time measurement of the surface profile of a rotating disc using the distance measurement system with the colour sensing photodiode detector.
fixed test surface was placed 8 cm from the holographic lens behind a two bladed chopper rotating at 500 r.p.m. Both the fixed surface and the chopper blades were white painted surfaces and hence the distance seen by the sensor oscillates rapidly. The output from the photodiode sensor was then monitored using a storage oscilloscope. The square wave output voltage which was obtained can be seen in Fig. 8.21 (a) for a separation of 2.0 cm between the the chopper and the fixed surface and (b) for a separation of 1.0 cm. Some distortion can be seen due to low pass filtering of the output from the voltage divider and some small glitches occur at the edges of the blades. However, once the output has settled it is an accurate indication of the separation between the fixed and rotating surfaces and it is possible to detect small differences in the orientations of the two chopper blades.

The system was also used to measure the surface profile of a rotating disc. The disc was divided into four quadrants, one of which was 0.25 mm higher and another 0.5 mm higher than the surrounding area. When this disc is rotated in front of the sensor the distance between the surface and the lens varies rapidly in the repeating sequence: 7.000, 7.025, 7.000, 7.050 cm. These features can be seen clearly in Fig. 8.22 which was obtained from the storage oscilloscope.

8.10 SUMMARY.

An efficient on-axis holographic lens has been fabricated with suitable properties for a distance measurement sensor. The efficiency and numerical aperture are a considerable improvement over a Fresnel zone plate. A compact position measurement sensor has been demonstrated which can be operated remotely using optical fibres.

The sensor can be used to determine the distance of a mirror or a bright, diffusely reflecting surface over a range of 6 cm and is
insensitive to the alignment or reflectivity of this surface. The factors affecting the resolution of the sensor have been investigated and the resolution reaches a maximum of 40 μm at the centre of the measurement range.

It has also been shown that it is possible to measure the distance of a surface along different axes and thereby determine the orientation of the surface, but adopting this system results in a loss of efficiency and resolution.

Finally, the system was shown to be capable of making rapid real-time measurements of moving surfaces.
A study has been made of the use of dichromated gelatin as a holographic recording material. Sufficient control over environmental conditions and processing procedures has been achieved to enable a quantitative study of the recording properties of the material to be made.

Initially, the bulk properties of the material were investigated. It was found that processing induced a change in the thickness and refractive index of the material and that a uniform exposure affected the amplitude of these changes. A simple model was developed which related the thickness (and therefore the density) to the refractive index. The changes in the bulk properties of uniformly exposed dichromated gelatin were found to be very similar to those occurring in recorded and processed holograms, where there is always a uniform component in the exposure. The recording response of the material has been investigated for planar reflection gratings by comparison with coupled wave theory. A mathematical model of the recording characteristic has been found to give good agreement with the observed behaviour, such as the saturation at high exposures and the formation of harmonic intermodulation gratings in double exposure holograms. However, there were some differences between the angular response of the planar gratings and the response calculated theoretically using uniform grating coupled
SUMMARY AND CONCLUSIONS.

Wave theory. It was found that the agreement between experiment and theory could be improved substantially if the grating parameters were allowed to vary with depth in the theoretical model. Using this improved model it was possible to examine the non-uniformities in planar reflection gratings and it was found these were strongly affected by processing.

The formation of real-time gratings in the dichromated gelatin have been investigated and a dynamic, time dependent form of coupled wave theory has been developed which gives good agreement with the experimental results. It has been possible to show, quite unambiguously, that the real-time gratings are pure absorption gratings, although some phase modulation may appear later if the gratings are exposed to a humid atmosphere. This is the first time that the mechanism behind the real-time gratings has been fully explained.

The copying of silver halide holograms into dichromated gelatin has been studied, with particular interest in the level of noise in the copy holograms. It has been shown that the scatter present during copying is recorded and that this can, in some circumstances, lead to strong diffraction. However, despite this, the level of noise in the copies is substantially lower when replayed in white light. It has been found that highly efficient noise gratings can be fabricated in dichromated gelatin by recording the scattered light field from a silver halide emulsion. These 'synthesised noise gratings' were found to have similar properties to those reported of silver halide noise gratings. It is hoped that some of the behaviour which has been observed will help in the development of a better understanding of noise gratings in general.

