

# Fluorescence Microscopy Investigation on Residual Stresses in Alumina-based Ceramics

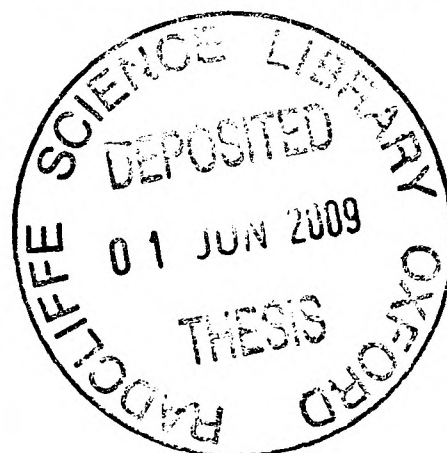
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## Abstract

Grinding/polishing and indentation induced residual stresses were measured by confocal  $\text{Cr}^{3+}$  fluorescence microscopy with high spatial resolution ( $\sim 2 \mu\text{m}$ ), obtaining local stress variation information rather than the mean stress averaged over a large sampling volume as is measured by other techniques.

Due to the translucency of alumina materials, a substantial portion of the fluorescence signal comes from beneath the surface of the specimen. A probe response function (PRF) was developed taking account of microscope resolution, refraction, absorption and scattering, to quantitatively describe where the collected signal came from. It described the fluorescence intensity variations against defocus distance very well for a range of materials including sapphire, ruby, polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites.

Large variations in the residual stresses on ground and polished surfaces were observed, owing to the surface fracture and pullouts. The broad peaks and narrow peaks separated from the spectra collected near the ground/polished surfaces physically represented the two distinct regions in the ground region: a plastically deformed surface layer and the elastically deformed material underneath. A model for the residual stress field taking into account the pullout was proposed using an array of virtual dislocations. The model agreed with the experimental results well when the PRF was included. Tensile stresses were detected on the ground surfaces of polycrystalline aluminas and 2 vol.% SiC nanocomposite, but not on the polished surfaces of polycrystalline aluminas or ground surfaces of 5 and 10 vol.% SiC nanocomposites. This was explained in terms of difference in the amount of pullouts on the surfaces. The depth of deformation was deeper in the ground polycrystalline alumina compared to the polished condition; the depth of deformation in alumina and the  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were similar ( $\sim 1 \mu\text{m}$ ) while the compressive stresses in the nanocomposites were greater owing to the reduction in pullout. The main difference between ground alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites was the brittle fracture behavior rather than the plastic deformation.

Line scans and area mapping were carried out on 1 kg loaded Vickers indentations of alumina-based ceramics. Tensile stresses were found at the tips of radial cracks and lateral cracks and compressive stresses were found around the indent impression. The line scan results in the elastic regions agreed qualitatively with Yoffe's model and the quantitative discrepancy was attributed mainly to the cracking that relaxed the stresses. The differences in residual stresses between alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were small if measured with high spatial resolution but it would be exaggerated with lower resolution.



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## **Preface**

The work presented in this thesis was carried out by the author in the Department of Materials, University of Oxford under the supervision of Dr. Richard I. Todd. The work in this thesis is original and no part of this thesis has previously been submitted for a degree at this or other university.

Part of this work has been presented by either poster or oral talk in the following conferences:

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# Chapter 1

## Introduction

### 1.1 Background

Alumina ceramics are the most widely used structural ceramic materials because of their excellent hardness, strength and wear resistance. Often their surfaces are diamond ground before use and the grinding process will introduce surface damage like cracks, pullouts and residual stress to the surface. This surface damage will certainly degrade the performance of alumina ceramics. As a result, it is important to understand the origin of this damage before we can find solutions to alleviate it. The surface stress is probably the key to this understanding as the initiation and propagation of cracks and grain pullouts can both be related to stresses.

Surface stresses in alumina materials became more interesting as researchers found that by adding a small amount of fine SiC, the surface quality after grinding can be significantly improved [1] and the pullout sizes also decrease [2]. Improved strength of  $\text{Al}_2\text{O}_3/\text{SiC}$  over pure alumina was also observed [3], and surface stresses are among the proposed strengthening mechanisms [1, 4].

There already exist some residual stress measurement techniques, such as destructive curvature [5] and hole drilling [6] methods, and non-destructive X-ray diffraction [7] and neutron diffraction [8] methods. The disadvantage of these techniques is their low



spatial resolution [9]: they are actually measuring the average stress over the large sampled volume, rather than the local stress. Confocal fluorescence microscopy is an ideal non-destructive technique to measure residual stresses in alumina ceramics: it is quick and cheap, and most importantly its higher spatial resolution enables the measurement of local stress.

## 1.2 Aims of the Project

This project aims to develop a better understanding of the nature and origin of surface damage in alumina-based ceramics, including single crystal alumina, polycrystalline alumina with different grain sizes, and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites with different SiC additions. The residual stresses caused by indentation and grinding/polishing have been mapped using  $\text{Cr}^{3+}$  fluorescence microscopy. A better understanding is made possible by the higher spatial resolution of the confocal Raman microscope ( $\sim 2 \mu\text{m}$ ), and careful curve fitting to determine the peak positions. The experimental results have been compared with existing models, and new models have been proposed if there is none available.

There are still lots of unsolved issues about surface damage of alumina-based ceramics, among which is the strengthening mechanism of alumina/SiC nanocomposites over pure alumina [3]. By measuring the grinding induced surface stress in alumina and nanocomposites, this project also aims to provide some clues to this puzzle.

### 1.3 Outline of the Thesis

This thesis is split into 8 chapters, including the brief introduction in this chapter. A review of previous research on surface damage in alumina based ceramics is given in Chapter 2. Also included are an introduction to residual stress and commonly used techniques to investigate residual stress. Materials preparation and experimental procedures are described in Chapter 3, with the emphasis on introducing the  $\text{Cr}^{3+}$  fluorescence microscopy technique and the problems and solutions with curve fitting. In Chapter 4, the spatial resolution is first measured by experiments on different materials. Then some mathematical models, called probe response functions (PRFs), are proposed to explain how the fluorescence intensity varies in the depth direction. A stress model in which the residual stresses are represented by a distribution of near surface dislocations is proposed in Chapter 5. The stress function is convoluted with the PRFs established in Chapter 4 to predict the fluorescence peak shifts that are actually measured. The depth of stressed layers and the stress level within these layers are obtained from the fitting of the convolution to the experimental results. The same fitting technique is applied to  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites in Chapter 6, to compare the grinding induced residual stress between alumina and nanocomposites. In addition to grinding and polishing, residual stresses induced by indentation are investigated in Chapter 7. The experimentally measured stresses around indentations are compared with classical stress models [10, 11]. Conclusions from this work are summarized in Chapter 8, and some suggestions for future work are also given in this chapter.

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## Chapter 2

### Literature Review

#### 2.1 Introduction

Alumina is the most commonly used structural ceramic material due to its extraordinary properties such as high surface hardness and fracture strength, low friction, high wear resistance, high temperature resistance and chemical stability. The performance and life-span of ceramic materials are affected greatly by their surface quality. Both the manufacturing and subsequent treatment, like grinding or abrasion, will introduce surface damage to the material. One typical example is for alumina as it is normally diamond ground before use and its main applications include abrasives and cutting tools [1]. Hence it is of great importance to investigate how the surface damage degrades its performance and life-span, and how to minimize the surface damage. The possible surface damage includes cracks, residual stresses, surface roughness and plastic deformation, of which the cracks and the residual stresses affect the surface condition and hence the materials performance and life-span most significantly [2]. Research progress on surface damage and surface residual stresses will be reviewed extensively in section 2.2 and section 2.3.

The surface stresses induced by indentation and grinding/polishing will be experimentally investigated in this research. Understanding the surface stress will certainly help the understanding of the crack initiation and propagation processes.

Surface stresses in alumina materials became more interesting as researchers found that by the addition of a small amount of SiC, the fracture strength is improved over pure alumina [3-7]; the surface quality after grinding is also significantly improved [8-10]. The typical microstructure of  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites is composed of a polycrystalline alumina matrix with an average grain size of 1~5  $\mu\text{m}$ , and SiC particles with size ranging from 100 to 400 nm which exist either inside the matrix grains or on the grain boundaries. Surface residual stress is among the various strengthening mechanisms of nanocomposites over alumina that have been proposed [6-9, 11-14]. In Chapter 7, the grinding induced surface stresses in alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites will be compared, to examine the validity of this possible strengthening mechanism.

## **2.2 Surface Damage of Alumina-based Ceramics**

In this section, previous research on surface damage in alumina-based materials induced by polishing and grinding, single-point scratching and indentation will be summarized.

### **2.2.1 Grinding and Polishing**

The difference between grinding and polishing processes lies in that for grinding the abrasives are fixed in a grinding wheel or stage; while for polishing the abrasives are not fixed. The abrasives used for grinding are also coarser than those used for polishing.

There are two main types of model used to describe the machining mechanism in ceramics, namely, indentation fracture models and ductile regime grinding models [15], although tribochemical reactions also play a role in some conditions [16, 17]. In the indentation fracture model, each abrasive particle is assumed to act as an indenter. Two types of crack are induced by these multiple indenters: a median crack that is perpendicular to the surface and a lateral crack that is parallel to the surface [18, 19]. The median crack is commonly regarded to be responsible for strength deterioration and the lateral crack is associated with material removal. In the ductile grinding model, the material removal mechanism is purely plastic deformation (by ploughing or cutting) [20]. To realize this material removal mechanism, the depth of cut needs to be precisely controlled which is very difficult in practice. In general, the material removal is due to a mix of both brittle fracture and ductile deformation.

Materials of high fracture strength, toughness and density will exhibit greater ductile deformation during grinding, and the degree of ductile deformation will depend on the geometry of the abrasive particles and the depth of cut per grain [15]. Ductile deformation can be developed in a brittle material when a light load and abrasive grit with small tip radius are used [21].

Grinding induced damage in alumina is dependent on grit size and the brittleness of the ceramics themselves. Damage penetrates deeper in less brittle ceramics under a given grinding condition, and larger grit size also results in deeper damage [22]. The

grain size of alumina also has a significant influence on the grinding behavior. Xu et al. found that [23] the density of grinding induced microcracks increases with the grain size. They claimed that the intra-grain twin/slip and grain boundary microcracks, rather than lateral and median cracks, are mechanism responsible for the grinding induced surface damage. The wear rate (wet erosive wear and sliding wear) of alumina also has grain-size dependence, where alumina of smaller grain size has better wear resistance [8, 17, 24, 25]. However, the exact mechanism behind this grain size dependence is not fully understood. The grain size also affects the mild to severe wear transition in that the time to transition decreases with increasing grain size [26, 27].

Subsurface deformation of ground and polished alumina and alumina/SiC nanocomposites has been investigated by TEM and FIB (focused ion beam) milling. Basal twinning and pyramidal slip are responsible for the deformation of alumina [1, 28-30]. Inkson [30] suggested that the basal twins might be confined by the grain boundaries and the plastic deformation is then limited, which results in the intergranular fracture and grain pullout for alumina during the abrasive wear (grinding and polishing). Wu et al. [31] used TEM to examine ground surfaces of  $\text{Al}_2\text{O}_3$  and 5 vol.%  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites and they found that the  $\text{Al}_2\text{O}_3$  surface suffers from intergranular fracture and grain pullout while the nanocomposites suffer from grooves and intragranular fracture, in agreement with the previous results reported by Winn and Todd [32]. The deformation in  $\text{Al}_2\text{O}_3$  is localized close to the surface with



dislocations distributed within only 1.5  $\mu\text{m}$  from the surface. The deformation mechanism is mainly basal twinning with penetration of only one grain depth into the surface. The penetration of basal twins terminates at their intersection with the first grain boundary they meet and the twin pile-ups lead to cracks that were observed along grain boundaries or interfaces of basal twins. In  $\text{Al}_2\text{O}_3$  nanocomposites the dislocation flow is the main deformation mechanism and twinning was rarely found. Dislocations penetrate to a depth of 10  $\mu\text{m}$  (about 3-4 grains), and they were possibly activated both on the surface and by the intragranular SiC particles. Microcracks were observed below the surface of the nanocomposites and the cracks are mainly intragranular. Wu et al. [10] found that finer diamond grit size used for machining will decrease the depth of dislocations. Details are given in Table 2.1. Larger uncertainty of this depth was found in nanocomposites.

Table 2.1 Depth of dislocation on ground and polished  $\text{Al}_2\text{O}_3$  and 5 vol. %  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites [10]

| Surface Treatment Conditions |                        |                          |                          |                          |
|------------------------------|------------------------|--------------------------|--------------------------|--------------------------|
|                              | Ground                 | 8 $\mu\text{m}$ polished | 3 $\mu\text{m}$ polished | 1 $\mu\text{m}$ polished |
| $\text{Al}_2\text{O}_3$      | <1.0 $\mu\text{m}$     | >1.5 $\mu\text{m}$       | 0.5-1.5 $\mu\text{m}$    | <0.3 $\mu\text{m}$       |
| Nanocomposites               | 3.0-10.0 $\mu\text{m}$ | 1.5-3.0 $\mu\text{m}$    | >1.0 $\mu\text{m}$       | 0.5-1.5 $\mu\text{m}$    |

Kara and Roberts [33] found that grain size has a strong effect on the polishing of alumina but the effect is less strong for nanocomposites. Surface roughness at a given polishing stage increases with increasing grain size for both alumina and alumina



nanocomposites, although the grain size dependence for nanocomposites is less apparent than alumina. The nanocomposites have a better surface finish than that of alumina using the same polishing conditions, mainly due to the suppression of grain pullout [7].

The wear behavior of  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites has also been intensively investigated, since researchers found that the wear resistance of monolithic alumina can be significantly improved by addition of a small amount of nano-sized SiC particles [8, 32, 34-37]. They basically investigated the effects of alumina grain size [25, 38, 39], SiC content (vol.% SiC ranging from 1.8 to 15) [8, 25, 32, 35, 39-41], SiC particle size (12 nm~2000 nm) [8, 36, 42, 43] and SiC location (intergranular or intragranular) [8, 32, 44] on the wear resistance. The improvement in wear resistance of  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites over monolithic alumina is attributed to the reduction in pullout diameter and fraction of pullout [8, 45].

### 2.2.2 Scratching

Single-point scratching is commonly used to understand the material removal mechanism of ceramics. Much work has been done on the topography of scratched surfaces, and material removal mechanisms.

Xu et al. [46, 47] proposed a material removal mechanism in scratching of alumina. The process includes intragranular microfracture and chipping formation through

crack propagation along the twin/slip planes, intergranular microfracture and grain pull-out, and removal of large segments by propagation of lateral cracks. They showed that interaction between parallel scratches leads to an increase in the material removal, which is also dependent on the grain size, load and distance between the parallel scratches. They claim that lateral and median cracks are not the main mechanism behind the materials removal in coarse-grained alumina. Irrespective of the grain size, grain dislodgement is always the predominant mechanism for material removal.

Zarudi et al. [48] found solid proof of plastic deformation of alumina at room temperature indicated by the plastic zones during the scratching process. The plastic zones are grain size sensitive, with twins and higher density of dislocations in the coarse-grained alumina and much less dislocations in the fine-grained alumina. Their further research showed that there are five independent slip and twin systems in alumina [49], i.e., one prism plane slip system,  $\{1\bar{1}20\}\langle\bar{1}100\rangle$ ; two basal slip systems:  $(0001)\frac{1}{3}[1\bar{1}20]$  and  $(0001)\frac{1}{3}[1\bar{2}10]$ ; one basal twin with  $K_1=(0001)$  and shear direction  $\eta_1=[\bar{1}100]$ ; and one rhombohedral twin system with  $K_1=(10\bar{1}1)$  and  $\eta_1=[\bar{1}012]$ .

Wu et al. [50] used TEM and FIB milling to characterize the subsurface scratches made by a  $120^\circ$  cone indenter loaded to 1N in polycrystalline alumina and 5 vol.%  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites. They found for polycrystalline alumina the plastically

deformed zone extends to a maximum depth of about 4  $\mu\text{m}$  and is contained within the first layers of grains near the scratched surface; however, this plastic zone can reach  $\sim 7 \mu\text{m}$  below the scratched grooves and extends beyond the first layer of the grains in the  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites. In both ceramics, a similar 3D crack pattern is found around the scratches (Fig 2.1): a crack-free compressive zone under the groove base (zone I,  $\sim 3 \mu\text{m}$ ); a microcrack zone below and around zone I (zone II,  $\sim 3 \mu\text{m}$ ); and zone III, a deep crack zone below zone I and II. The residual stresses measured by  $\text{Cr}^{3+}$  fluorescence are higher in the nanocomposites than those in the polycrystalline alumina.

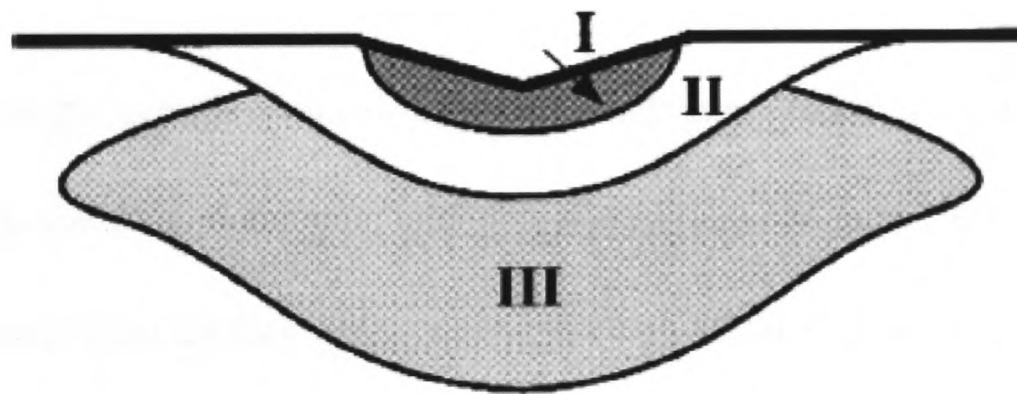


Fig 2.1 Schematic diagram of surface cracking types underneath scratches  
(I) crack deficient zone; (II) microcracking zone;  
(III) deep lateral and median crack zone (after [50])

### 2.2.3 Indentation

Understanding of fracture mechanisms is very important for brittle materials like alumina, and it is usually based on the cracking development. Lots of work has been done concerning crack initiation, its propagation and modeling the stress field around cracks. One commonly used method to introduce a crack (or flaw) is by indentation [51], in which a sharp or blunt indenter is loaded onto the surface of materials.

Various types of crack may be produced in the material surface [52], as shown in Fig 2.2. Cook and Pharr classified all crack types into five categories [52]: 1) cone cracks, generated by elastic loading of spherical or flat-punch indenters. They spread away from the surface at some angle to the load axis. For sharp indenters like Vickers or Knoop diamonds, a residual plastic zone is generated in the surface by the indentation, known as elastic-plastic contact [53]. There are four types of crack that can be generated by sharp indenters: 2) radial cracks, parallel to the load axis and initiating from the edge of plastic contact (usually the indentation corner); 3) median cracks, also parallel to the load axis and in the form of circle or segmented circle. It is believed that median cracks are generated below the plastic deformation zone. 4) half-penny cracks, possibly formed by radial crack propagation downward or median crack growth toward the surface; 5) lateral cracks, parallel to the surface and in the form of a circle. Like median cracks they are also generated beneath the plastic zone. Another two types of crack are also commonly observed though not investigated extensively, i.e., secondary radial cracks and shallow lateral cracks. The particular cracks observed in an indentation depend on the material, indenter and load. Understanding of the cracks generated by indentations comprises understanding of the crack morphology, size and initiation sequence. Both analytical and numerical models have been constructed to give stress field solutions and to predict the cracking events during the loading and unloading process, which are introduced in the following section. Experimental investigations on the Vickers indentation induced residual stresses in alumina ceramics will be covered in section 2.3.

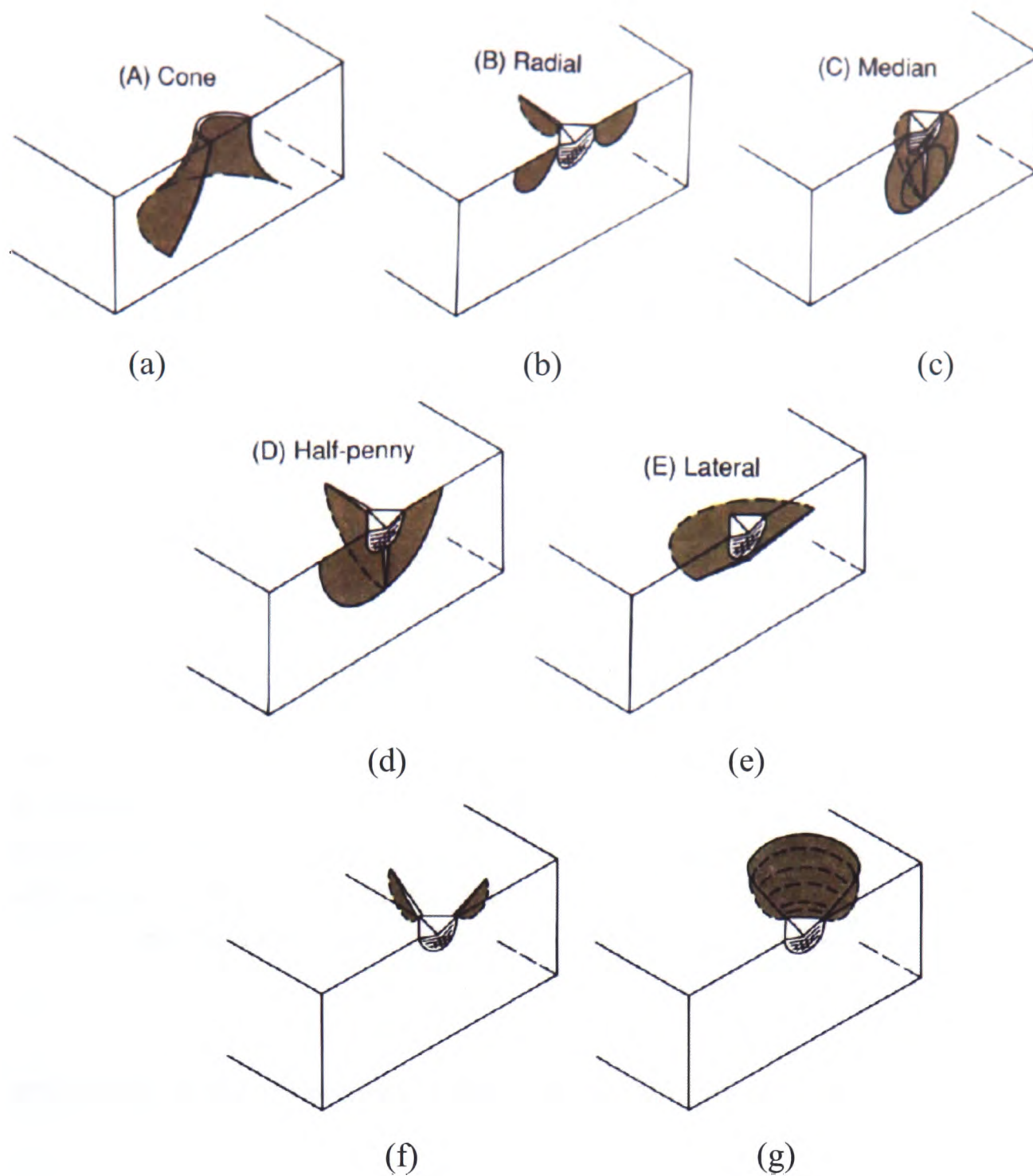


Fig 2.2 Schematic diagram of idealized crack morphologies  
 (a) cone type; (b) radial type; (c) median type; (d) half-penny type  
 (e) lateral type; (f) secondary radial type; (g) shallow lateral type (after [52])

### 2.2.3.1 Vickers Indentation

Vickers indentation is widely used to measure the hardness of materials, including brittle ceramics. The typical evolution of plastic zone and cracking systems under a sharp indenter is shown in Fig 2.3.