A theoretical model of holographic optical elements has been developed which combines vector coupled wave theory with a ray tracing approach. Using this model, it has been possible to show the effect of
the polarisation of light on the diffraction efficiency of holographic lenses and, for example, that off-axis lenses are more sensitive to polarisation.

Finally, an optical method of distance measurement has been investigated which utilises the dispersive properties of an on-axis holographic lens. The theoretical model developed earlier has been used to optimise the lens for this application. The method has been shown to be capable of making remote, real-time measurements of the distance of a mirror or a diffusely reflecting surface. A resolution of 40 µm was achieved at a distance of 9 cm.
THE MODULATION OF HARMONIC INTERMODULATION GRATINGS.

In this analysis we obtain expressions for the amplitude of the refractive index modulation of harmonic intermodulation gratings which occur in double exposure holograms due to the non-linearity of the recording characteristic.

The exposure at a depth x in a double exposure hologram is

\[ E(x) = E_1(x) + E_2(x) \] (I-1)

where

\[ E_1(p) = E_1 \left[ (1+r) + 2 r \cos(\vec{K}_1 \cdot \vec{p}) \right] \] (I-2)

\[ E_2(p) = E_2 \left[ (1+r) + 2 r \cos(\vec{K}_2 \cdot \vec{p}) \right] \] (I-3)

where

\[ E_1 = I_1 t_1, \quad E_2 = I_2 t_2 \] (I-4)

and where \( I_1 \) and \( I_2 \) are the intensities of the reference beam of each exposure, \( t_1 \) and \( t_2 \) are the exposure times, \( r \) is the beam ratio of both exposures, \( \vec{K}_1 \) and \( \vec{K}_2 \) are the grating vectors and \( \vec{p} \) is the position vector.

The recording characteristic of the recording material is assumed to be of the form (3.6)

\[ n(p) = N_0 \left[ 1 - \exp\left( -E(p)/E_0 \right) \right] \] (I-5)
where \( n(p) \) is the change in refractive index due to exposure.

Substituting for \( E(p) \) using (I-1) we obtain

\[
n(p) = N_0 \left[ 1 - \exp\left(-\frac{(E_1 + E_2)(1 + r)}{E_0}\right) \times \exp\left(-2\frac{rE_1}{E_0}\cos(\vec{k}_1 \cdot \vec{r})\right)\right] \times \exp\left(-2\frac{rE_2}{E_0}\cos(\vec{k}_2 \cdot \vec{r})\right) \tag{I-6}
\]

Using the relationship [ABR65]

\[
\exp(z\cos\theta) = I_0(z) + 2 \sum_{k=1}^{\infty} I_k(z) \cos(k\theta) \tag{I-7}
\]

where \( I_k(z) \) are modified Bessel functions of order \( k \).

we can write

\[
n(p) = N_0 \left[ 1 - \exp\left(-\frac{(E_1 + E_2)(1 + r)}{E_0}\right)\right] \times \left[ I_0(z) + 2 \sum_{q=1}^{\infty} I_q(z) \cos(q\vec{k}_1 \cdot \vec{r})\right] \times \left[ I_0(v) + 2 \sum_{m=1}^{\infty} I_m(v) \cos(m\vec{k}_2 \cdot \vec{r})\right]
\]

where \( z = -2\frac{rE_1}{E_0} \) and \( v = -2\frac{rE_2}{E_0}. \) Using the relationship

\[
\cos A \times \cos B = \frac{1}{2} \cos(A-B) + \frac{1}{2} \cos(A+B) \tag{I-9}
\]

we obtain the expression

\[
n(p) = N_0 \left[ 1 - \exp\left(-\frac{(E_1 + E_2)(1 + r)}{E_0}\right)\right] \times \left[ I_0(z) I_0(v) + \right.
\]