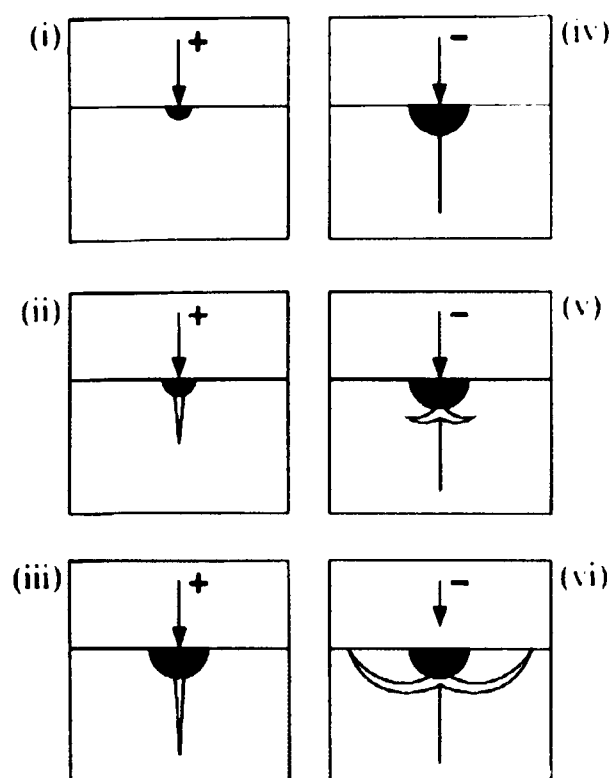


Fig 2.3 Cracking evolution under a sharp indenter during loading (+) and unloading (-) cycle (after [54])

(i) Plastic deformation occurs; (ii) at a critical load a median crack forms underneath the plastic zone, subsurface radial cracks are also formed; (iii) crack propagate downward on increased loading; (iv) the median crack closes up; (v) residual stresses expand radial cracks and initiate lateral cracks near the base of the plastic zone; (vi) cracks continue to expand until load is completely removed.

The stress field around a Vickers indentation has been investigated intensively. Both analytical and numerical solutions will be summarized here.

### 1) Analytical Solution

Ductile behavior is possible for brittle materials when they suffer from a sufficiently high pressure. A plastically deformed zone is expected to form underneath the sharp indenter. Also, due to the high pressure required, the ductility is restricted to a small area of contact with the indenter and the material can be regarded as elastic elsewhere. The classic plasticity theory developed for metals would not apply to brittle materials. The volume of the material cannot always be taken as unchangeable and the elastic



effects become decisive. Yoffe's linear elastic theory is commonly used to predict the stress field around the indentations in brittle materials [55].

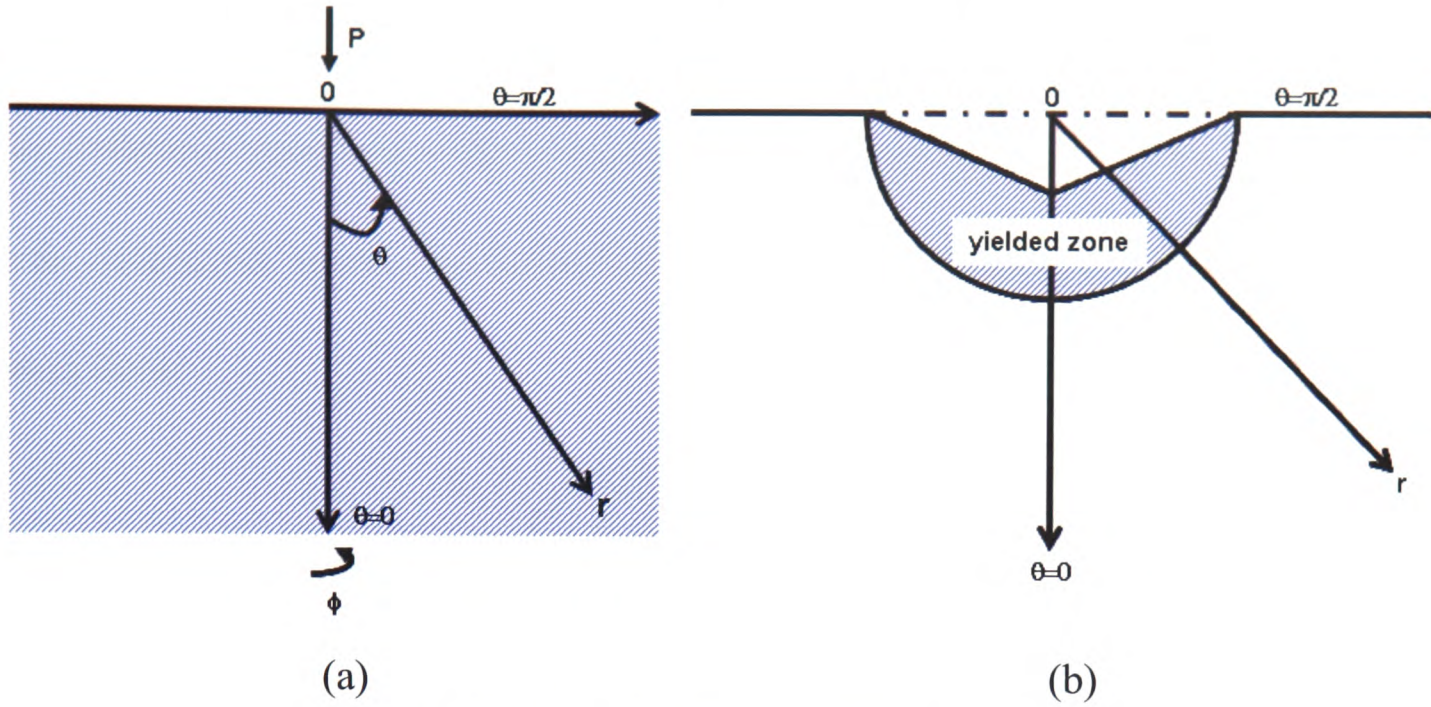


Fig 2.4 (a) Coordinate systems  $(r, \theta, \phi)$ ;  
(b) schematic diagram of yielded zone (after [55])

During indentation the Boussinesq solution describes the basic elastic field of the indenter assuming a point load,  $P$ , on the flat surface of a semi-infinite half-space.

The stress components are given in terms of spherical polar coordinates (Fig 2.4) as:

$$\sigma_r = \frac{P}{2\pi r^2} [1 - 2\nu - 2(2 - \nu) \cos \theta] \quad (2.1 \text{ a})$$

$$\sigma_\theta = \frac{P}{2\pi r^2} \frac{(1 - 2\nu) \cos^2 \theta}{(1 + \cos \theta)} \quad (2.1 \text{ b})$$

$$\sigma_\phi = \frac{P(1 - 2\nu)}{2\pi r^2} \left[ \cos \theta - \frac{1}{1 + \cos \theta} \right] \quad (2.1 \text{ c})$$

$$\tau_{r\theta} = P \frac{(1 - 2\nu) \sin \theta \cos \theta}{2\pi r^2 (1 + \cos \theta)} \quad (2.1 \text{ d})$$

where  $\nu$  is the Poisson ratio.

Two types of maximum tensile stress are predictable from the formula, i.e.,  $\sigma_r$  on

the surface ( $\theta = \pi/2$ ), which may lead to a ring crack; and  $\sigma_r = \sigma_\theta$  on the z axis ( $\theta=0$ ), which leads to median cracks.

The deformed zone below the indenter changes shape and size though still attaching to the half elastic space. It is equivalent to a surface inclusion and causes a blister-type stress field:

$$\sigma_r = \frac{B}{r^3} 4[(5-\nu)\cos^2\theta - (2-\nu)] \quad (2.2 \text{ a})$$

$$\sigma_\theta = \frac{-B}{r^3} 2(1-2\nu)\cos^2\theta \quad (2.2 \text{ b})$$

$$\sigma_\phi = \frac{B}{r^3} 2(1-2\nu)(2-3\cos^2\theta) \quad (2.2 \text{ c})$$

$$\tau_{r\theta} = \frac{B}{r^3} 4(1+\nu)\sin\theta\cos\theta \quad (2.2 \text{ d})$$

where  $B$  is a constant as a measure of the strength of the field.

A combination of the above mentioned stress fields gives the external field (elastic stress field outside the plastic zone) when the indenter is under load, considering continuity across the yielded zone boundary ( $\nu=0.25$  is assumed).

$$\sigma_r = \frac{P}{4\pi r^2} (1-7\cos\theta) + \frac{B}{r^3} (19\cos^2\theta - 7) \quad (2.3 \text{ a})$$

$$\sigma_\theta = \frac{P}{4\pi r^2} \frac{\cos^2\theta}{(1+\cos\theta)} - \frac{B}{r^3} \cos^2\theta \quad (2.3 \text{ b})$$

$$\sigma_\phi = \frac{P}{4\pi r^2} \left(\cos\theta - \frac{1}{1+\cos\theta}\right) + \frac{B}{r^3} (2-3\cos^2\theta) \quad (2.3 \text{ c})$$

$$\tau_{r\theta} = \frac{P}{4\pi r^2} \frac{\sin\theta\cos\theta}{1+\cos\theta} + \frac{B}{r^3} 5\sin\theta\cos\theta \quad (2.3 \text{ d})$$

Deduced from the Yoffe's stress field, the elastic stresses are large when  $r$  is small,



hence the cracks tend to initiate just outside the plastic zone (compressive stress inside). The value of  $B$  decides the position of highest tensile stress.  $B$  is proportional to the increase of volume of the deformed zone.  $B$  is small in a porous material or underneath a large-angled indenter.  $B$  decides the strength of the blister field (residual stress field). It affects the sign of the stress component during the loading process and hence the crack types. The commonly observed indentation induced cracks like ring and cone crack, median cracks, radial cracks, lateral cracks during the loading and unloading phase can be explained using Yoffe's model.

## 2) Numerical Solution

Chiang et al. [53] developed a model extending the spherical cavity solution taking into account the reduced half-space constraint. In their approach a spherical cavity (radius  $a$ ) is under a pressure of  $p$ , which creates a plastic zone (radius  $b$ ). The stress solution must satisfy the boundary conditions, which is a stress-free surface and approximately uniform shear stress within the plastic zone. The stress components are given as:

During loading:

$$\frac{\sigma_r^{pl}}{p} = \left[ \frac{3 \ln(r/a)}{1 + 3 \ln \beta} \right] - 1 \quad (\beta > r/a > 1) \quad (2.4 \text{ a})$$

$$\frac{\sigma_t^{pl}}{p} = \frac{3 [\ln(r/a) + 1/2]}{1 + 3 \ln \beta} - 1 \quad (2.4 \text{ b})$$

$$\frac{\sigma_r^{el}}{p} = \frac{-\beta^3}{(r/a)^3 (1+3\ln \beta)} \quad (2.4 \text{ c})$$

$$\frac{\sigma_t^{el}}{p} = \frac{\beta^3}{2(r/a)^3 (1+3\ln \beta)} \quad (r/a > \beta) \quad (2.4 \text{ d})$$

where  $\sigma_r$  is the radial stress,  $\sigma_t$  is the tangential stress; the superscripts  $pl$  and  $el$  represent plastic and elastic area;  $r$  is the distance from the cavity center.  $\beta$  ( $\beta=b/a$ ) is the relative plastic zone size.

After unloading:

$$\frac{\sigma_r^{rpl}}{p} = \left[ \frac{3\ln(r/a)}{1+3\ln \beta} \right] - 1 + \frac{1}{(r/a)^3} \quad (\beta > r/a > 1) \quad (2.5 \text{ a})$$

$$\frac{\sigma_t^{rpl}}{p} = \frac{3[\ln(r/a) + 1/2]}{1+3\ln \beta} - 1 - \frac{1}{2(r/a)^3} \quad (2.5 \text{ b})$$

$$\frac{\sigma_r^{rel}}{p} = \frac{1}{(r/a)^3} \left[ 1 - \frac{\beta^3}{1+3\ln \beta} \right] \quad (2.5 \text{ c})$$

$$\frac{\sigma_t^{rel}}{p} = \frac{1}{2(r/a)^3} \left[ \frac{\beta^3}{1+3\ln \beta} - 1 \right] \quad (r/a > \beta) \quad (2.5 \text{ d})$$

where  $\sigma^r$  refers to the residual stress.

The conclusions deduced from Chiang's field are the existence of the peak tension at the elastic/plastic boundary, rapidly transiting into compression within the plastic zone. The cracks usually initiate in the vicinity of the elastic/plastic boundary and terminate in the elastic zone after some extension. Note that both Yoffe's and Chiang's models ignore the stress modification caused by the cracking. Very extensive and complicated numerical integration is required in Chiang's model, as a result it is less

widely used compared to Yoffe's analytical model. For the same reason, it is also difficult to quantitatively compare the stress field predicted by Yoffe and Chiang.

The finite element method (FEM) has also been proposed to predict the stress field around indentations in brittle materials. FEM has the advantage of solving complex problems, but it is also more demanding on computation and less generally informative of situations other than those modeled. For example, Zhang and Subhashi [56] proposed an elastic-plastic-cracking model to analyze the indentation cracking in the brittle materials. This model assumes tensile cracking and compressive yielding behavior of brittle material and is claimed to be capable of predicting the development of median cracks during the loading phase and the development of lateral cracks during the unloading phase. Plasticity was assumed and the brittle material can yield in the loading phase, generating the irreversible plastic deformation zone below the surface. It is the tensile stress induced by the mismatch between the elastic and plastic deformation that leads to the formation of lateral cracks during the unloading phase. The size of the damage zone was found to correlate linearly with a non-dimensional brittleness parameter,  $(EY / \sigma_f^2)^{1/3}$ , where  $E$  is the Young's modulus,  $Y$  is the yield stress and  $\sigma_f$  is the fracture stress. Niezgoda et al. [57] applied FEM to simulate the microindentation process in alumina, based on elasticity theory. They showed that the maximum stresses are in the upper region of the corner made by the Vickers indenter, under the force point and in the middle of the symmetric plane. More reports on using FEM to model the stress field around indentations can be found in the literature

[58-62]. They basically focus on the stress field development and the crack initiation but not on the crack evolution during the indentation process.

### 2.2.3.2 Hertzian Indentation

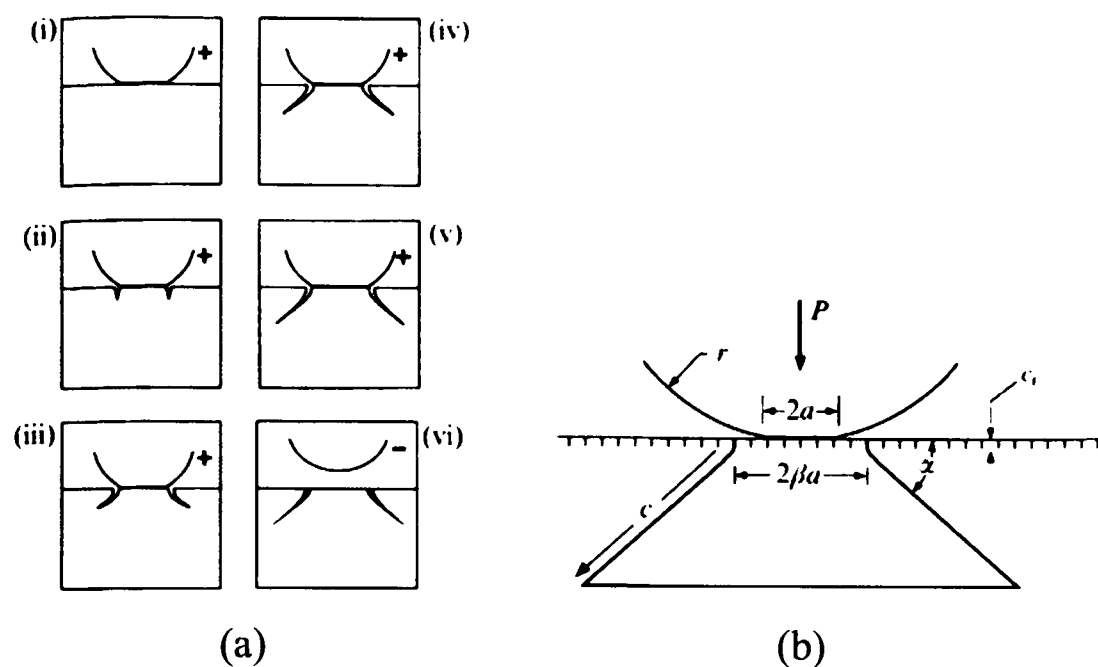


Fig 2.5 Hertzian cone crack system (after [54] )

(a) Evolution of cone cracks during loading (+) and unloading (-) cycle. (i) Surface flaws subjecting to tensile stresses outside contact zone; (ii) Formation of “ring” crack; (iii) ring “crack” grows downward upon further loading; (iv) “ring” crack becomes unstable and forms cone crack; (v) cone continues to grow at further loading; (vi) cone crack closes on unloading.

(b) Indentation Geometry.

The basic idea of Hertzian indentation is to use a hard sphere loaded onto a surface. The Hertzian contact is purely elastic under small and medium loads. The cracking system for Hertzian indentation is shown in Fig 2.5. Under a sufficiently high load, pre-existing cracks propagate to form a ring type crack and may develop into a cone upon further loading [54]. The stress field around Hertzian indentation was investigated by Roberts et al. [63]: the tensile radial stress is close to the surface; this stress decreases sharply with distance from the contact center and also decreases

with depth, finally becoming compressive. Hertzian indentation can be used to measure fracture toughness [64] and also surface residual stress [63].

Wu et al. [10] used Hertzian indentation to measure the residual stresses in machined alumina and 5 vol.%  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites using the relationship between stress intensity, residual stress and the depth of deformation. The results were sensitive to the depth of damage measured from the TEM micrograph, and the compressive stresses were determined to be about 1.5 GPa in the surface for both alumina and nanocomposites, although the depth of the stressed layer in nanocomposite is thicker (10  $\mu\text{m}$ ) than that in alumina (<3  $\mu\text{m}$ ).

## **2.3 Residual Stresses Measurement**

In this section, a brief review of residual stresses is given, including the definition of residual stress and the main measurement techniques, with emphasis on the  $\text{Cr}^{3+}$  fluorescence microscopy method which is being used in this research.

### **2.3.1 Definition of Residual Stresses**

Withers et al. [65, 66] reviewed the origin and nature of residual stresses and pointed out that all residual stresses are the results of misfit. They defined residual stress as stress “which remains in a body that is stationary and at equilibrium with its surroundings”. The misfit leading to the residual stresses can be of mechanical, thermal or plastic origin or come from phase transformation, and it occurs between

different phases, or different parts of the material that have different mechanical or thermal properties. In terms of the misfit regions, residual stresses can be classified into three types, namely, type I: long distance scale; type II: grain scale; type III: near-atomic scale. In reality stress is a complicated tensor field which cannot be measured at a point, and instead must be measured over a finite sampling volume. For most stress measurement techniques that have been developed so far, the sampling volume is larger than the grain size. As a result, it is the type I stress that is normally measured. For polycrystalline alumina materials, type II residual stresses always exist and are the main source of the practically measured stress. In the case of cooling down after treatment from high temperature, variation of thermal expansion among different grains due to crystallographic anisotropy leads to variation in constraints, and hence the generation of residual stresses [67]. Understanding of the residual stresses in polycrystalline alumina is of great importance. Sufficient residual stresses across the grain boundary may cause microcracking. They are also believed to affect the crack propagation path, which could lead to R-curve behavior [68].

### **2.3.2 Overview of Residual Stress Measurement Methods**

There are many methods available to measure residual stress (reviewed in [65, 69]), dividing into destructive and non-destructive ones. Destructive methods include the curvature method [70, 71], hole drilling method [72], crack compliance method [73] and contour method [74]. They rely on inferring the original stress from the displacement by partially or completely removing materials to relax stresses.

Non-destructive methods usually measure the stress indirectly by measuring some parameters that are related to the stress. They include the diffraction method (X-ray diffraction [75], neutron diffraction [76] and synchrotron X-ray [69]), magnetic and electrical method [69], ultrasonic method [77], thermoelastic [78] and photoelastic method [79], and piezo-spectroscopic method (optical fluorescence [80, 81], Raman [82] and cathodoluminescence [83]). Except for the piezo-spectroscopic method, all the other methods have the disadvantage of low spatial resolution [69] due to their large sampling volume.

For alumina materials, since doping with trace amounts of chromium ions can lead to strong fluorescence peaks (R lines),  $\text{Cr}^{3+}$  fluorescence microscopy is very suitable to investigate the residual stress induced by indentation, polishing and grinding. The details of  $\text{Cr}^{3+}$  fluorescence microscopy will be introduced in the following section. The cathodoluminescence method, which is capable of measuring stress with nanoscale resolution, is also introduced here. Relevant research results on residual stresses for alumina ceramics obtained by other techniques are also summarized for comparison.

### 2.3.3 X-ray Diffraction

Levin et al. [75] used X-ray diffraction to measure the thermal residual microstresses in  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites with different additions of SiC (10-25 wt. %), combining the peak shift and line broadening information. The average microstress in



the alumina matrix increases with the SiC content, ranging from 200 to 550 MPa (tensile); the average microstress in SiC decreases from 1800 MPa to 1300 MPa (compressive) with increasing SiC content. The results are in good agreement with a theoretical prediction [84]. Only one diffraction peak was used in Levin et al.'s work and hence the crystallographic anisotropy was not considered. As a result, they measured the interphase stress only and did not measure the intergrain stress [76].

Lange et al. [85] used X-ray diffraction technique to measure the diamond grinding (320 grit) induced residual stresses in the polycrystalline alumina ( $\sim 2 \mu\text{m}$ ). They estimated the compressive stresses to be in the range 135 to 170 MPa and the compressive surface layer that was assumed to deform plastically extended  $\leq 15 \mu\text{m}$  in depth.

Tanner et al. [86] investigated the polishing (down to  $1 \mu\text{m}$  diamond slurry) induced surface damage in alumina and 5 vol.%  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites using high-resolution grazing-incidence parallel-beam X-ray powder diffraction and TEM. They found a correlation between the depth of damage observed from TEM, and the depth at which the FWHM (full width at half maximum) of the X-ray Bragg peaks fell to  $1/e$  of the value at the surface. Using the same technique, they estimated that a uniform compressive strain of  $\sim 0.1\%$  (corresponding to approximately 400 MPa) existed close to the 150 diamond grit ground surface of nanocomposite [9]. No such strain was found in ground monolithic alumina or polished surfaces of



nanocomposites.

### **2.3.4 Neutron Diffraction**

Todd et al. [76] used neutron diffraction to measure the thermal stresses in  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites (SiC content ranging from 4 to 35 vol. %). The deeper penetration of neutrons avoids the stress relaxation and grinding stresses on the surface and the results are more representative of the bulk material. Different diffraction peaks were measured to include the crystallographic anisotropy effect. The average stress in alumina matrix is tensile and increases with increasing SiC content (ranging from 80 to 680 MPa), and the compressive stresses in SiC particles approach 2 GPa. These measurements are very close to Levin et al.'s results measured from X-ray diffraction [75].

### **2.3.5 $\text{Cr}^{3+}$ Fluorescence Microscopy**

#### **2.3.5.1 Principle**

Fluorescence is a member of the luminescence family of processes in which molecules emit light from electronically excited states induced by a physical, mechanical or chemical mechanism. Generation of luminescence through excitation of a molecule by ultraviolet or visible light photons is termed photoluminescence. Fluorescence is one of the two categories in photoluminescence. It is the property of some atoms and molecules to absorb light at a particular wavelength and then to emit light of a longer wavelength after an interval termed lifetime. Fluorescence

differentiates with phosphorescence in that the latter has a much longer excited state lifetime.

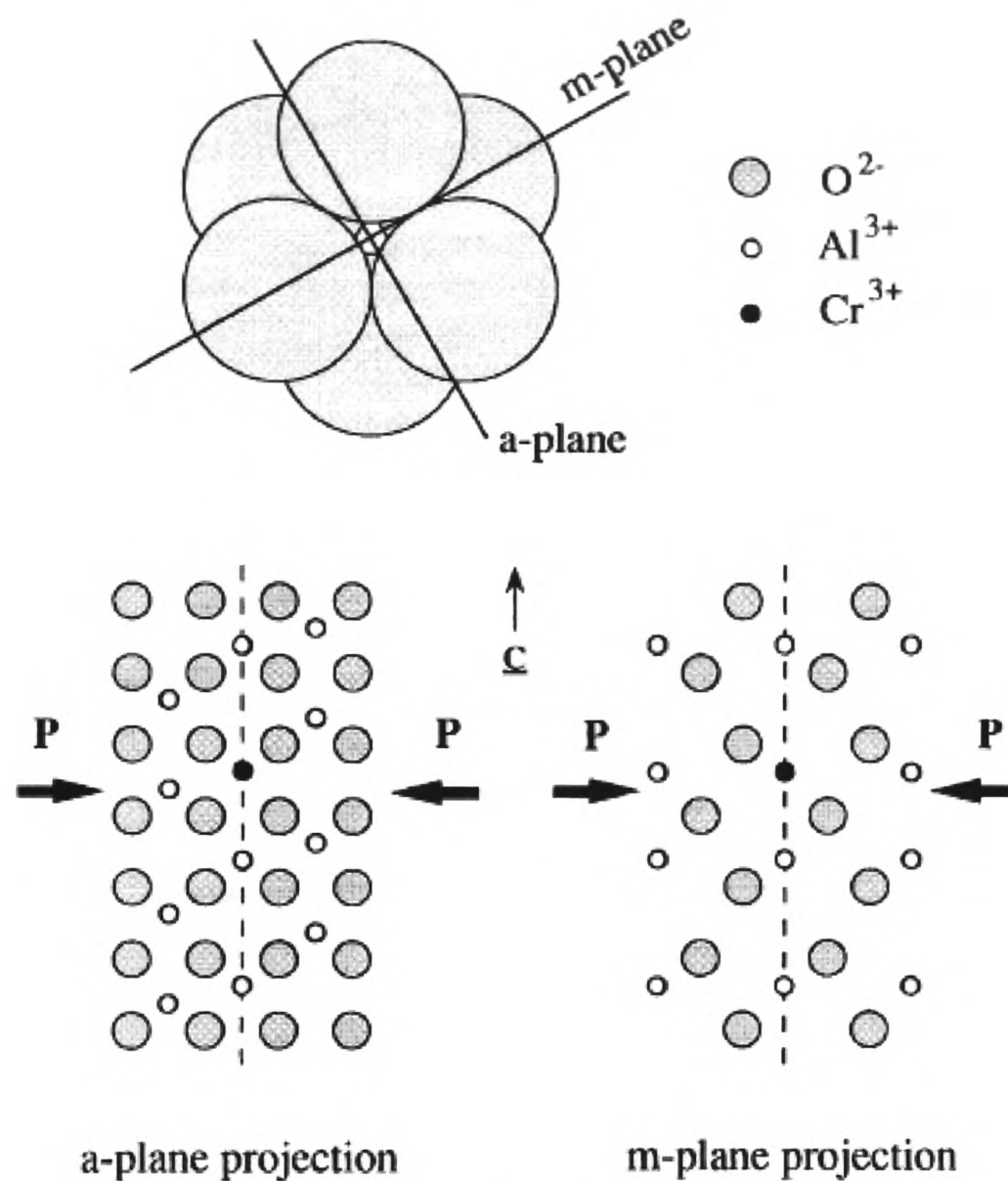


Fig 2.6 Schematic show of the local ionic structure around  $Cr^{3+}$  and the effect of applied stress (after [87])

Chromium in alumina is a chromophore which can give rise to the characteristic red fluorescence of ruby, known as the R-line [88, 89]. The fluorescence comes from electronic transitions in the  $Cr^{3+}$  generated by excitation. Electrons excited from the ground states can lose their energy by radiation.  $Cr^{3+}$  can substitute for  $Al^{3+}$  in alumina and octahedrally coordinate with neighboring  $O^{2-}$ . The octahedron is not regular and is elongated along the  $c$  axis of the lattices. This leads to the splitting of R lines into the doublet of R1 and R2 lines, with wavenumber being around 14402 and 14432

$\text{cm}^{-1}$  [87, 90-92], respectively. Upon applying stress, the crystal field around  $\text{Cr}^{3+}$  is distorted and the potential energy of  $\text{Cr}^{3+}$  changes, and so do the energies of the radiative transitions. The shift of frequency with stress is called the piezo-spectroscopic (PS) effect and is commonly used as a pressure gauge.

The PS effect can be expressed by the tensorial equation [91]:

$$\Delta\nu = \Pi_{ij}\sigma_{ij}^c \quad (2.6)$$

where  $\Pi_{ij}$  are the PS coefficients;  $\Delta\nu$  is the R line shift and  $\sigma_{ij}^c$  are the stresses, defined in the crystal structure coordinates.

More generally, the stresses in the crystal structure are given in terms of the applied stress:

$$\sigma_{ij}^c = a_{im}a_{jn}\sigma_{mn} \quad (2.7)$$

where the matrix  $a_{ij}$  represents the orthogonal transformation between the crystal structure coordinates and the applied stress field,  $\sigma_{ij}$ .

The general relation describing the frequency shift-stress relationship is then expressed as:

$$\Delta\nu = \Pi_{ij}a_{im}a_{jn}\sigma_{mn} \quad (2.8)$$

For single crystal sapphire, the non-diagonal components of the PS coefficients are zero, together with the rotational symmetry of sapphire about the c axis ( $\Pi_{11} = \Pi_{22}$ ), the R line shift is simplified as [91]:

$$\Delta\nu = \Pi_{11}(\sigma_{11} + \sigma_{22} + \sigma_{33}) + (\Pi_{33} - \Pi_{11}) \times (a_{31}^2 \sigma_{11} + a_{32}^2 \sigma_{22} + a_{33}^2 \sigma_{33}) + 2(\Pi_{33} - \Pi_{11}) \times (a_{31}a_{32}\sigma_{12} + a_{31}a_{33}\sigma_{13} + a_{32}a_{33}\sigma_{23}) \quad (2.9)$$

Under a purely hydrostatic pressure, i.e.,  $\sigma_{11} = \sigma_{22} = \sigma_{33} = -P$ , the expression is further simplified as:

$$\Delta\nu = (2\Pi_{11} + \Pi_{33})P \quad (2.10)$$

where  $\Delta\nu$  is in  $\text{cm}^{-1}$  and  $P$  is in GPa.

Apart from shift due to pressure, the R lines also shift with  $\text{Cr}^{3+}$  concentration, which is less extensively investigated. A linear dependence of R lines shift on  $\text{Cr}^{3+}$  concentration is reported [93]:

$$\Delta\nu = 99c_m \quad (2.11)$$

where  $\Delta\nu$  is the measured wavenumber shift in  $\text{cm}^{-1}$ , and  $c_m$  is the  $\text{Cr}^{3+}$  concentration (in wt.%).

Temperature can also affect R-line shift. Both R1 and R2 lines shift to smaller wavenumbers when temperature increases. A change in temperature can cause a line shift of  $\sim -0.14 \text{ cm}^{-1}/^\circ\text{C}$  and the this line shift decreases with increasing temperature [91]. The laser-heating induced temperature increase in alumina-based material is found to be negligible.

#### 2.3.5.2 PS Coefficients Calibration

The precise knowledge of the PS coefficients is of decisive importance to the residual

stress measurement. A great amount of work has been done to determine or calibrate the PS coefficients. The most popular methods to determine the PS coefficients are using a diamond (or a sapphire) anvil cell [94] or a four-point flexure bending configuration [95], by which the sample is under a known stress and the corresponding spectral shift is recorded. The former method cannot give knowledge of PS along a selected orientation, or the individual PS tensor components; while the latter method is not feasible for thin film materials. Recently, biaxial loading methods using jig configurations like ball-on-ring, ring-on-ring and three-ball pressure have been developed for thin samples [96].

He and Clarke [87] evaluated the PS tensors for  $\text{Cr}^{3+}$  doped sapphire using uniaxial compression and shear tests. They measured the frequency shift along the three principal crystallographic and shear directions. They found that the PS coefficient along the  $a$  axis is different to that along the  $m$  axis, which is contrary to the rotational symmetry of sapphire about the  $c$  axis, and is attributed to the breaking of symmetry by the deformation. The difference compared to previous literature measurements can be seen from Table 2.2.

Table 2.2 Summary of PS coefficients for sapphire

| PS coefficients for sapphire (cm <sup>-1</sup> /GPa) |                               |            |                      |            |            |
|------------------------------------------------------|-------------------------------|------------|----------------------|------------|------------|
|                                                      | Previous results <sup>*</sup> |            | He & Clarke's result |            |            |
|                                                      | $\Pi_{11}$                    | $\Pi_{33}$ | $\Pi_{11}$           | $\Pi_{22}$ | $\Pi_{33}$ |
| R1                                                   | 3.0                           | 1.8        | 2.56                 | 3.50       | 1.53       |
|                                                      | 3.2                           | 1.4        |                      |            |            |
|                                                      | 2.7                           | 1.8        |                      |            |            |
| R2                                                   | 2.8                           | 2.3        | 2.65                 | 2.80       | 2.16       |
|                                                      | 2.8                           | 1.9        |                      |            |            |
|                                                      | 2.4                           | 2.2        |                      |            |            |

<sup>\*</sup> Source of the cited PS coefficients can be found in [87]

Thus they drew the conclusion that there exists a significant difference between the PS coefficients of the R1 and R2 line.

$$\begin{aligned}\Delta \nu_{R1} &= 2.56\sigma_{11}+3.50\sigma_{22} +1.53\sigma_{33} \\ \Delta \nu_{R2} &= 2.65\sigma_{11}+2.80\sigma_{22} +2.16\sigma_{33}\end{aligned}$$

(2.12)

They also found that the R1 line shifts nonlinearly with the applied stress along the *a* axis.

Pezzotti developed a procedure to calibrate the PS coefficients using a four-point bending jig [95], as shown in Fig 2.7. The ceramic sample is mounted on the jig and put under the optical microscope. Load is applied and the jig is moved while the

spectrum is recorded. He assumed a uniaxial stress,  $\sigma_{uni}$  is applied to the sample and rewrote the PS effect expression for polycrystalline aluminas as:

$$\langle \sigma_v \rangle = \Pi_{ave} \langle \sigma_{uni} \rangle \quad (2.13)$$

where  $\sigma_{uni}$  is the average of the diagonal PS tensors,  $\Pi_{ave} = (\Pi_{11} + \Pi_{22} + \Pi_{33})/3$ .

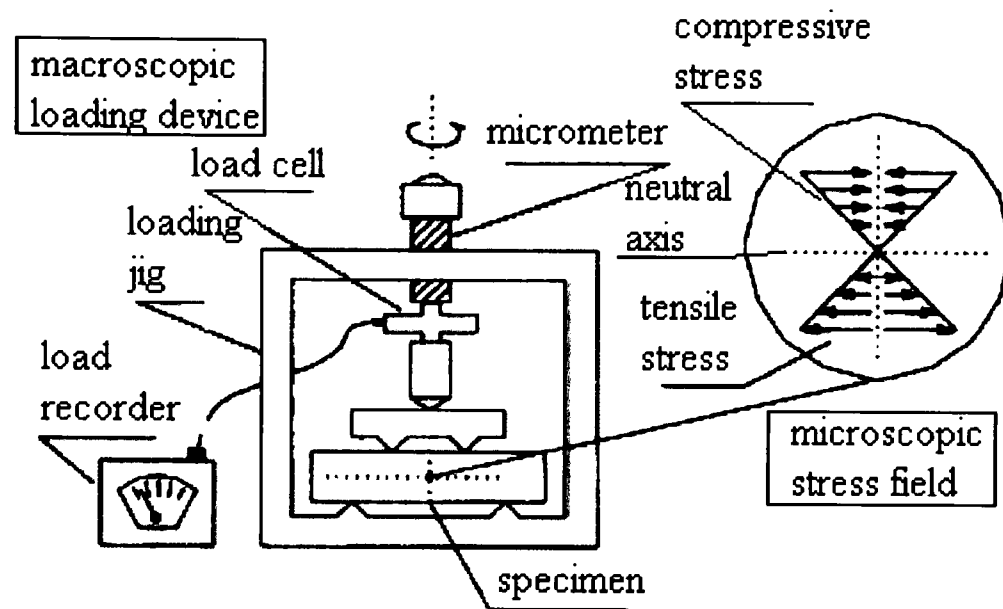


Fig 2.7 Experimental setup for PS coefficients calibration (after [95])

Using the standard four-point bending elastic beam equation, the applied load is converted to stress distribution. Then the recorded spectral shifts of R lines are plotted as a function of the applied stress. The slope of the plot,  $\Pi_{ave}$ , is precisely calculated. The uniaxial PS coefficient  $\Pi_{ave}$  of the R1 band was measured to be in the range between 2.53 and 2.66  $\text{cm}^{-1}/\text{GPa}$ , depending on the grain sizes of polycrystalline alumina [97], which is consistent with those reported using different techniques.

Zhu et al. [98] developed a calibration method to determine the PS coefficients making use of the tensile stress field generated at the indentation crack tips. They rewrote the PS effect as:



$$\Delta \nu = -(2\Pi_{11} + \Pi_{33})p - (1/3)(\Pi_{11} - \Pi_{33})(-\sigma_{11} - \sigma_{22} + 2\sigma_{33}) \quad (2.14)$$

where  $p = -(1/3)(\sigma_{11} + \sigma_{22} + \sigma_{33})$ .

The stress field developed ahead of the crack tip under mode I loading [54] can be given as:

$$\left. \begin{aligned} \sigma_x &= \frac{K_I}{\sqrt{2\pi r}} \cos \frac{\theta}{2} \left[ 1 - \sin \frac{\theta}{2} \sin \frac{3\theta}{2} \right] \\ \sigma_y &= \frac{K_I}{\sqrt{2\pi r}} \cos \frac{\theta}{2} \left[ 1 + \sin \frac{\theta}{2} \sin \frac{3\theta}{2} \right] \\ \tau_{xy} &= \frac{K_I}{\sqrt{2\pi r}} \sin \frac{\theta}{2} \cos \frac{\theta}{2} \cos \frac{3\theta}{2} \end{aligned} \right\} + \text{higher-order terms} \quad (2.15)$$

where  $K_I$  is the stress intensity factor;  $r$  and  $\theta$  are the polar coordinates locating at an arbitrary point ahead of the crack tip, with their origins at the crack tip.

Assuming a surface crack ( $\sigma_{33} = 0$ ) on the  $c$ -plane of a sapphire, the spectral shift on the crack propagation direction just ahead of the crack tip ( $\theta=0$ ) is given as:

$$\Delta \nu(r) = \frac{2K_I \Pi_{11}}{\sqrt{2\pi r}} + \text{higher-order terms} \quad (2.16)$$

Determination of  $\Pi_{11}$  can be directly obtained from the slope of a plot of  $\Delta \nu$  against  $\frac{2K_I}{\sqrt{2\pi r}}$ , provided the crack tip stress intensity factor  $K_I$  is precisely known, which is actually not possible. Their calculated  $\Pi_{11}$  (from crack tip opening displacement measurement) is smaller than that reported by other researchers using macroscopic calibration methods [87, 99].



### 2.3.5.3 Applications in Alumina Ceramics

Ma and Clarke [67] measured the residual stresses in polycrystalline alumina with different grain sizes (2-16  $\mu\text{m}$ ) using  $\text{Cr}^{3+}$  fluorescence. The residual stress is created as a result of the constrained anisotropic thermal contraction along the  $a$ -axis and the  $c$ -axis (thermal expansion coefficients at  $a$ -axis and  $c$ -axis are  $8.3 \times 10^{-6}$  and  $9.0 \times 10^{-6}$   $\text{K}^{-1}$ , respectively), and is tensile along the  $c$ -axis and compressive along the  $a$ -axis. The average residual stresses in the  $c$ -axis were measured to be in the range of 120 to 250 MPa (increasing with grain size) although a variation of about 50 MPa was seen for each grain size. Ortiz Merino and Todd [100] measured the thermal stresses in  $\text{Al}_2\text{O}_3/x$  vol.% SiC ( $x=2,5,10$ ) nanocomposites using  $\text{Cr}^{3+}$  fluorescence microscopy. The tensile stresses in the alumina matrix increased with SiC content from 36 MPa (2 vol.%) to 170 MPa (10 vol.%) and correspondingly the compressive stresses in the SiC particles decreased from 1770 MPa to 1530 MPa (because there is less matrix to constrain them). These results are smaller (by  $\sim 15\%$ ) than those measured by diffraction methods [75, 76].

Molis and Clarke [91] used the  $\text{Cr}^{3+}$  fluorescence to investigate stress inside a 10N loaded Vickers indentation impression and its vicinity for a single crystal sapphire ( $[\bar{1}010]$  oriented). The impression is under compression with the peak compressive stress in the indentation center (-237 MPa at the corner and -370 MPa in the center). At the radial crack tips tensile stresses (93 MPa) are detected. The stress field in the vicinity of the indentation is more complex. Along the distance away from the

indentation where no obvious crack could be observed, compressive stresses decrease approximately exponentially (from -1730 MPa at 12  $\mu\text{m}$  away from the edge of the indentation to -2 MPa at a distance of 157  $\mu\text{m}$ ). However, in regions close to the indentation (outside) where lateral cracks were observed, the stress field is of tension and the tensile stress decreases with the distance from the indentation edge (from 53 MPa on the edge to 7 MPa at a distance of 75  $\mu\text{m}$ ). The lateral cracks were believed to relieve the elastic constraints on the indentation so that the compressive stresses in the impression are small. A uniaxial stress state was assumed for the stress calculation and the results should therefore be regarded as semi-quantitative. The complexity of the fluorescence lines in the close vicinity was addressed in this work in that apart from the line shift and broadening, an apparent shoulder to the R1 line was observed due to the superimposing of different stress states.