\[
2I_0(v) \sum_{q=1}^{\infty} I_q(z) \cos(q\vec{k}_1 \cdot \vec{r}) + 2I_0(z) \sum_{m=1}^{\infty} I_m(v) \cos(m\vec{k}_2 \cdot \vec{r}) + \left.
\right]
\]

\[
2 \sum_{q=1}^{\infty} \sum_{m=1}^{\infty} I_q(z) I_m(v) \cos((q\vec{k}_1 - m\vec{k}_2) \cdot \vec{r}) + 2 \sum_{q=1}^{\infty} \sum_{m=1}^{\infty} I_q(z) I_m(v) \cos((q\vec{k}_1 - m\vec{k}_2) \cdot \vec{r}) \right] \tag{I-10}
\]
It can be seen that as a result of a cosinusoidal exposure we obtain an infinite series of harmonics of the two fundamental gratings, $q\hat{K}_1$ and $m\hat{K}_2$, but also an infinite series of intermodulation gratings of the form, $(q\hat{K}_1 \pm m\hat{K}_2)$. The amplitude of the refractive index modulation of the fundamental and the harmonic gratings can be found from (I-10).
APPENDIX II.

COUPLED WAVE THEORY FOR NON-UNIFORM GRATINGS.

In this analysis we obtain coupled wave equations for the two wave case in which the refractive index modulation, the average refractive index and the fringe spacing are functions of depth. The absorption constant and the absorption modulation are not made functions of depth since in dichromated gelatin they are both small and have little effect. The complex permittivity is therefore

\[ \varepsilon_r(r) = \varepsilon'_r(x) - j\varepsilon''_r(x) + [\varepsilon'_r(x) - j\varepsilon''_r(x)] \cos \phi \quad (\text{II.1}) \]

where

\[ \phi = \int_0^x K_x(s) \, ds + K_y \, y \quad (\text{II.2}) \]

and where \( K_x(s) \) represents the variation in the fringe spacing with depth, \( \varepsilon'_r(x) \) is the variation in the average permittivity with depth, \( \varepsilon'_r(x) \) is the variation in the permittivity modulation with depth, \( \varepsilon''_r(x) \) represents the average absorption constant and \( \varepsilon''_r(x) \) the absorption modulation. We assume that the \( y \) component of the grating vector is constant since the gelatin is restrained from movement in this direction by the rigid glass substrate.

We first find solutions to the scalar wave equation where there is no coupling i.e. \( \varepsilon'_r(x) = 0 \) and \( \varepsilon''_r(x) = 0 \).

\[ \nabla^2 E + \beta(x)^2 E = 0 \quad (\text{II.3}) \]
and try solution of the form

\[ E = A \exp(-j\beta_a L(x,y)) \]  

(II-4)

where \( \beta_a \) is the average value of the propagation constant in the medium.

Substituting this into the wave equation we obtain

\[ -2j\beta_a \nabla V L - j\beta_a A^2 L - \beta_a^2 A^2 (\nabla L)^2 + \beta(x)^2 A = 0 \]  

(II-5)

Equating real parts separately to zero

\[ -\beta_a^2 (\nabla L)^2 + \beta(x)^2 = 0 \]  

(II-6)

A possible solution to this would be

\[ L(x,y) = f(x) + \rho_y / \beta_a \]  

(II-7)

Hence

\[ \nabla L = \frac{\partial f}{\partial x} \hat{x} + \rho_y / \beta_a \hat{y} \]  

(II-8)

\[ (\nabla L)^2 = \left[ \frac{\partial f}{\partial x} \right]^2 + \left[ \frac{\rho_y}{\beta_a} \right]^2 \]  

From (II-6)

\[ \left[ \frac{\partial f}{\partial x} \right]^2 = \left[ \beta(x)/\beta_a \right]^2 + \left[ \frac{\rho_y}{\beta_a} \right]^2 \]  

(II-9)

and therefore

\[ f(x) = \frac{1}{\beta_a} \int_0^x [\beta(x)^2 - \rho_y^2] \frac{1}{2} \, dx \]  