Banini et al. [101] used  $\text{Cr}^{3+}$  fluorescence to map (2D) the residual stress around a 25 N loaded Vickers indentation in a ruby crystal. They showed that the largest compressive stress ( $\sim 2$  GPa assuming hydrostatic stress state) is not located in the indentation center but close to the edge. At the tips of the radial and lateral cracks tensile stresses ( $< 0.2$  GPa) were detected. A line scan through the indentation center was also carried out and the maximum compressive stress was 0.8 GPa located at the edge of the indentation. The stress decreases approximately following the  $A/r^3$  relationship ( $r$ : distance from the indentation center;  $A$ : constant), which is in qualitative agreement with Yoffe and Chiang's models [53, 55]. Note that in Yoffe's

model (Equation 2.2),  $A = Bf(\theta)$ ,  $A$  is a constant at fixed  $\theta$  ( $\theta=90^\circ$  when the line scan is carried out on the surface); in Chiang's model (Equation 2.5), the stress dependence on  $1/r^3$  is a constant ( $A$ ) in the elastic zone. However, the value of  $A$  was determined to be  $9.07 \times 10^{-6}$ , smaller than that in Yoffe's ( $A=7.17 \times 10^{-4}$ ) and Chiang's ( $A=1.38 \times 10^{-4}$ ) models. The discrepancy was attributed to the cracking which caused the stress relaxation. In Chapter 7 (section 7.4), experiments results from this work will be compared with Yoffe's model in detail.

Wu et al. [102] used  $\text{Cr}^{3+}$  fluorescence to study stresses around Vickers indentations (0.5-10 N) and single point scratches in alumina and  $\text{Al}_2\text{O}_3/x$  vol. % SiC ( $x=1, 5, 10$ ) nanocomposites. Assuming a biaxial stress state, the maximum compressive stresses measured in the polycrystalline alumina increase with load and reach 460-560 MPa at 10 N. The compressive stresses measured in nanocomposites are much higher than that in alumina and reach a magnitude of 1000-2000 MPa at loads  $\geq 3\text{N}$  with little effect of SiC content. Inside the indent impressions, a larger degree of line broadening was found in the nanocomposites compared to the polycrystalline alumina. They correlated the line broadening with the dislocation distribution and claimed that the lower levels of line shift and broadening in alumina is due to the lower dislocation density after indentation. They observed the line shift showed no clear trend inside the indent impressions but gave no explanation on this. This issue is actually due to the complexity of the stress states and hence the complexity of curve fitting to determine the peak positions close to the indent impression center, which will be emphasized in

## Chapter 7.

**2.3.6 Cathodoluminescence**

The principle of cathodoluminescence (CL) is quite similar to that of photoluminescence (PL), with the difference being that the energy input is an electron beam rather than a visible or ultraviolet light beam [103]. As the electron beam illuminates the sample, the primary electrons lose their energy by creating electron-hole (e-h) pairs. These e-h pairs then recombine through radiative or non-radiative processes, of which the radiative recombination will lead to occurrence of lines on the CL spectrum. The radiative recombination may come from electronic states in the perfect crystal (intrinsic) or from those in the defects or impurities (extrinsic). The defects and impurities that produce extrinsic luminescence are commonly known as luminescence centers, and every luminescence center has a characteristic emission spectrum in the particular host crystal. CL analysis can provide information on crystal structure, chemical composition, defect concentration and internal stress field.

Measurement of residual stress using CL is also based on PS effects, which have been discussed in detail in section 2.3.5. For many sources of CL there exists a linear relationship between CL wavelength  $\Delta\lambda$  and the stress tensor  $\sigma_{ij}$  [104]:

$$\Delta\lambda = \Pi_{ij}\sigma_{ij} \quad (2.17)$$

where  $\Pi_{ij}$  are the PS coefficients, nm/GPa.

By assuming the second-order terms in  $\Pi_{ij}$  are negligible and the material to be isotropic, Equation 2.17 can be simplified as:

$$\Delta\lambda_{av} = \Pi\sigma_{av} \tag{2.18}$$

where  $\Delta\lambda_{av}$  is the average R-line shift,  $\sigma_{av}$  is the average stress and  $\Pi$  is the trace of the PS tensor.

The values of the PS coefficients for alumina materials are summarized in Table 2.3 [104], determined by calibrations with known stress fields for CL bands of several ceramics.

Table 2.3 Experimentally determined PS coefficients [104]

| Materials                                   | details                                                            | $\Pi^*$ (nm/GPa) |           |
|---------------------------------------------|--------------------------------------------------------------------|------------------|-----------|
|                                             |                                                                    | <i>R1</i>        | <i>R2</i> |
| Cr-doped Sapphire                           | a-axis                                                             | -0.157           | -0.140    |
|                                             | m-axis                                                             | -0.101           | -0.096    |
|                                             | c-axis                                                             | -0.0861          | -0.1072   |
| Al <sub>2</sub> O <sub>3</sub> poly-crystal | Averaged experimental results from<br>an area of 5 $\mu\text{m}^2$ | -0.115           | -0.118    |

\* trace of the PS tensor

The advantage of CL over PL is its much higher spatial resolution. However, many factors can affect the spatial resolution of SEM-based CL, including beam-specimen

interaction, signal-noise reduction, carrier redistribution via diffusion and drift, sample geometry, contamination, mechanical vibration, and electronic noise and astigmatism [105]. Todd et al. experimentally measured the spatial resolution of CL to be 550 nm [106], which is in agreement with the prediction from the modified Grün range [107].

Recently Pezzotti [108] used  $F^+$ -center cathodoluminescence, rather than R lines to measure the stress dependence of *c*-plane sapphire.  $F^+$ -center CL emission was reported to have a more intense band than ruby lines by Ostertag et al. [103] in 1991, and this was confirmed in Pezzotti's investigation. Using this intense band makes the spectral collection efficiency higher. The PS coefficient was measured to be  $\Pi = 1.18 \pm 0.03$  nm/GPa, under the biaxial stress field in a ball-on-ring jig.

## 2.4 Spatial Resolution

### 2.4.1 Definition of Resolution

In the optical microscope system, the lens magnifies an object because of its geometric optical property. Due to the wave property of light, the image formed by a point of light is a distribution of points in an area on the image plane, rather than just a single point. The distribution has a higher probability at center and lower probability at the periphery, which can be described by a point spread function (PSF) [109].

Resolution and contrast are two components characterizing image quality. Resolution

is defined as the minimum separation between two objects that can be sufficiently distinguished, or that produces a certain level of contrast [110]. Contrast is defined for two objects with equal (overall) intensity as the difference between their maximum intensity and the minimum intensity that occurs at some distance between them, or is the noise level in the real measurement that limits the ability to use the available resolution [109, 110].

The PSF can be used to quantify the resolution, as two self-luminous points observed under a microscope will appear distinguishable only if they are far enough apart for their PSFs to be distinct, or more specifically, to produce a reasonable dip between the two peaks (Fig 2.7). As can be seen from the definition of resolution, it is quite ambiguous to define the separation distance. When the two peak maxima start to be non-discernible or the contrast becomes zero, the distance between them is defined as the contrast cut-off distance. The Sparrow criterion regards the resolution to be equal to this cut-off distance [110]. The most commonly used Rayleigh criterion, however, requires that the separation to make the two points resolved to be one resel (Fig 2.8) [109], which is the radius of the first dark fringe in the diffraction pattern, or half the Airy disk. The dip is 26.4% of the contrast.

According to the PSF, the radius of the first dark fringe is

$$r_{resel} = 0.61\lambda / NA \quad (2.19)$$

where  $\lambda$  is the wave length;  $NA$  is the numerical aperture.



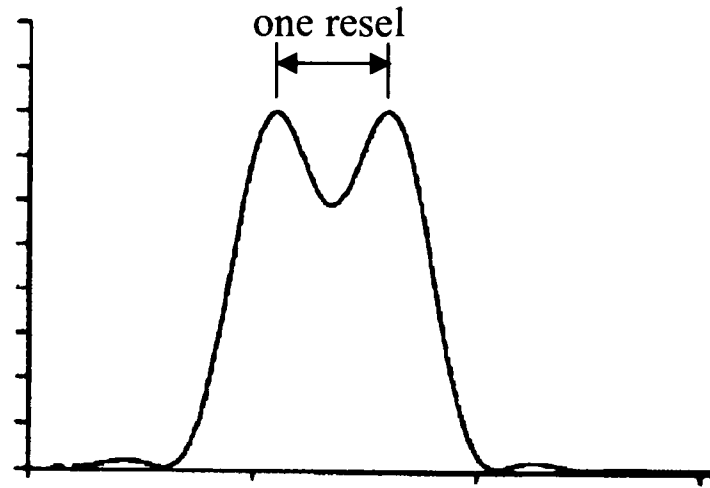


Fig 2.8 Two points separated by one resel (after [109])

Equation 2.19 is also the definition of lateral resolution,  $r_{later}$ . The definition of axial resolution can be deduced from similar reasoning, but is more complicated due to the fact that the axial diffraction pattern of a point source is not disk shaped. It can be estimated by taking the distance from the point of maximum intensity to the first point of minimum intensity of the central bright region along the optic axis [111], which is given by:

$$r_{ax} = 2\lambda n / (NA)^2 \quad (2.20)$$

where  $n$  is the refractive index.

The concept of axial resolution is closely related to depth of field, defined as the distance from the nearest object plane in focus to the farthest object plane that is also in focus. Depth of field is commonly defined as half of the axial resolution:

$$d = \lambda n / (NA)^2 \quad (2.21)$$

Shillaber gave a more precise equation for it in [112] as:



$$d = \frac{\lambda \sqrt{n^2 - (NA)^2}}{(NA)^2} \quad (2.22)$$

However, in some literature axial resolution or depth resolution is connected with depth of focus, which refers to the distance between the farthest and the nearest image plane that are in focus [113, 114].

As can be seen from the definitions in Equations 2.19 and 2.20, the resolutions are decided by the laser wavelength ( $\lambda$ ). The highest lateral resolution for a conventional microscope is about  $\lambda/2$ . This diffraction limit can be overcome by the development of near-field scanning optical microscopy (NSOM) [115]. The basis of this technique is to place an aperture of size smaller than the wavelength of the passing light very close to the sample (less than the wavelength). The spatial resolution of NSOM is not limited by the wavelength, but the size of the aperture. So far the best resolution obtained using visible light is about  $\lambda/40$  (~12 nm) [116], although most reported NSOM work uses an aperture of  $\lambda/3$ - $\lambda/5$  (150-250 nm) considering the sharp decreasing of signal intensity as the aperture decreases.

### 2.4.2 Confocal Microscopy

The basic idea of the confocal approach is to use a spatial filter (aperture, pinhole) to eliminate out-of-focus signal that comes from the specimen, as shown in Fig 2.9. High quality images can be obtained by reducing the unwanted background information.

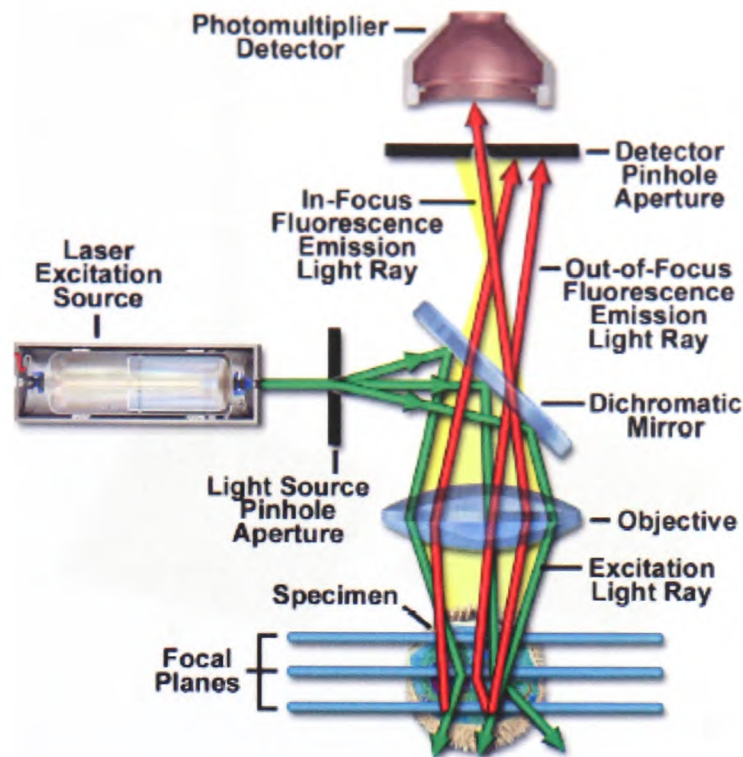


Fig 2.9 Schematic diagram of the optical pathway in confocal microscopy [117]

The confocal volume, within which the object is confocal to the source and the pinhole thus contributing to the detected signal, determines the resolution of the confocal microscope. Since both the illumination volume and the observed volume can be described by the PSF, the volume that is both illuminated and observed can be described using the product of two PSFs [109] assuming identical optics are used for illumination and observation:

$$p_{conf}(\zeta, \rho) = p(\zeta, \rho) \times p(\zeta, \rho) \quad (2.23)$$

where  $p_{conf}(\zeta, \rho)$  and  $p(\zeta, \rho)$  are the PSFs for confocal microscopy and conventional optical microscopy.

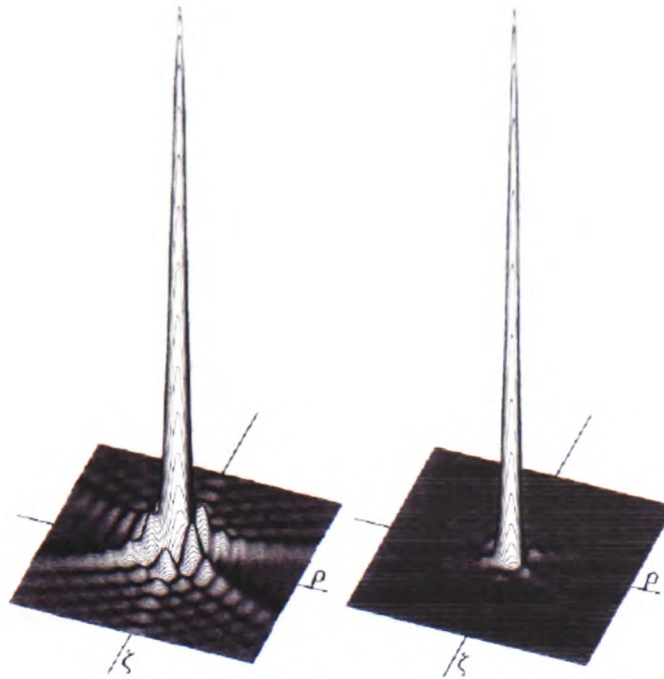


Fig 2.10 The PSF  $p_{conf}(\zeta, \rho) = p(\zeta, \rho) \times p(\zeta, \rho)$  for confocal microscopy (right).

On the left is the PSF  $p(\zeta, \rho)$  for the conventional case (after [109])

The addition of a confocal aperture mathematically means convolution of two functions ( $p(\zeta, \rho)$ ) then multiplying by the aperture (see [109] for more details). The lateral and axial resolution can be similarly deduced from Equation 2.23 [109], assuming a dip of 26.4% of contrast between two adjacent peak maxima.

$$\begin{aligned} r_{later-conf} &= 0.44\lambda / NA \\ r_{ax-conf} &= 1.5n\lambda / (NA)^2 \end{aligned} \tag{2.24}$$

As  $p_{conf}(\zeta, \rho)$  has sharper peaks compared to  $p(\zeta, \rho)$  (see Fig 2.10), it is easy to understand that the resolution of confocal microscopy is better than that of the conventional microscopy.

One important feature of a confocal microscope is the capability to investigate

transparent materials throughout the volume with signal coming just from the area of interest. That is why confocal microscopy is most widely used in the field of biology. For the same reason, in this research confocal microscopy is used to investigate stresses in the transparent sapphire and translucent polycrystalline alumina.

## 2.5 Laser Beam-Specimen Interactions

It has been emphasized in section 2.3.5 that for the  $\text{Cr}^{3+}$  fluorescence microscopy technique, the reliable measurement of residual stress is based on an accurate knowledge of spectral shift. The peak positions are obtained from curve fitting the spectral data, or mathematically describing the line shape. Efforts have been made on modeling the line shape for improving the fitting accuracy [94]. Apart from the peak position and intensity, important information on the physical state of the system (probe size and spatial property gradient of sample) can be acquired from the fitting parameters describing the line shape; inversely, taking the specific system properties into account will help in modeling the spectral line shape [118].

### 2.5.1 Probe Geometry

It is necessary to describe the geometry of the sampling probe before considering the specimen-probe interactions. There are normally two parameters used to characterize the probe geometry: probe size and probe sensitivity [118]. Probe size refers to the effective sample volume that contributes information to the collected spectrum, and probe sensitivity describes how efficiently a volume element within the probe can

convey sample information to the collection optics.

The beam is usually taken to have a Gaussian transverse intensity profile, and the beam radius  $r$  is a function of distance  $z$  from the focal plane:

$$\frac{r_z}{r_0} = \left[ \left( \frac{z}{c} \right)^2 + 1 \right]^{1/2} \quad (2.25)$$

where  $r_0$  is the in-plane focal spot radius,  $r(z=0)$ ;  $c$  is the Rayleigh range that represents a characteristic beam size parameter:

$$\frac{c}{r_0} = \cot \left[ \sin^{-1} \left( \frac{NA}{n} \right) \right] \quad (2.26)$$

Quantification of the probe is complicated by the combining effect of the response of sample to the source beam and collection optics such as slits, apertures and lenses. Normally quantification of the probe makes use of the probe response function, or more specifically, the axial probe response,  $B(z)$ . The probe response function is calibrated by the probe sensitivity function,  $S(z)$ . When the signal origin is limited to a narrow region, which can be regarded as a delta function,  $B(z)$  is proportional to  $S(z)$ . This applies to materials of strong absorption, or opaque materials like silicon. The probe length,  $2p_l$ , is defined as the FWHM (full width at half maximum) of the probe response function (see Fig 2.11 (a)). For transparent materials or materials with negligible absorption like sapphire, the signal origin is uniform. As the probe is scanned along the beam axis, the sensitivity function  $S(z)$  describes the integral of the probe response,  $B(z)$ .  $B(z)$  can hence be recovered by differentiation of  $S(z)$ , and the

probe length is also defined as the FWHM of  $B(z)$ . In practice,  $S(z)$  can be obtained using the depth scan method: the specimen is put on a motorized stage which can move upwards and downwards across the sample surface (laser is focused on the surface, in this case,  $\Delta z$  (as in Fig 2.11)= $z$ ). The spectra collected at different depth are collected and analyzed to generate  $S(z)$ , i.e., the  $I(z) \sim z$  curve, where  $I(z)$  is the intensity at depth  $z$ .

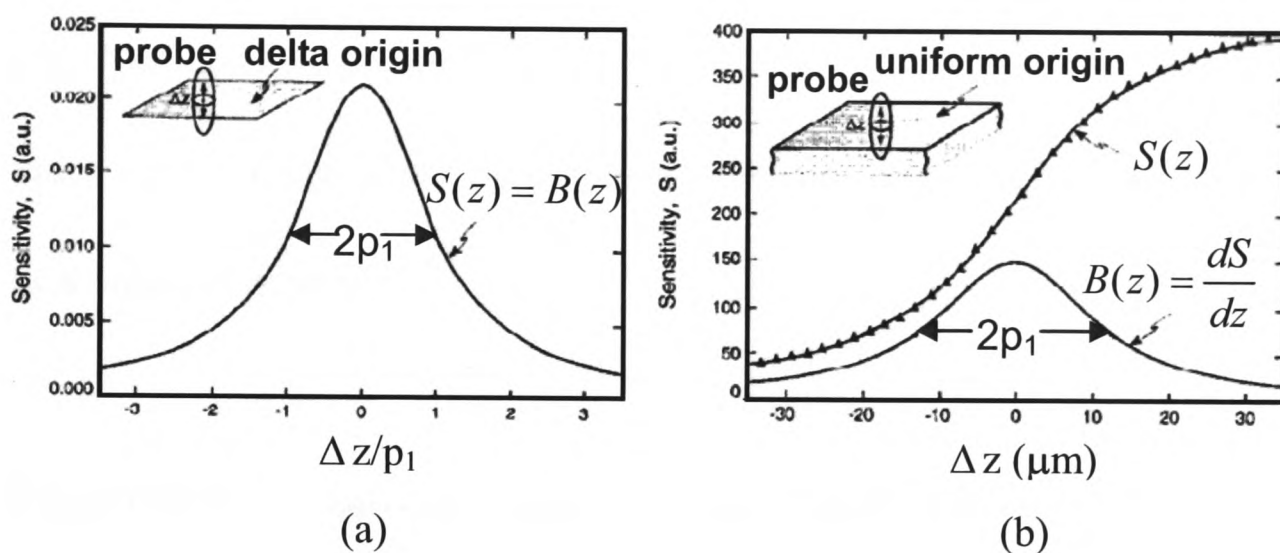


Fig 2.11 Calibration of the axial ( $z$ ) probe response,  $B(z)$ , using (a) infinitesimal signal origin; (b) uniform signal origin (after [118])

### 2.5.2 Probe-Specimen Interaction

Stoneham used a statistical method to calculate the inhomogeneous line broadening [119]. If the effect of spatial property gradient to the line shape is considered, the measured line shape can be expressed as [118]:

$$I(\omega, r_0) \propto \int_{-\infty}^{+\infty} \phi\{\omega - \omega_0[\epsilon(r)]\} B(r - r_0) dr \quad (2.27)$$

where  $\omega$  is the frequency and  $r_0$  is the probe origin position relative to the sample surface.

Details of the equation can be found in [118], but simply put it can be regarded as the

convolution of spatial property distribution  $\Phi(\omega)$  and probe response function  $B(r)$ .

Line shape can be calculated upon solution of the equation above, provided that the information of probe geometry and spatial properties distribution is already known.

### 2.5.3 Modeling Line Shape

The probe-specimen interaction model can be solved using analytical or numerical methods. Numerically, solution to Equation 2.27 will require Fourier transform techniques. By making some assumptions, the model can be simplified and an analytical solution is possible.