(II-10)

Hence

\[ L(x,y) = \frac{1}{\beta_a} \int_0^x \left[ \beta(x)^2 - \rho_y^2 \right] \frac{1}{2} \, dx + \frac{\rho_y}{\beta_a} y \]  

(II-11)

and
\[ \nabla L \approx \rho_x/\beta_a \hat{i}_x + \rho_y/\beta_a \hat{i}_y \]  
(II-12)

where \( \rho_x \) and \( \rho_y \) are the \( x \) and \( y \) components of the propagation vector of the wave in a medium with propagation constant \( \beta_a \).

We now introduce coupling. Assuming the variation in \( \epsilon'_0(x) \) is small \( \beta(x) \approx \beta_a \). From (II-1) the wave equation can be written

\[ \nabla^2 E + \left( \beta(x) \right)^2 - 2j\alpha_0 \beta_a + 2\kappa(x) \beta_a \left[ \exp(j\phi) - \exp(-j\phi) \right] E = 0 \]  
(II-13)

where \( \alpha_0 = \frac{\beta_a \epsilon''_0}{2\epsilon'_0} \) and \( \kappa(x) = \beta_a \left[ \frac{\epsilon''_{10}(x) - j\epsilon''_{11}}{4\epsilon'_0} \right] \)

Assuming the electric field in the hologram is of the form

\[ E = R \exp\left\{ -j\beta_{a\text{r}} L_r(x,y) \right\} + S \exp\left\{ -j\beta_{a\text{s}} L_s(x,y) \right\} \]  
(II-14)

where

\[ L_r = \frac{1}{\beta_a} \int \left[ \beta(s) \right] \frac{1}{2} ds + \frac{\rho_y}{\beta_a} y \]  
(II-15)

\[ L_s = \frac{1}{\beta_a} \int \left[ \beta(s) \right] \frac{1}{2} ds + \frac{\sigma_y}{\beta_a} y \]

and where we have chosen the diffracted wave in the hologram to be defined by beta value theory (1.11). Substituting into the wave equation and neglecting the higher order terms \( \nabla R, \nabla S, \nabla L_r, \nabla L_s \), all derivatives of the slowly varying terms \( \epsilon''_{10}(x), \epsilon''_{11}(x), \kappa(x) \) and all higher diffracted orders we obtain

\[ \begin{align*}
[-2j\beta_a VR_{L_r} - \beta_a R(\nabla L_r)^2 + \beta^2(x) R - 2j\alpha_0 \beta_a R] \exp\left\{ -j\beta_{a\text{r}} L_r \right\} + \\
[-2j\beta_a VS_{L_s} - \beta_a S(\nabla L_s)^2 + \beta^2(x) S - 2j\alpha_0 \beta_a S] \exp\left\{ -j\beta_{a\text{s}} L_s \right\} + \\
2\kappa(x) \beta_a [R \exp\left\{ -j(\beta_{a\text{r}} L_r - \phi) \right\} + S \exp\left\{ -j(\beta_{a\text{s}} L_s + \phi) \right\}] = 0
\end{align*} \]  
(II-16)
Equating coefficients of the exponential terms and using (II-8) and (II-12) the coupled equations are obtained

\[
\frac{\rho}{\beta_a} \frac{\partial R}{\partial x} + \alpha R + j\kappa(x)S \exp(j\beta_a \gamma) = 0 \tag{II.17a}
\]

\[
\frac{\sigma}{\beta_a} \frac{\partial S}{\partial x} + \alpha S + j\kappa(x)R \exp(-j\beta_a \gamma) = 0 \tag{II.17b}
\]

where \( \gamma = \frac{1}{\beta_a} \int_0^x \kappa(s) \, ds - \beta_a L_r + \beta_a L_s \) \tag{II.18}

Note that since \( \gamma(x) \) is a function of both \( \kappa(x) \) and \( \epsilon'_\infty(x) \) it will not be possible to distinguish the effect of these two variables.
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