Lipkin and Clarke assumed the spatial property distribution to be one-dimensional and bilinear, and both the probe response function and unperturbed line shape [119] are taken to be Lorentzian in shape, Equation 2.27 is then simplified to [118]:

$$I(\omega, z_0) \propto \int_{-\infty}^{+\infty} \frac{dz}{\left[ (z - z_0)^2 + p_1^2 \right] \left[ (\omega - m_i z)^2 + p_2^2 \right]} \quad (2.28)$$

where  $z_0$  is the location of the focal plane;  $p_1$  and  $p_2$  are the FWHM of the probe response function and unperturbed line-shape distributions;  $m_i$  ( $i=1,2$ ) are the slopes of spatial property distribution.

Atkinson et al. [120, 121] took the absorption effect into account when modeling the line shape, and ignored the spatial property variation within the material (this was addressed by Baldwin and Batchelder [122] and other researchers [123, 124]). They gave the laser intensity at peak frequency as:



$$I_{obs}(\Delta\omega_p) \propto \int_0^\infty \exp(-2\alpha z) \frac{p^2}{(z - z_0)^2 + p^2} dz \quad (2.29)$$

where  $\alpha$  is the absorption coefficient,  $p$  is the half probe length.

For sapphire with almost no absorption, Equation 2.29 becomes:

$$\frac{I_{obs}}{I_{obs,max}} = \frac{1}{2} - \frac{1}{\pi} \arctan\left(-\frac{z_0}{p}\right) \quad (2.30)$$

Equation 2.29 is a very simple and clear expression and many further line shape modelings are based on it. However, it actually does not describe the line shape satisfactorily, as can be seen from the verification examples for sapphire, ruby and silicon (especially for sapphire and ruby) in the original paper [120]. The collection efficiency due to the confocal settings is also not included in Equation 2.29.

The advantage of confocal microscopy is its capability to efficiently cut off light scattered from outside the laser focus area, which is especially important for transparent materials since the laser penetration depth in them can be very large. For confocal microscopy the collection efficiency varies according to the optical geometry.

Pezzotti et al. used the concept of collection solid angle,  $\Omega$  [97, 122, 125, 126], describing the inner transmitted zone for the scattered/emitted light, to quantify the collection efficiency as a function of the depth position,  $z$ .



$$\Omega = \begin{cases} 2\pi \left( 1 - \frac{z}{\sqrt{z^2 + \rho_0^2}} \right) (\rho_{\max} \geq \rho_0) \\ 2\pi \left( 1 - \frac{z}{\sqrt{z^2 + \rho_{\max}^2}} \right) (\rho_{\max} \leq \rho_0) \end{cases} \quad (2.31)$$

$$\rho_{\max} = \frac{z}{\sqrt{(n^2 - 1) + \left( \frac{f}{D/2 - \rho_{\max}} \right)^2 n^2}}$$

where  $\rho_{\max}$  is the maximum transverse ray aberration from the optical axis;  $\rho_0$  is the radius of the virtual image of the confocal aperture;  $D/f$  is the diameter/focal length of the objective lens;  $n$  is the refractive index of the sample. A diagram explaining these parameters is given in Fig 2.12.

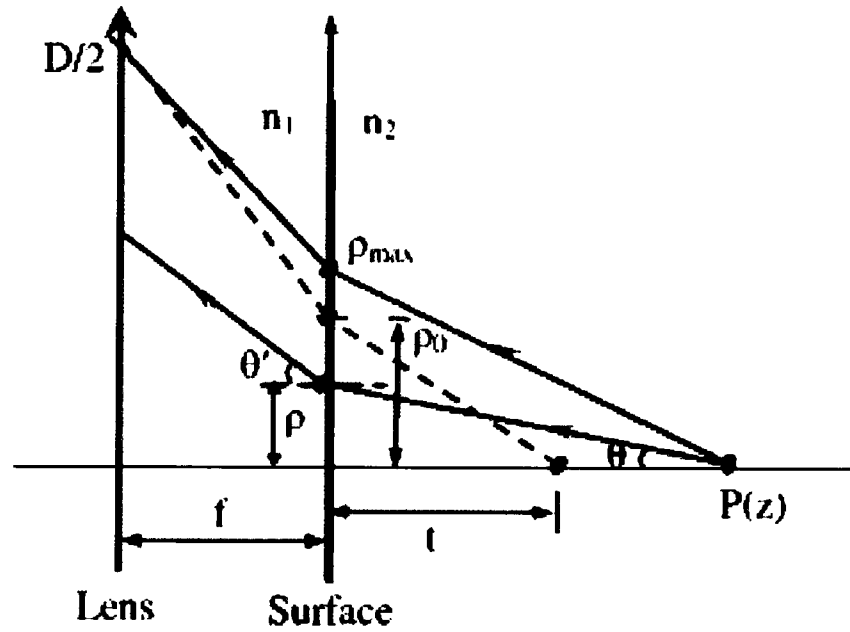


Fig 2.12 Schematic diagram showing the refraction of the rays from the illuminated point  $P$  at depth  $z$  (after [126])

Assuming that the focal plane is placed on the sample surface, the observed intensity in a confocal configuration can be expressed as:

$$I_{obs}(\omega_p) \propto \int_0^z \Omega(\rho_{\max} \leq \rho_0) \exp(-2\alpha z) \frac{p^2}{z^2 + p^2} dz + \int_z^\infty \Omega(\rho_{\max} \geq \rho_0) \exp(-2\alpha z) \frac{p^2}{z^2 + p^2} dz \quad (2.32)$$

where  $t$  is the threshold in-depth below which the collection solid angle  $\Omega$  starts to decrease sharply.

$$t = \rho_0 \sqrt{(n^2 - 1) + \left( \frac{f}{D/2 - \rho_0} \right)^2} n^2 \quad (2.33)$$

For polycrystalline materials, due to the inhomogeneous scattering resulting from grain boundary effects, the collection solid angle will be replaced by the collection cross section [97].

Equation 2.32 relies much on the geometry of the confocal settings and assumes the confocal aperture to be circular. It does not strictly apply when the confocal aperture is in rectangular shape. In addition, it is practically difficult to quantify  $\rho_{\max}$  and  $\rho_0$  in Equation 2.31.

## 2.6 Summary

### 2.6.1 Surface Damage of Alumina-based Ceramics

Grinding/polishing, scratching, and indentation (Vickers and Hertzian) induced surface damage in alumina ceramics have been investigated in the past, with topics on the materials removal mechanism (plastic deformation and brittle fracture), grinding/polishing behavior (wear rate, pullout size and fraction, depth of damage, grain size dependence), deformation mechanisms (twin and dislocation), fracture mechanism (intergranular and intragranular fracture), cracking development (radial crack, lateral crack, median crack or cone crack) and residual stress measurement.

Plastic deformation in alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites have been investigated previously by TEM and it was reported that the depth of dislocations in the ground  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites (3~10  $\mu\text{m}$ ) is much deeper than the depth of twins and dislocations in the ground alumina (<1.0  $\mu\text{m}$ ). However, the density of dislocations varies at different depths and is higher near the surface. In Chapter 5 and 6, the depth of plastic deformation in the ground/polished alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites predicted from a dislocation model will be compared with the TEM results.

### **2.6.2 Residual Stress Measurement Using $\text{Cr}^{3+}$ Fluorescence Microscopy**

Residual stress measurement techniques include TEM, FIB milling, X-ray diffraction, neutron diffraction,  $\text{Cr}^{3+}$  fluorescence microscopy and cathodoluminescence.  $\text{Cr}^{3+}$  fluorescence microscopy has the advantage of high resolution (~2  $\mu\text{m}$  using the confocal settings of the microscope), higher accuracy (relied on the spectra lines exclusively) and being simpler and cheaper. Previous investigations on indentation induced residual stress using  $\text{Cr}^{3+}$  fluorescence microscopy suggested a big difference between alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites. However, the results were obtained using a very bad spatial resolution (>10  $\mu\text{m}$ ). As a result, they were not representative of the surface residual stress since a substantial amount of signal came from far below the surface. In Chapter 7, the real surface stress without being averaged within the sampling volume will be recovered by the convolution method.

Quantitative comparison of the experimentally measured residual stresses around the

indentation to Yoffe's model was rarely seen in the previous investigations. This will be done in Chapter 7 to check the agreement between the experimental results and Yoffe's prediction, and to analyze the reasons behind the discrepancy.

Previous investigations on grinding/polishing or indentation induced residual stresses using  $\text{Cr}^{3+}$  fluorescence microscopy did not look into the curve fitting of spectra lines carefully, especially for spectra lines close to the indentation center or near the ground/polished surface, where asymmetrical broadening was observed. This will be examined carefully in Chapters 5-7.

### **2.6.3 Spatial Resolution**

High resolution is one of the advantages for  $\text{Cr}^{3+}$  fluorescence microscopy, enabling the detection of local stress variation. The principle of how confocal microscopy can improve the spatial resolution was introduced in detail, starting from the concept of resolution and point spread function (PSF).

### **2.6.4 Probe Response Function**

The translucent nature of alumina materials requires the understanding of the probe response function (PRF), or the function to describe the intensity-defocus distance curves. None of the existing models takes into account all the factors including refraction, absorption, scattering, resolution broadening and collection efficiency. A new PRF will be proposed in Chapter 4 that includes all these factors and will be used

to describe the intensity-defocus distance curves for different materials, including single crystal sapphire and ruby, polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites. By convoluting the PRF with the stress function that will be developed in Chapter 5, the predicted volume averaged stresses can be used to compare with the experimentally measured depth profiles through ground and polished polycrystalline aluminas and ground  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites (Chapter 5 and Chapter 6). In Chapter 7, the PRF is convoluted with the stress function based on Yoffe's model to predict the apparent stress distribution around the Vickers indentation.

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## Chapter 3

## Materials Preparation and Experimental Procedures

### 3.1 Materials Preparation

#### 3.1.1 Single Crystal Alumina

Single crystal sapphire and ruby were used in this work mainly for understanding the resolution of the instrument as standard transparent materials. Sapphire was also used to investigate indentation induced residual stress. The surface orientation of the sapphire was (012); the two ruby samples had surface orientations of (0001) and  $(11\bar{2}0)$ . Crystallographic orientations of single crystal sapphire are shown in Fig 3.1. Surface orientation is important for  $\text{Cr}^{3+}$  fluorescence microscopy measurement, as the piezo-spectroscopic coefficient used to convert measured wavenumber shift to stress is dependent on the crystallographic orientation. This will be covered in section 3.5.

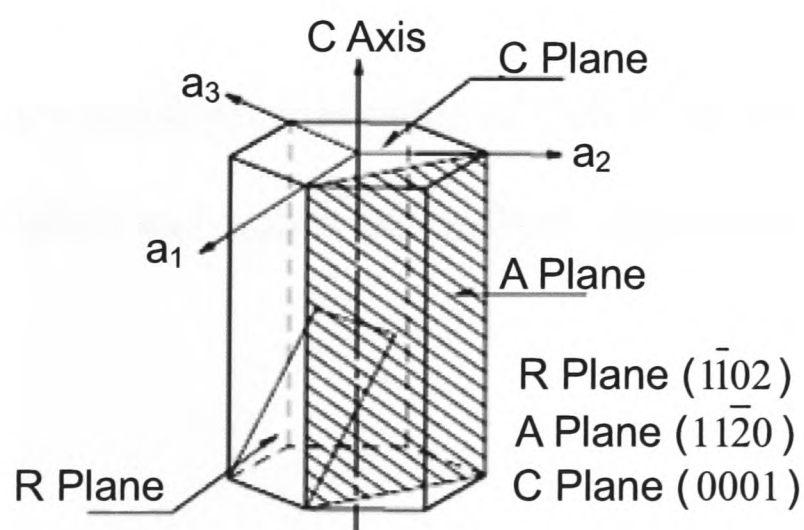


Fig 3.1 Crystallographic orientations in single crystalline alumina [1]



Pieces of commercial sapphire and ruby (one side already polished) were sequentially polished down to  $1/4\ \mu\text{m}$  grit finish. The surface was first lapped using diamond slurry with  $25\ \mu\text{m}$  grit size for 0.5 hour. Then  $6\ \mu\text{m}$  and  $3\ \mu\text{m}$  diamond slurry were used to further polish sample surfaces, both steps lasting for around 1 hour. All lapping and polishing was carried out on a Kent III polishing machine using Kemet plates (Engis Ltd., UK) and an external load of 15 N.  $1\ \mu\text{m}$  polishing was the key step to obtain a smooth surface for single crystal alumina, which was carried out on an ASF soft cloth (Kemet, UK) for at least two hours. Use of  $1/4\ \mu\text{m}$  diamond paste further improved the final polishing. Polished specimens were annealed for 1 hour at  $1550\ ^\circ\text{C}$  in air to remove surface stresses introduced by polishing.

To measure the lateral resolution of the instrument, a piece of polished ruby and another piece of polished sapphire were diffusion bonded by hot pressing. Samples were loaded in a 25 mm graphite die and graphite foils were used to separate samples from spacers. Hot pressing was performed in argon atmosphere with a pressure of 5 MPa and held at a maximum temperature of  $1500\ ^\circ\text{C}$  for 0.5 hour. The surface across the bonded interface was lapped and polished under the same route as introduced above.

### 3.1.2 Polycrystalline Alumina

For polycrystalline specimens, AKP50 (Sumitomo, Japan) alumina powder was used with an average particle size of  $\sim 0.2\ \mu\text{m}$  and a reported chemical purity of 99.995%



$\alpha$ -Al<sub>2</sub>O<sub>3</sub>. The alumina powder was mixed with distilled water (volume ratio of water to powder was 4:1), chromium nitrate (0.05 wt.%), MgO (0.25 wt.%) and dispersing agent (Dispex A40, Allied Colloids, UK; 2.1 wt.%) in an attrition mill (Szegvari 01 HD attritor, USA) with zirconia-lined bucket containing yttria stabilized zirconia (Y-TZP milling media balls with diameter of ~3 mm). After attrition milling for 2 hours at a speed of 300 rpm, the slurry was transferred into a flask and moved immediately to a freeze tank (Edward Shell Freezer, UK) and held there for 2 hours with the flask rotating smoothly. The frozen slurry was dried for 24 hours by subliming frozen water in the vacuum condition (Edwards Micro Modulyo freezing drier, UK). All the dried powders were passed through a 106  $\mu$ m sieve, and then calcined at 600 °C for 1 hour.

The powder was first uniaxially pressed at 50 MPa followed by cold isostatic pressing at 200 MPa to obtain a green compact. Pressureless sintering in air with different parameters was used to produce dense pellets with different microstructures. The heating and cooling rates were program set as 10 °C/min unless otherwise stated.

To make specimens for microstructural observation and fluorescence microscopy measurement, cross-sections of the as-sintered samples were obtained by diamond saw (Capco35, UK) cutting. The surface was sequentially polished (MOTOPOL2000, Buehler) by lapping using 25  $\mu$ m grit diamond paste on an epoxy resin bonded alumina wheel until flat, with a wheel speed of 150 rpm and a force of 10 N per

sample, followed by 6  $\mu\text{m}$  grit polishing on a tin-lead wheel, and then 3  $\mu\text{m}$  on a soft cloth and finally 1  $\mu\text{m}$  on a soft cloth.

In Chapter 6, grinding induced residual stress in polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were compared. To exclude the grain size effect, alumina and nanocomposites of similar grain size were used. For this purpose, a ground 5.6  $\mu\text{m}$  sized polycrystalline specimen was kindly provided by Mr. Apichart Limpichaipanit of the Department of Materials, University of Oxford. It was made by hot pressing at 1700°C for half an hour, and the starting powder was also AKP 50 alumina.

### 3.1.3 $\text{Al}_2\text{O}_3/\text{SiC}$ Nanocomposites

The  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites samples used in this work were kindly provided by Mr. Apichart Limpichaipanit. The detailed powder processing procedure can be found in [2] and [3]. The starting materials were AKP 50 alumina (Sumitomo, Japan, 200 nm) and SiC UF45 (Lonza, Germany, 260 nm). The attrition milling, freeze drying and calcination process were the same as described for preparation for polycrystalline alumina. Dense specimens were fabricated by hot pressing for half an hour at 25 MPa in an argon atmosphere at temperatures between 1400 and 1700 °C. The heating rate was between 30 and 50 °C per minute. The polishing route was the same as that for polycrystalline alumina.

## 3.2 Microstructural Characterization

### 3.2.1 Grain Size Measurement

Polished polycrystalline alumina were thermally etched at 1450 °C for 1 hour in air and Al<sub>2</sub>O<sub>3</sub>/SiC samples were thermally etched at 1450 °C for 30 minutes in vacuum to reveal grain boundaries. The microstructure was then studied by scanning electron microscope (SEM). Samples were gold sputter coated for 90 seconds. Electro-conductive silver was applied to prevent surface charging.

Average grain sizes were measured directly from SEM (JSM 6300 and JSM 840F, JEOL, Japan) images using the linear intercept method [4]:

$$G = 1.56 \frac{L}{MN} \quad (3.1)$$

where  $G$  is the average grain size;  $L$  is total length of the intercepting lines;  $M$  is the magnification of the micrograph;  $N$  is the count number of grain boundary intersections. The number 1.56 is a geometrical factor to convert two dimensional linear intercepts to three dimensional (real) conditions.

At least 200 intercepts were counted for each measurement.

### 3.2.2 Density Measurement

The densities of the materials were measured by the Archimedes method. Samples were put in an oven at 150 °C for 2 hours to give a dry weight,  $W_{\text{dry}}$ . Next they were

immersed in boiling distilled water for 1 hour to fill any open pores with water. Then they were weighed whilst immersed in distilled water ( $W_{wet}$ ). Finally the samples were dried with damp filter paper to remove surface water ( $W_{damp}$ ). The relative sample density  $\rho_R$  is given by:

$$\rho_R = \frac{100}{\rho_T} \left( \frac{W_{dry} \rho_{H_2O}}{W_{damp} - W_{wet}} \right) \quad (3.2)$$

where  $\rho_T$  is theoretical density and  $\rho_{H_2O}$  is the density of distilled water (taken as 3.986 and 1.0 here, respectively). All measurements were repeated at least three times to give an averaged value.

The processing and microstructural information of polycrystalline alumina and  $Al_2O_3/SiC$  nanocomposites that were investigated in this work are summarized in Table 3.1.

### 3.3 Grinding and Polishing

To investigate the residual stress induced by grinding and polishing, polycrystalline alumina was 45  $\mu m$  grit ground on an epoxy resin bonded alumina wheel for 15 minutes, with a wheel speed of 150 rpm and a force of 10 N per sample. After fluorescence microscopy measurements, they were later lapped using 25  $\mu m$  grit diamond on an alumina wheel followed by 6  $\mu m$  grit polishing on a tin-lead wheel and then 3  $\mu m$  on a cloth. The 3  $\mu m$  polished surfaces were also used for residual stress measurements as a contrast to the 45  $\mu m$  diamond ground surfaces.

Table 3.1 Summary of processing and microstructural information of polycrystalline alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites

| Materials                      | Sintering Condition                                             | Average Grain<br>Size (μm) | Relative<br>Density (%) |
|--------------------------------|-----------------------------------------------------------------|----------------------------|-------------------------|
| Al <sub>2</sub> O <sub>3</sub> | ST <sup>1</sup> =1450 °C/1h; PS <sup>2</sup> ;<br>0.25 wt.% MgO | 2.4                        | 98.9                    |
| Al <sub>2</sub> O <sub>3</sub> | ST=1550 °C/1h; PS;<br>0.25 wt.% MgO                             | 4.2                        | 99.5                    |
| Al <sub>2</sub> O <sub>3</sub> | ST=1700 °C/0.5h; HP <sup>3</sup> ;<br>0.25 wt.% MgO             | 5.6                        | >99.9                   |
| Al <sub>2</sub> O <sub>3</sub> | ST=1720 °C/2h; PS;<br>3 °C/min                                  | 8.0                        | 98.4                    |
| 2 vol.% SiC                    | ST=1550 °C/0.5h; HP;<br>0.25 wt.% MgO                           | 6.2                        | 99.8                    |
| 5 vol.% SiC                    | ST=1550 °C/0.5h; HP;<br>0.25 wt.% MgO                           | 5.3                        | 99.6                    |
| 10 vol.% SiC                   | ST=1700 °C/0.5h; HP;<br>0.25 wt.% MgO                           | 4.9                        | 99.8                    |

<sup>1</sup> ST=sintering temperature

<sup>2</sup> PS=pressureless sintering

<sup>3</sup> HP=hot pressing

Grinding of  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites was similar to the grinding of polycrystalline alumina, except the applied force was 22.2 N per sample; the wheel speed was 300 rpm and the duration was 4 minutes. This is in accordance with the requirements for a standard abrasive wear test used previously [3]. For comparison, an alumina specimen with similar grain size was ground under the same condition.

Ground surfaces were examined by SEM (JSM 840 F, JEOL, Japan) and optical microscope (Carl Zeiss, UK).

### **3.4 Indentation**

A microindenter (MH2, Matsuzawa Co., Japan) was used to generate indentations on the surface of materials with a Vickers diamond tip. The indentations were made under a load of 1 kg with 10 seconds of contact time. Micrographs of indentations were taken using an optical microscope (Carl Zeiss, UK).

### **3.5 $\text{Cr}^{3+}$ Fluorescence Microscopy**

#### **3.5.1 Piezo-spectroscopic Effect**

The residual stress was investigated using  $\text{Cr}^{3+}$  fluorescence microscopy, based on the piezo-spectroscopic (PS) effect [5, 6]. The chromium impurity in alumina can give rise to the characteristic ruby lines, R1 and R2. Upon applying stress, R-lines will shift relative to the stress-free position. The shift of frequency with stress is called the piezo-spectroscopic effect. Details of PS effect have already been covered in section

2.3.3 of Chapter 2.

The residual stress is related to the measured R-line shift by the PS coefficients [7]:

$$\Delta \nu = \Pi_{ij} \sigma_{ij}^c$$

(3.3)

where  $\Pi_{ij}$  are the PS coefficients;  $\Delta \nu$  is the R line shift;  $\sigma_{ij}^c$  are the stresses defined in the crystal structure coordinates.

As introduced in Chapter 2, temperature will affect R-line shift. In this work, the room where the Raman microscope (on which fluorescence is activated) is located is kept temperature constant, minimizing the environmental temperature increase.

3.5.2 Renishaw Raman Microscope

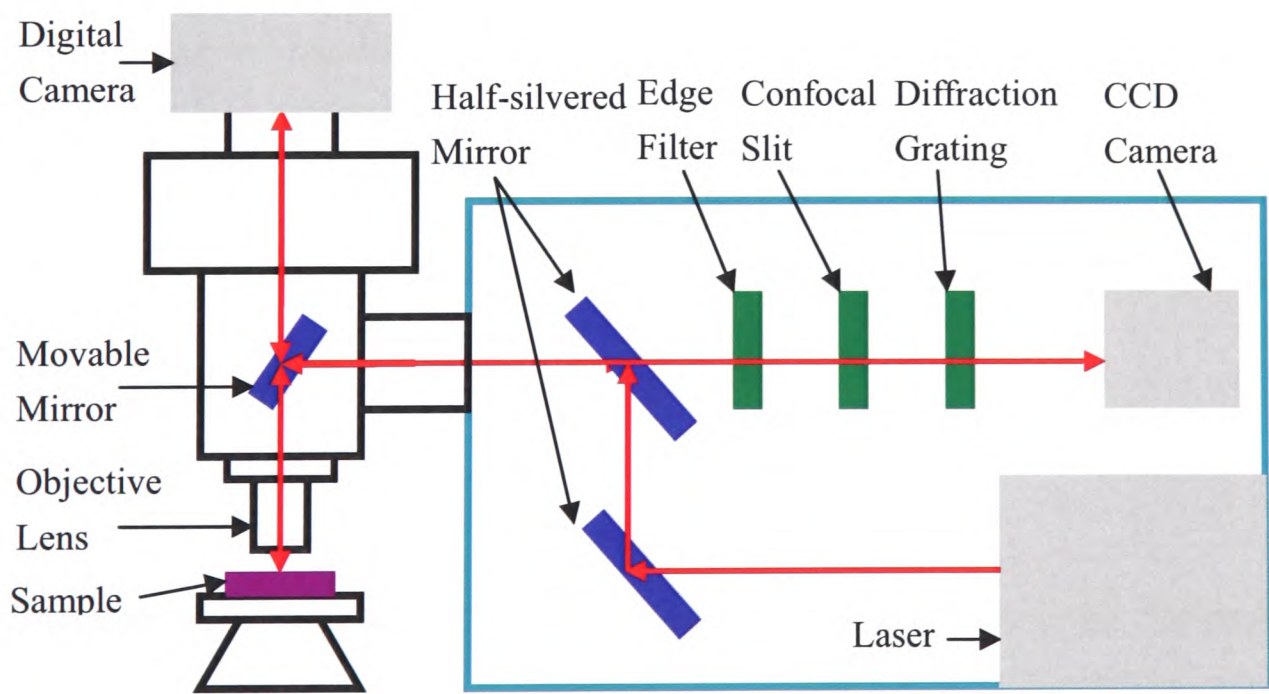


Fig 3.2 Structure of Renishaw Raman Microscope

The spectroscopic measurements were conducted using a Raman microscope (System



1000, Renishaw, UK). The 632.8 nm red line of a 50 mW He-Ne laser was used to excite the fluorescence. Four neutral density filters (NDF) are available to reduce the laser power to 1%, 10%, 25% and 50% of the maximum power. The fluorescent intensity of alumina-based materials depends on the  $\text{Cr}^{3+}$  concentration of the material, and can be adjusted by controlling the laser power (NDF) or the detector time/accumulation number. Higher intensity is generally desirable for signal/noise of the collected spectrum and hence accuracy of curve fitting; however there is always a compromise between stronger intensity and shorter detecting time. There is also an intensity maximum beyond which the CCD will be saturated and drive in a non-linear manner. As a result, the exact experimental settings were case by case.

The microscope is equipped with four objective lenses: 5×, 20×, 50× and 100×. The two commonly used objective lenses, 50× and 100× are of numerical aperture (NA) of 0.75 and 0.90, and working distances of 370  $\mu\text{m}$  and 270  $\mu\text{m}$ , respectively. The working distance determines how far the laser can reach below the surface of the transparent materials.

One important feature of this Raman microscope is its confocal capability. By reducing the size of the confocal slit from 50  $\mu\text{m}$  to 10  $\mu\text{m}$ , most of the out-of-focus signal can be eliminated and the collected signal comes almost exclusively from the focused region. For convenience, the working condition for confocal slit width of 50  $\mu\text{m}$  and 10  $\mu\text{m}$  are called non-confocal mode and confocal mode. Apart from the



confocal slit width, to achieve confocal operation the height of the image area used for capturing grating spectra was reduced from 20 pixels in non-confocal mode to 2 pixels in the confocal mode, and the high-magnification 100× lens is also required. The confocal setting was used for most of the fluorescence microscopy measurements in this work unless otherwise stated.

### **3.5.3 Spectroscopic Measurement**

When the laser is activated and interacts with the sample, the characteristic spectrum from the focused position will be generated and collected by the CCD (charge coupled device) camera. The spectrum is then analyzed using the curve fitting programme (GRAMS/32, Galactic, USA) to get information like peak intensity (area and height), peak position and peak width.

Thanks to the motorized sample stage (Prior Scientific, UK) and the controlling software (GRAMS/32), versatile automatic scanning can be carried out in one dimension (line scan and depth scan), two dimensions (area mapping) and three dimensions (volume mapping).

(a) Line Scan (x&y scan): by assigning the starting and ending position, the laser will scan the set line with the chosen step size. Spectra are collected at each point by the CCD camera and can be analyzed later by the curve fitting programme. Line scans were used in measuring the lateral resolution, and the stress distribution on the surface

of an indented material, normally a line normal to the edge of the indent and passing through the indent center.

(b) Depth Scan (z scan): also one dimensional scan but in the depth direction. The laser is focused on the surface of the measured sample at the beginning and the z position is set to zero. When the laser is focused below surface (into the specimen), the sign of z is positive,  $z > 0$ ; when the laser is focused above surface (in the air) the sign of z is negative,  $z < 0$ . Depths scan were used to detect stress in the ground/polished surfaces and also in the elastically deformed regions around the indentation.

(c) Area mapping (x&y scan): basically is a series of x scans on different y scales (raster pattern or bi-directional pattern). Area mapping was used to map the stress around indent prints and found out to be a powerful tool to relate stress distribution to microstructure such as crack tips.

(d) Volume mapping: series of area maps on different z scales. Volume mapping was used to detect the stress variation at different depths around indentations and collect information such as the size of the plastic zone. However, it takes a very long time to generate a volume mapping and its application is limited.

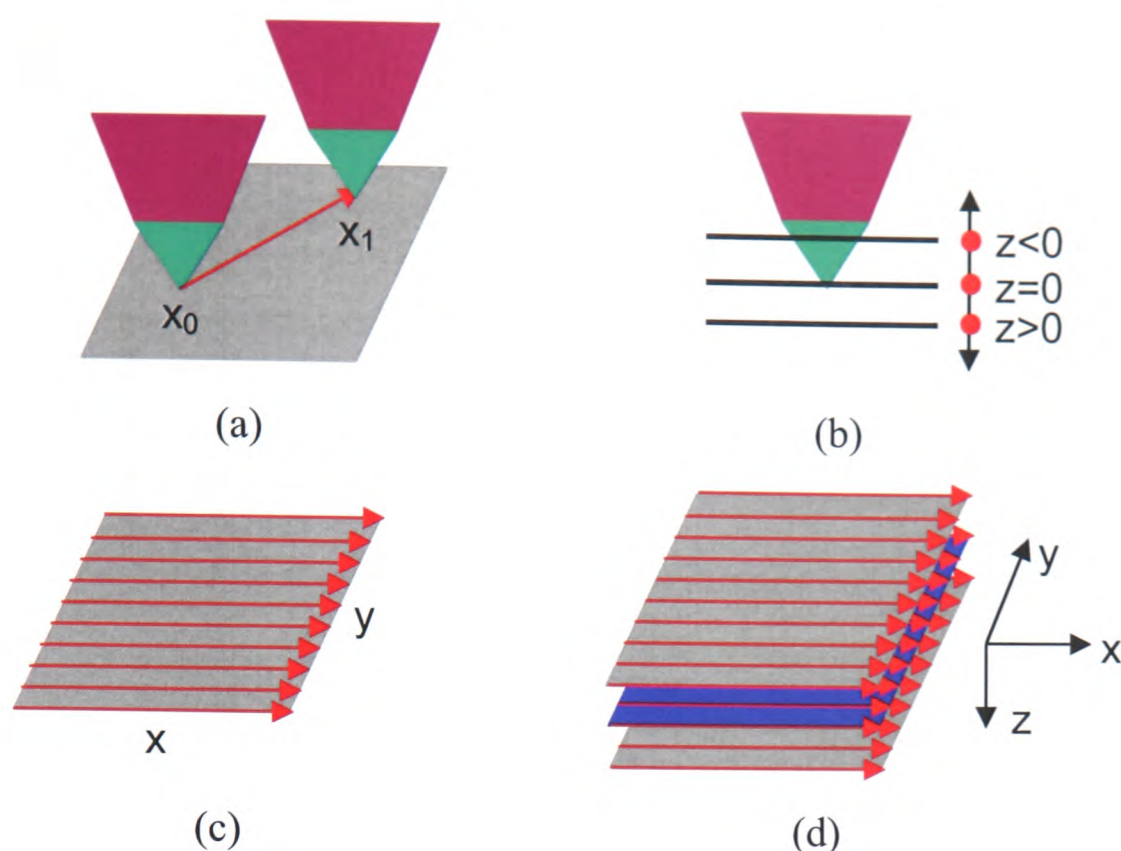


Fig 3.3 Schematic of (a) line scan; (b) depth scan; (c) area mapping and (d) volume mapping

### 3.5.4 Curve fitting

Curve fitting is of significant importance as the sign and the magnitude of stress depend exclusively on the shift of peak positions. The accuracy of the peak positions depends on the curve fitting, or the fitting functions.

When collecting the fluorescence spectrum from alumina-based materials, the center position was set to be  $14400\text{ cm}^{-1}$ . The collected spectrum ranges from  $14145$  to  $14671\text{ cm}^{-1}$ . Since only the R1 and R2 peaks are of interest, a region from  $14250$  to  $14550\text{ cm}^{-1}$  was cropped for curve fitting (see Fig 3.4). This will simplify the background treatment and a linear baseline was sufficient.



coefficient of determination,  $R^2$ . It is regarded as a good fitting when  $R^2 > 0.999$ . A good fitting is shown in Fig 3.5, where the fitted curve almost completely overlaps with the spectrum.

In Fig 3.5, only two peaks, R1 and R2 were fitted. However, it was found that within the plastic zones formed by indentations, and in regions near the ground/polished surface, the collected spectra could not be fitted well with two peaks. An example is shown in Fig 3.6.

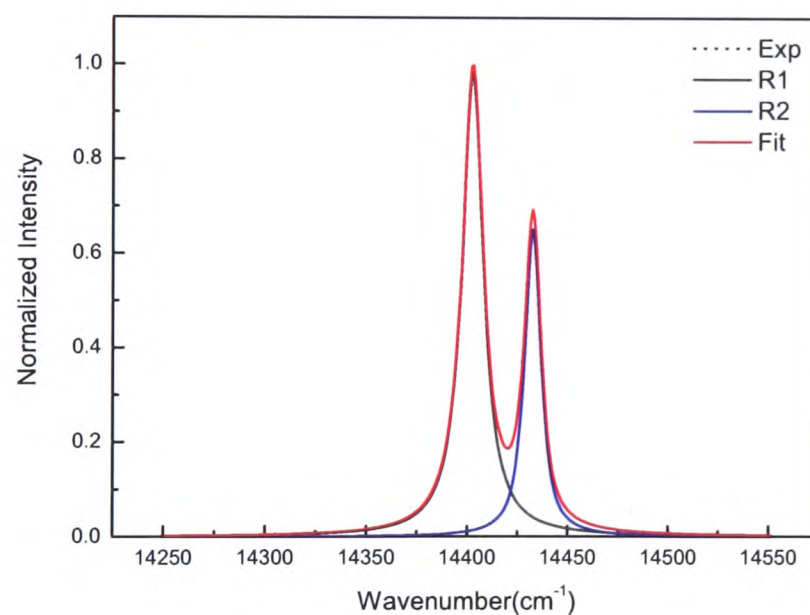


Fig 3.5 An example of a good fitting using mixed Gaussian and Lorentzian function to fit two peaks: R1 and R2

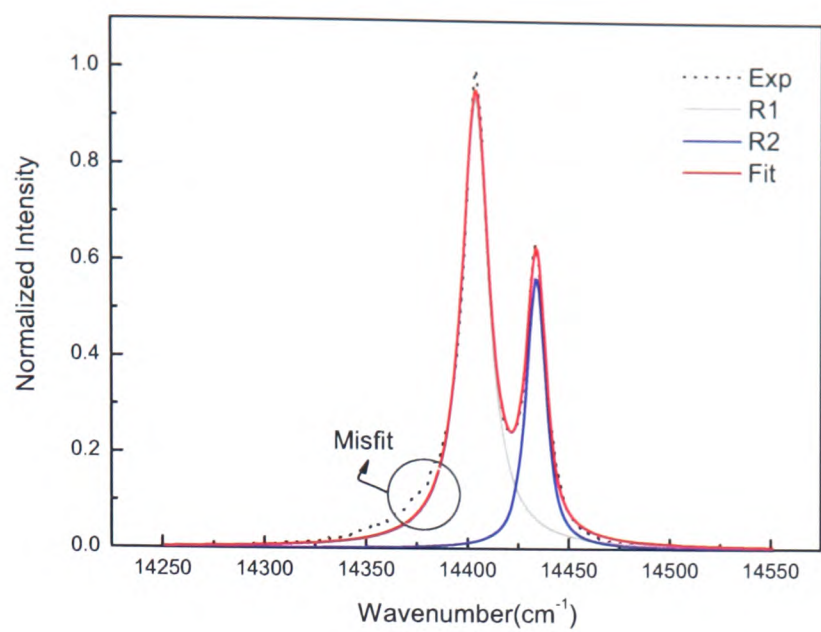


Fig 3.6 An example of an unsuccessful fitting using mixed Gaussian and Lorentzian function to fit two peaks: R1 and R2

It will be argued in Chapter 5 and Chapter 7 that the misfit is due to the tail part of the R1 peak as shown in Fig 3.6. The fitting can be improved by adding another set of R1 and R2 peaks, as seen from Fig 3.7.

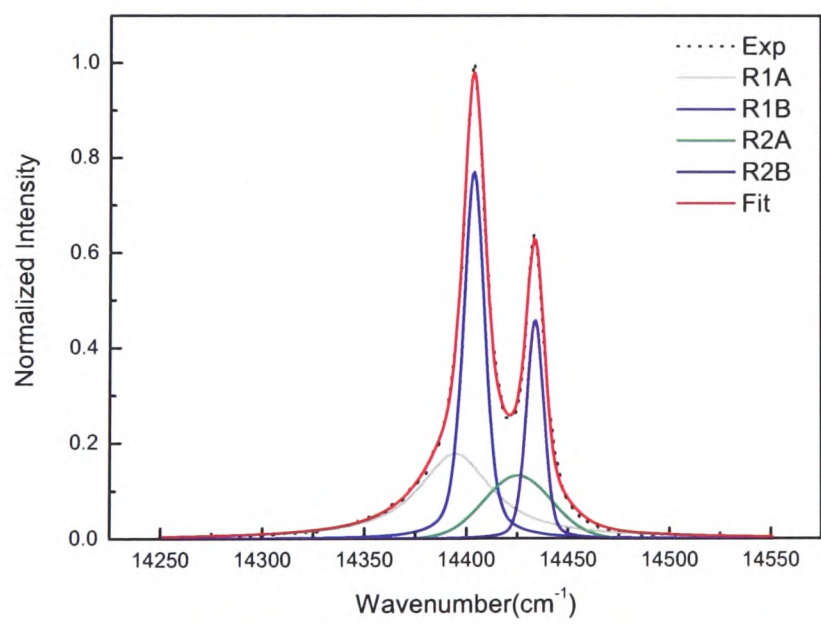


Fig 3.7 An example of a good fitting using mixed Gaussian and Lorentzian function to fit four peaks: two pairs of R1 and R2 peaks. The subscripts A and B represent broad peaks and narrow peak. The spectrum is the same as in Fig 3.6.



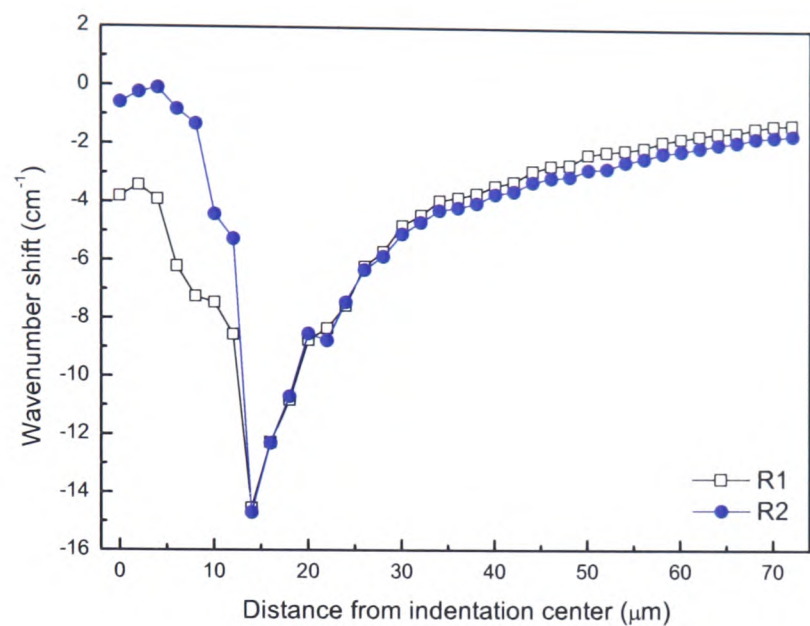
The physical meaning of these two pairs of R-lines will be discussed in detail in Chapter 5 and Chapter 7. Basically they represent two different stress states corresponding to plastically deformed regions and purely elastic regions. For convenience, the fittings using one pair of R-lines and two pairs of R-lines are called two-peak fitting and four-peak fitting from here onwards. For every peak there are 4 parameters to be determined (Equation 3.4), so there are in total 16 parameters to be determined for a four-peak fitting. Considering the broad peak as shown in Fig 3.7 is mathematically ill-defined, it can be imagined that there exist multiple fitting results, i.e, different combinations of the 16 parameters that have similar  $R^2$ . It is argued in Chapter 5 and Chapter 7 that the two sets of peaks (broad peaks and narrow peaks) are physically meaningful. There is a problem using GRAMS/32 to do the four-peak fitting as it cannot set effective constraints on the fitting parameters. The fitting results are good mathematically, but not physically. The fitting constraint issues have been discussed in references [9-11]. Potential constraints include R1/R2 area ratio, R1/R2 width ratio, R1/R2 height ratio, percentage of weight of Lorentzian function and separation of R1 and R2. These constraints are all from experiments and currently lack theoretical support. The other problem is that initial trials showed that when multiple constraints were used, some constraints became fixed to the boundary values set even they were supposed to be variable within a range.

In this work, mainly one constraint was finally used for the four-peak fitting: the area ratio of R1 to R2, for both sets of R-lines. The area ratio is set to a constant rather

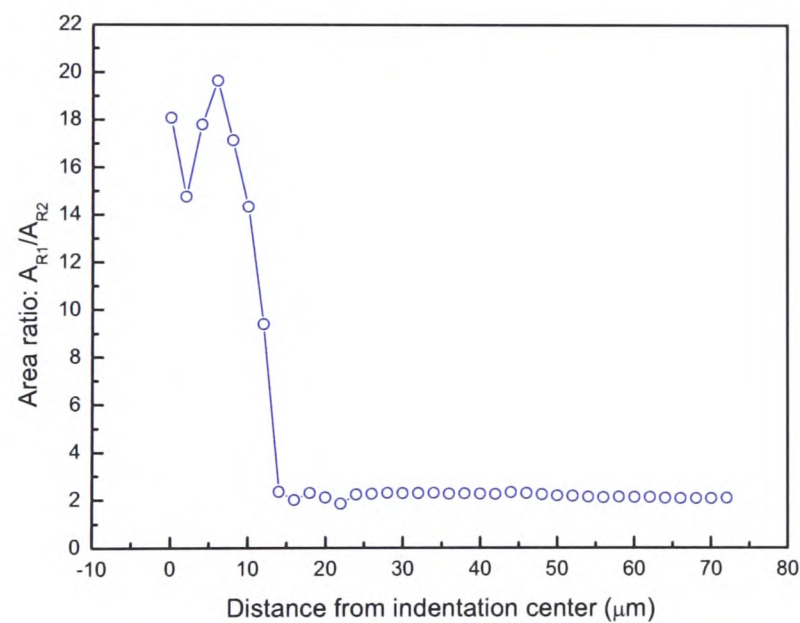
being allowed to vary. There are two reasons for setting this constant area ratio constraint: first, in contrast to the width and separation of R1 and R2, the intensity is almost independent of the stress [12]. Using intensity as a criterion is certainly more justified. According to [13], for single crystal ruby, the area ratio of R1 and R2 is a function of the angle between the  $c$ -axis and the spectrometer direction. The sample is not rotated during an experiment (line scan/depth scan/area mapping), so the angle is fixed and R1/R2 ratio is a constant. For polycrystalline  $\text{Al}_2\text{O}_3$ , the ratio is a function of absorption dichroic ratio or a function of excitation wavelength. The wavelength is constant at 632.8 nm, so the R1/R2 area ratio will also be constant; second, experiments show that area ratios are fairly constant for spectra collected from polished and annealed sapphire, and from the stress-free regions from indented (far away from indent center) or ground/polished material (i.e., far below the surface), and also for spectra collected from stressed regions but could be fitted well by two-peak fitting. An example is given in Fig 3.8, where a two-peak fitting was used to fit the spectra collected from a line scan through the indentation center on a sapphire specimen (details will be described in Chapter 7). The R1/R2 area ratio was almost constant ( $\approx 1.94$ ) at a distance of  $r > 12 \mu\text{m}$  ( $r$ : the distance from the indentation center) even stresses were not zero (non-zero wavenumber shift). For  $r \leq 12 \mu\text{m}$ , the R1/R2 area ratios were much bigger than 1.94 and the wavenumber shifts of R1 and R2 were quite different. The spectra with abnormal R1/R2 area ratios ( $r \leq 12 \mu\text{m}$ ) were then fitted using the four-peak fitting, with the area ratio set to be 1.94 (for both broad peaks and narrow peaks) as the constraint. Note that this area ratio varies between



experiments and between materials, but remains constant within each case. The value used comes from average area ratios of spectra collected from stress free regions of the material used in the individual experiment.



(a)



(b)

Fig 3.8 Wavenumber shifts of R1 and R1 (a) and R1/R2 area ratio (b) for spectra collected from a line scan through the indentation center on a sapphire specimen using the two-peak fitting

Another constraint, the separation of R1 and R2, was also used if it was found not in a reasonable range. Atkinson et al. [10, 11] suggested that the separation of R1 and R2

follows Equation 3.5:

$$c_{R2} - c_{R1} = 29.923 - 0.155\Delta\nu_{R2} \quad (3.5)$$

where  $c_{R1}$  and  $c_{R2}$  are the peak positions of R1 and R2 and  $\Delta\nu_{R2}$  is the R2 peak shift.

The separation increases from 29.9  $\text{cm}^{-1}$  at zero stress to 33.8  $\text{cm}^{-1}$  at a biaxial compressive stress of 5 GPa, and decreases to 28.4  $\text{cm}^{-1}$  at a biaxial tensile stress of 2 GPa [10].

The constrained four-peak fitting was carried out using an alternative fitting programme, Auto2Fit (7-D Software, China) based on the Levenberg-Marquardt algorithm plus its unique UGO (Universal Global Optimization) algorithm [14]. Sufficient numbers of iterations were run to make  $R^2 > 0.999$ . It needs to be pointed out that, even with the constraint, the positions of the broad peaks are still variable on different fitting runs, but given the very large shifts seen in these peaks the errors are still acceptable. The positions of narrow peaks are very reliable and can be reproduced quite well.

In principle, shifts from R1 and R2 peaks can both be used to measure residual stresses. In some references [6, 15], R2 is recommended as the R2 peak is considered to have better linearity. However, the higher intensity and hence the higher signal/noise ratio of R1 peak makes it a better choice to characterize the peak shift in the current work: the accuracy of peak position from curve fitting is higher for R1. In addition, when two stress states are superimposed, the weaker R2 peak, especially the

broad R2 peak, is more liable to be affected by tails of other peaks (refer to Fig 3.7). Therefore, all the peak fittings in the following chapters are based on R1 only unless otherwise stated.

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## Chapter 4

### Understanding the Spatial Resolution

#### 4.1 Introduction

Apart from being cheap and quick, another important advantage of using optical fluorescence to measure residual stress is its higher spatial resolution compared with other techniques [1]. By using a confocal setup to cut off signals from out-of-focus regions, higher resolution can be achieved. According to the manufacturer, the Raman microscope that was used to carry out the current fluorescence microscopy work has the best lateral and axial resolutions of 1  $\mu\text{m}$  and 2  $\mu\text{m}$ , respectively. However, the best resolutions can be only achieved in ideal conditions and they are very sensitive to conditions like laser alignment, vibration and temperature variation, as well as the optical properties of the material being examined. As a result, it is important to know the real resolution when doing the measurements. As introduced in Chapter 2, the spatial resolution of an optical microscope can be experimentally determined by measuring the FWHM (full width at half maximum) of a response curve, i.e., the intensity variation against distance from focus position. It can be done manually by measuring the width of the intensity curve at half of the maximum. Alternatively, it can be obtained from the fitting parameters by using some mathematical functions, like Gaussian, Lorentzian or mixed Gaussian and Lorentzian function, to fit the curve. There is a problem for the latter method in that due to the complexity of the actual intensity curves (see the following sections), they cannot be fitted very well with

simple functions. It might not be a big problem for estimating the FWHM if the misfit happens at the tail of the curve. However, functions that can describe the intensity curves accurately, called the probe response functions (PRFs), are needed to convolute with stress functions to predict the observed stress distribution, which will be covered in Chapter 5-7. PRFs for different alumina based materials, including single crystal sapphire and ruby, polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were developed in this chapter. Information such as resolution and absorption coefficient is included in the PRF. In addition, possible ways to improve the axial resolution were introduced and tried out based on the ray-tracing analysis.

## 4.2 FWHM

In this section, the lateral and axial resolution were measured by manual methods mentioned above, or from rough estimates from fitting parameters using a simple function such as a Gaussian or Lorentzian function to fit the intensity curve. In the end of this section, two special objective lenses, one with a built-in iris diaphragm, one oil immersion lens, were tried out to investigate the possibility of improving axial resolution.

### 4.2.1 Axial Resolution

The axial resolution was obtained by the depth scan method, as is described in section 3.5.3. The laser was made to focus on the sample surface, the motorized sample stage then moved the sample through the focal plane and a series of spectra collected at

different depths were recorded by the CCD camera and then analyzed by the curve fitting software (GRAMS/32), as a function of defocus distance (the distance away from the focal plane, also the distance away from the surface). For an opaque material, the axial resolution (FWHM) can be measured directly from the as-obtained intensity-defocus distance curve; for a transparent material, the FWHM is measured from the differentiated intensity-defocus distance curve (see section 2.5.1 in Chapter 2).

A silicon wafer is often used a standard material to test the resolution. It is an essentially opaque material and has a strong Raman peak centred around  $520\text{ cm}^{-1}$  shift. The axial resolutions were measured under different conditions, i.e., different confocal slit size; high magnification (100×) or low magnification (50×). The intensity-defocus distance curves are shown in Fig 4.1, and the measured FWHMs are given in Table 4.1.



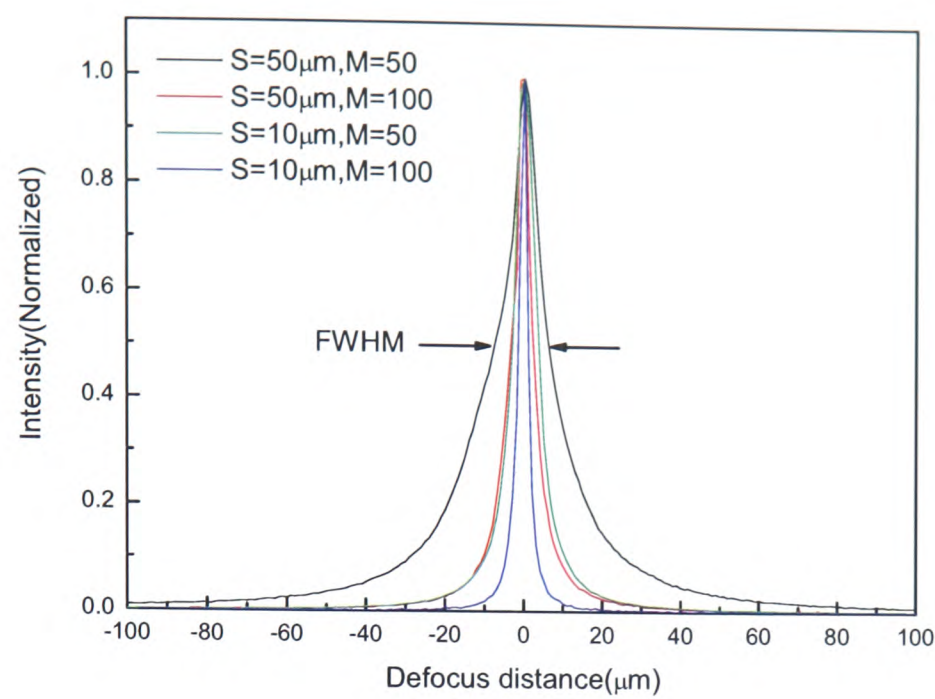


Fig 4.1 Raman intensity-defocus distance curve of Si wafer using different slit width (S) and magnification (M)

Table 4.1 Measured axial resolution from Si under different microscope settings (S: confocal slit width; M: magnification)

| Microscope setting | FWHM (μm) |
|--------------------|-----------|
| S=50 μm; M=50      | 13.8      |
| S=50 μm; M=100     | 5.6       |
| S=10 μm; M=50      | 6.8       |
| S=10 μm; M=100     | 2.6       |

The best axial resolution is 2.6 μm, close to what the manufacturer claims (2 μm), and can only be achieved using the confocal setting and under the highest magnification. This is also the standard setting used when measuring the residual stresses induced by polishing and grinding (Chapter 5 and 6) or indentation (Chapter 7), unless stated



otherwise. The FWHM was measured manually, as the seemingly simple curve cannot be fitted well using Gaussian, Lorentzian or mixed Gaussian and Lorentzian function. An example is given in Fig 4.2 where a Lorentzian function was used to fit the intensity-defocus distance curve obtained under the optimal microscope setting. Although the fitting is not very good above the surface, it will not significantly affect the FWHM. The fitted FWHM is  $2.5\ \mu\text{m}$ , very close to the manually measured one.

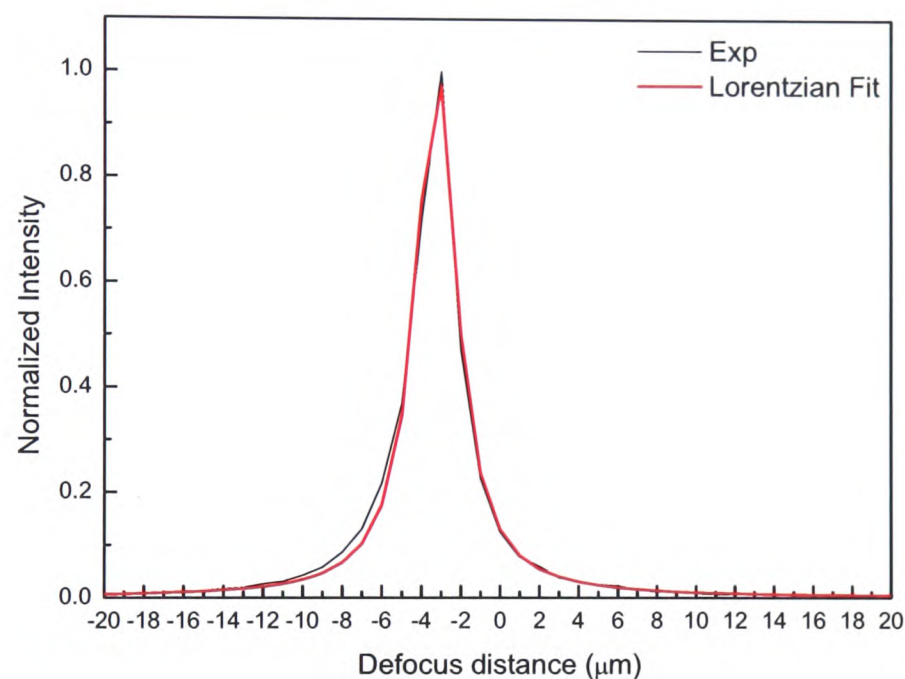


Fig 4.2 A Lorentzian fit to the intensity-defocus distance curve of Si ( $S=10\ \mu\text{m}$ ;  $M=100$ )

In principle, the best resolution is an instrumental parameter and is independent of the material. However, the measurement method of the FWHM might introduce some difference to the obtained resolution from different materials. The relevant response curve for transparent materials like sapphire and ruby is not a simple intensity-defocus distance curve; instead, it will be the differentiated one [2], as explained in section 2.5 of Chapter 2. If the transparent sample is thin enough, it is possible for the laser to

reach the bottom surface of the material. It would be of interest to know how much the resolution deteriorates when the laser reaches the bottom surface. The potential meaning of this resolution is: if the resolution at the bottom surface is good enough, it will be possible to measure the indentation induced stresses from the opposite side from that where the indentations are made (see Fig 4.3). This can potentially avoid the complicated plastic stress field and refraction due to the cracks and the sloping indentation surfaces when doing a depth scan through the indentation center.

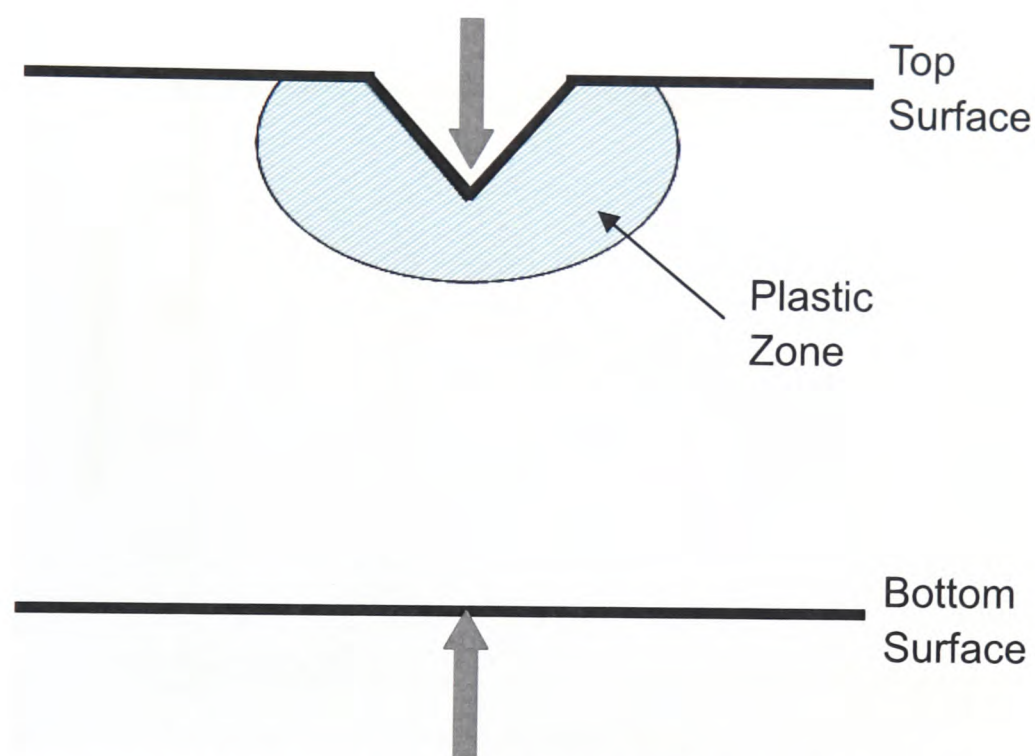
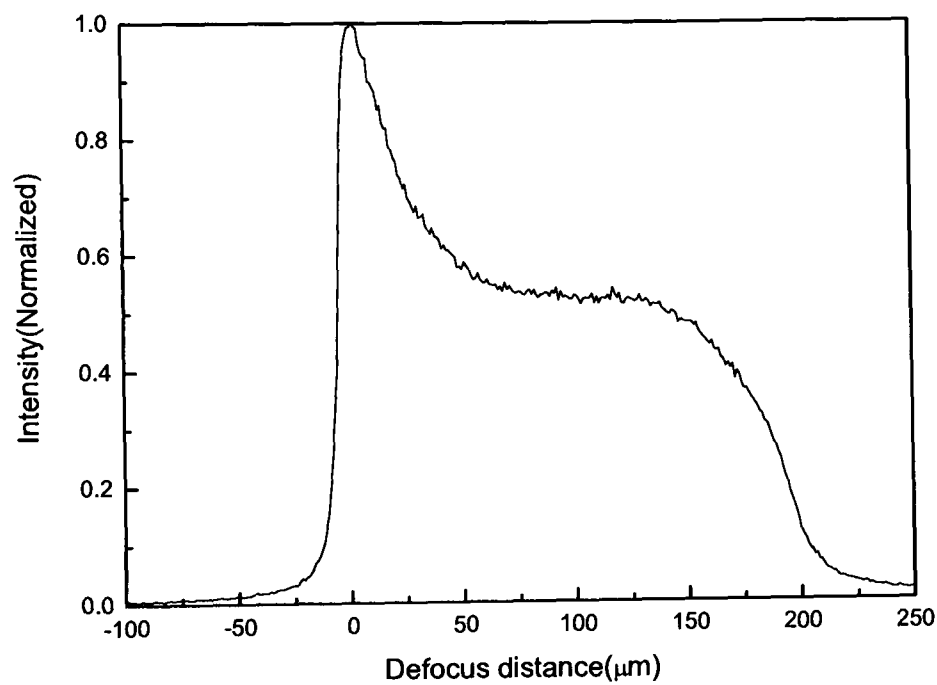


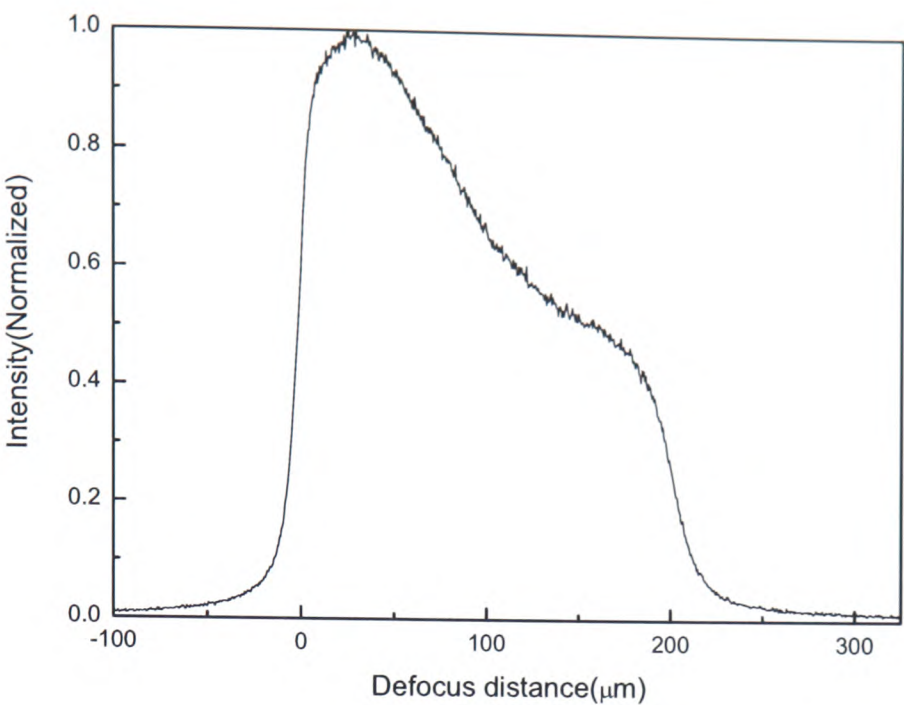
Fig 4.3 Schematic of depth scan through indent center from either top surface or bottom surface of a transparent material

Intensity-defocus distance curves from a 400  $\mu\text{m}$  thick sapphire (both sides polished down to 1  $\mu\text{m}$  grit) are shown in Fig 4.4, using both 100 $\times$  and 50 $\times$  lenses. Because of the refraction ( $n \approx 1.8$ , see Fig 4.9), the laser reaches the bottom surface when the apparent stage movement is about 220  $\mu\text{m}$ . For simplicity, only results from the R1

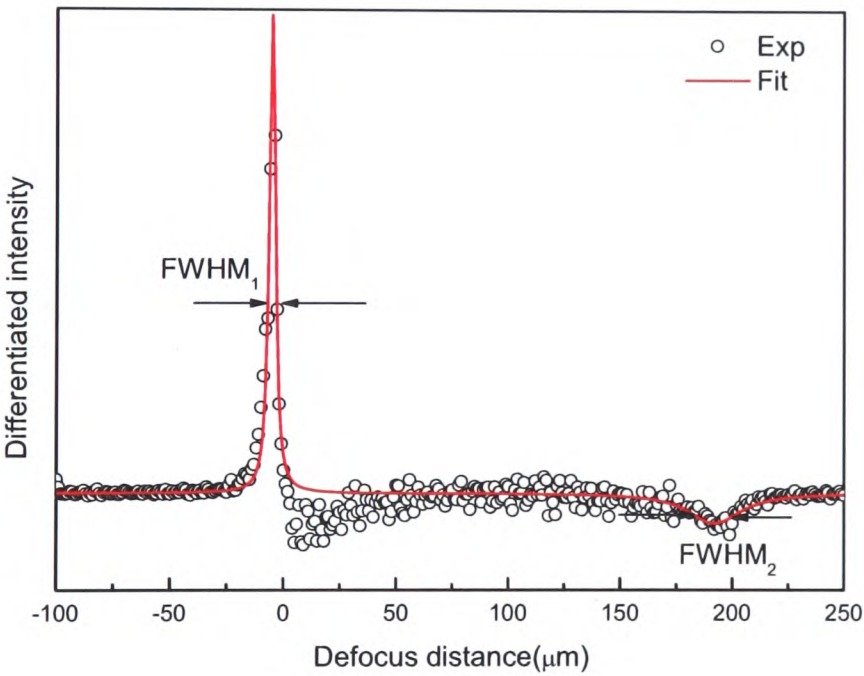
peak are shown here and afterwards. The FWHM was measured from the differentiated intensity-defocus distance curve, as shown on Fig 4.4 (c). The FWHMs measured on the top surface and bottom surface are called  $\text{FWHM}_1$  and  $\text{FWHM}_2$ , respectively and the values are given in Table 4.2. The measured resolutions on the top surface for  $100\times$  and  $50\times$  lenses are not as good as that measured from Si, and both of the resolutions measured on the bottom surface are around  $25\text{ }\mu\text{m}$ , which is very unsatisfactory.



(a)



(b)



(c)

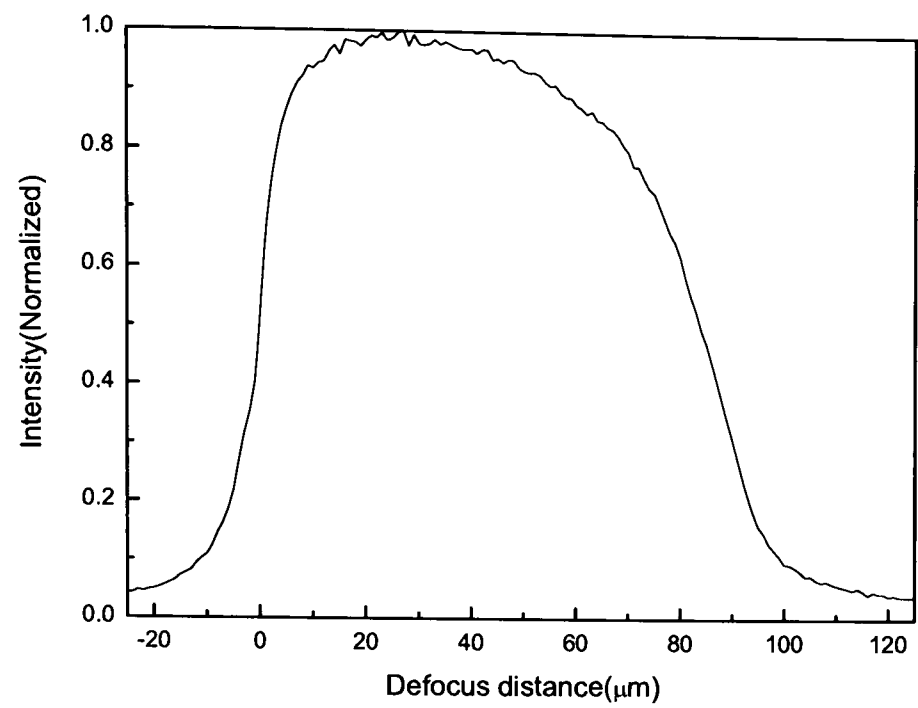
Fig 4.4 Fluorescence intensity-defocus distance curves of sapphire (400 μm)  
(a) 100×; (b) 50×; (c) is the differentiated curve (a) to measure the axial resolution



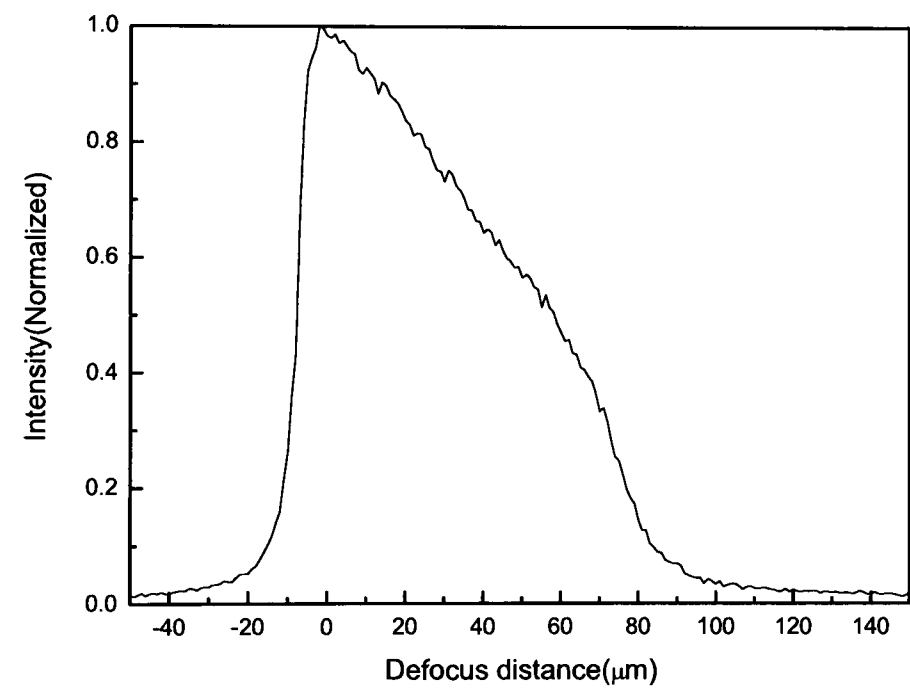
Table 4.2 Measured axial resolutions from sapphire (400  $\mu\text{m}$ ) under different magnifications, M=magnification

| Microscope setting         | FWHM ( $\mu\text{m}$ ) |                   |
|----------------------------|------------------------|-------------------|
|                            | FWHM <sub>1</sub>      | FWHM <sub>2</sub> |
| S=10 $\mu\text{m}$ ; M=100 | 3.2                    | 28.6              |
| S=10 $\mu\text{m}$ ; M=50  | 10.6                   | 24.3              |

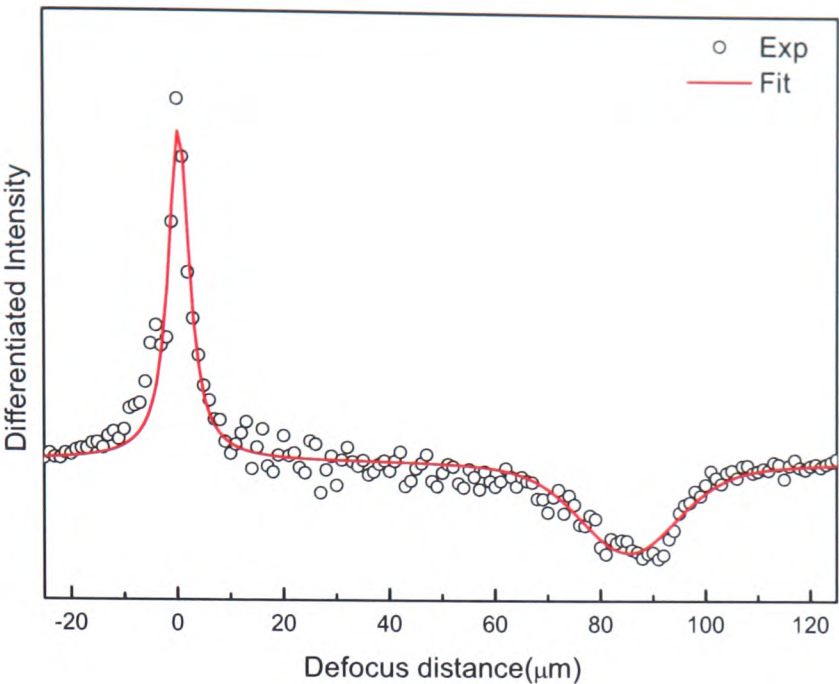
A much thinner sapphire sample (made for transmission electron microscopy, TEM examination) was also used to measure the axial resolution, as a reference test for the oil immersion lens which will be introduced in section 4.2.3. The results are given in Fig 4.5 and Table 4.3. The measured resolutions are quite similar to the results measured from thicker sapphire. Again, the resolution on the bottom surface is worse than 20  $\mu\text{m}$ , which discourages the idea proposed in Fig 4.3. Note that the resolutions on the bottom surface measured from thinner sapphire are better than those measured from thicker sapphire, which indicates the degradation of resolution is dependent on the thickness, or depth of the material.



(a)



(b)



(c)

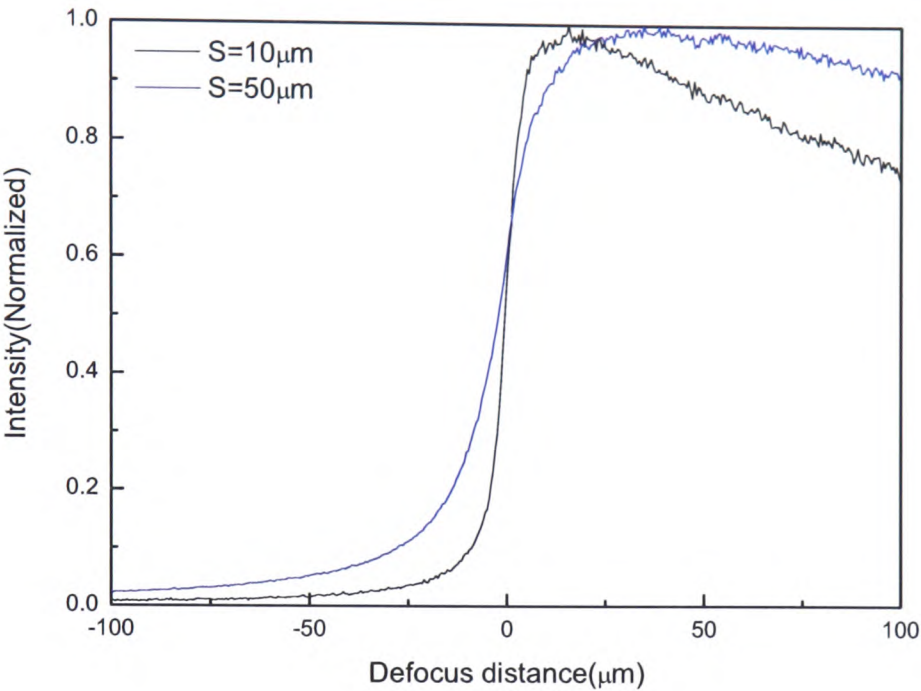
Fig 4.5 Fluorescence intensity-defocus distance curves of sapphire (TEM sample) (a) 100×; (b) 50×; (c) is the differentiated curve (a) to measure the axial resolution

Table 4.3 Measured axial resolutions from sapphire (TEM sample) under different magnifications, M=magnification

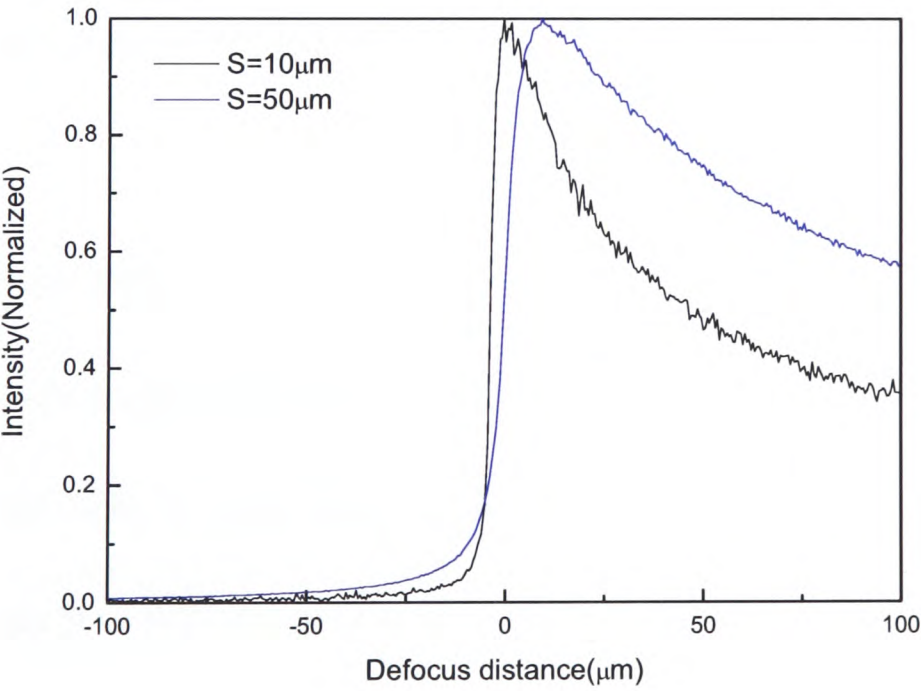
| Microscope setting | FWHM (μm)         |                   |
|--------------------|-------------------|-------------------|
|                    | FWHM <sub>1</sub> | FWHM <sub>2</sub> |
| M=100              | 3.2               | 23.9              |
| M=50               | 4.7               | 22.8              |

Compared with sapphire, ruby is less transparent and is doped with higher  $\text{Cr}^{3+}$  content. It is also a good example to check the validity of the proposed PRF for transparent materials that will be described in section 4.3. The thickness of the ruby sample used here ( $\sim 500\text{ }\mu\text{m}$ ) was much more than the working distance of the objective lenses even considering the refraction effect, hence the laser was not able to

reach the bottom surface. The measured intensity-defocus distance curves and resolutions on the top surface are given in Fig 4.6 and Table 4.4. They are very similar to the corresponding results measured from Si.



(a)



(b)

Fig 4.6 Fluorescence intensity-defocus distance curves of ruby (500  $\mu\text{m}$ )  
(a) 50 $\times$ ; (b) 100 $\times$



Table 4.4 Measured axial resolutions from ruby under different microscope settings  
(S: confocal slit width; M: magnification)

| Microscope setting | FWHM <sub>1</sub> (μm) |
|--------------------|------------------------|
| S=50 μm; M=50      | 15.0                   |
| S=50 μm; M=100     | 5.4                    |
| S=10 μm; M=50      | 6.4                    |
| S=10 μm; M=100     | 2.1                    |

Summarizing all the above measurements made on Si, sapphire with difference thicknesses and ruby, an axial resolution of 2 to 3 microns on the incident surface can be achieved when confocal settings and high magnification are used. FWHM measurements made on different materials are very close and they can be a reasonable estimate of the real resolution. The axial resolution degrades when the laser is focused below the surface, even in transparent materials such as sapphire and ruby.

4.2.2 Lateral Resolution

The lateral resolution can be measured from the intensity curve by scanning the laser across a material with a very sharp edge or feature, or scanning across a sharp interface between two different materials. The latter case is schematically shown in Fig 4.7, where a sapphire was diffusion bonded with a piece of ruby and the interface was aligned to parallel to X or Y axis of the sample stage.

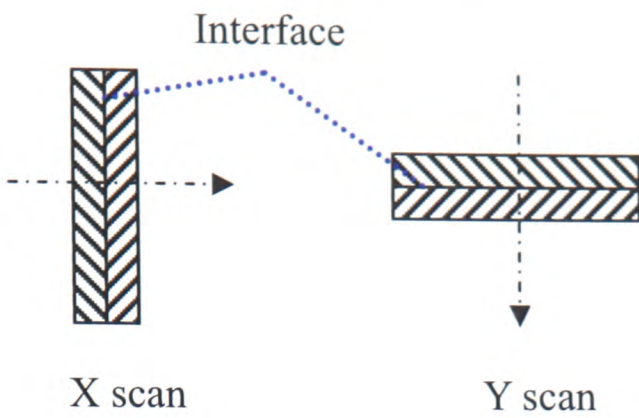
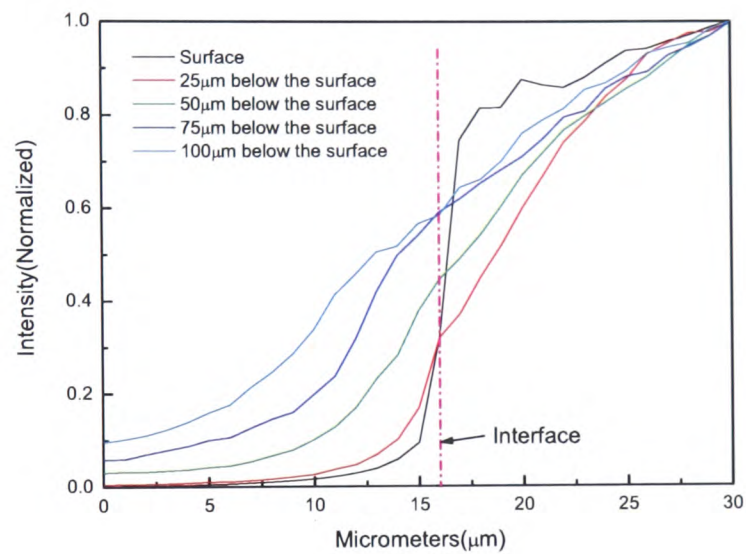
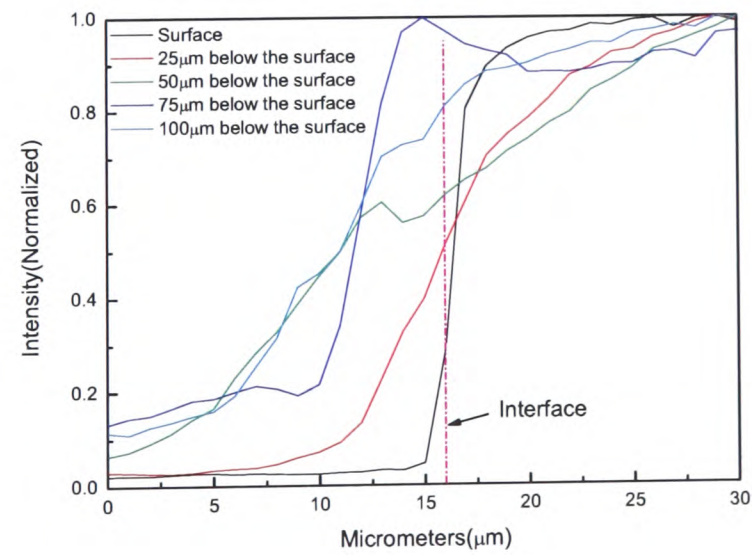


Fig 4.7 Schematic showing lateral scanning along X or Y axis



(a)



(b)

Fig 4.8 Fluorescence intensity curve from scanning the interface of diffusion bonded ruby/sapphire

(a) X scan; (b) Y scan

The intensity curves for both X scan and Y scan are shown in Fig 4.8. Lateral scans are also made at several distances below the surface to see how the lateral resolution degrades inside the material. After measuring the FWHM of the differentiated intensity curve, the lateral resolution on the surface is about 1.5  $\mu\text{m}$  (Table 4.5), for both the X and Y scans. It is slightly worse than the best lateral resolution claimed by the manufacturer (1  $\mu\text{m}$ ). The lateral resolution apparently gets significantly worse inside the material, but due to the differentiated intensity curves below surface being very noisy, no accurate numbers can be drawn but a rough estimate shows that at 25  $\mu\text{m}$  below the top surface the resolution has already degraded to approximately 9  $\mu\text{m}$ .

Table 4.5 Measured lateral resolution from a diffusion bonded ruby/sapphire

| Scan mode | FWHM ( $\mu\text{m}$ ) |
|-----------|------------------------|
| X Scan    | 1.6                    |
| Y scan    | 1.4                    |

4.2.3 Refraction Effect on Axial Resolution

The spatial resolution of the Raman microscope used for future investigation has been experimentally measured using the FWHM method described in the above sections. Some possible ways to improve the resolution, more specifically, the axial resolution, will be introduced and tried out in this section.

The idea is based on the ray-tracing analysis proposed by Everall [3]. Commercial microscopes are usually configured with metallurgical objectives that are designed to work in air. The tightly convergent laser beam will suffer refraction at the sample surface, i.e., the actual focal position is deeper than nominal focal point depth,  $\Delta$ , as illustrated in Fig 4.9.

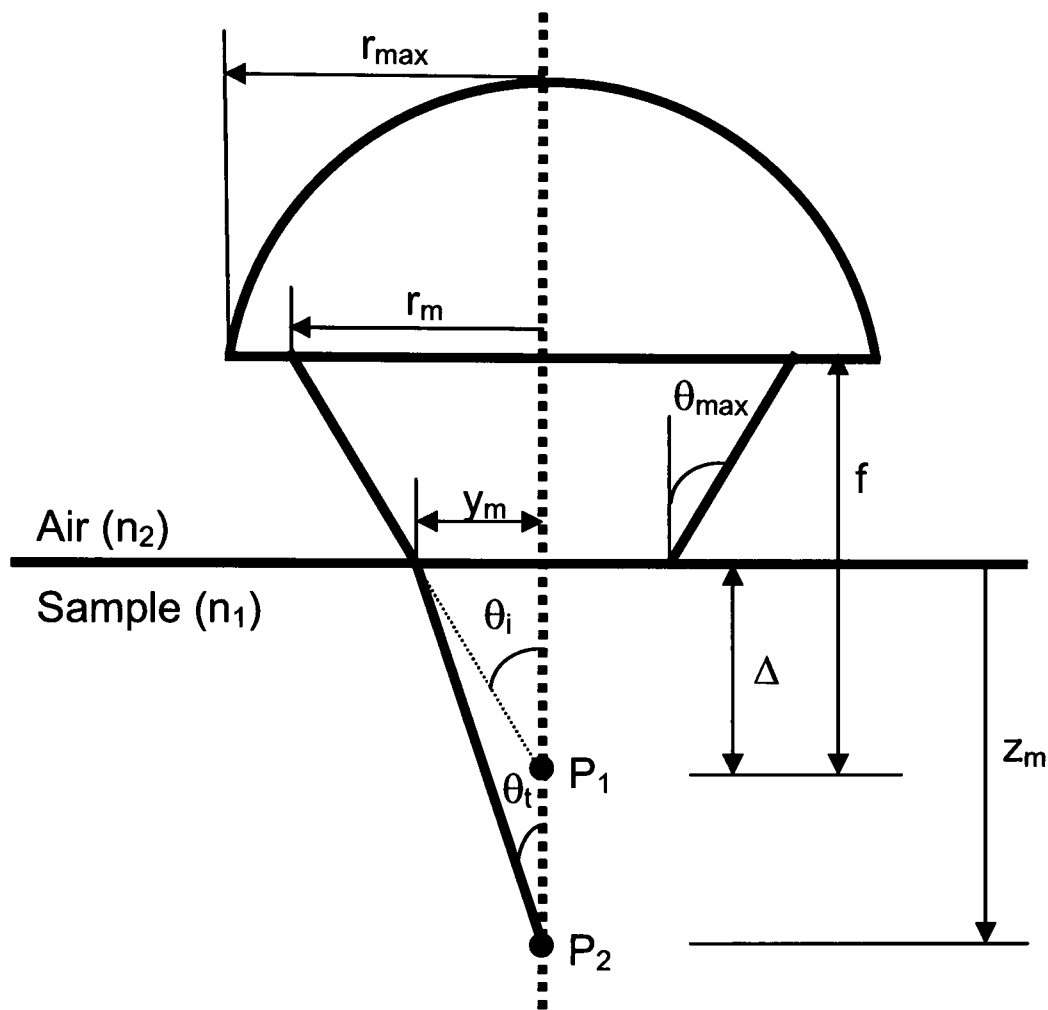


Fig 4.9 Definition of parameters used in ray-tracing analysis (after [3])

According to Everall's paper [3] which considered the effect of refraction at the surface on axial resolution, the distance of focus point below the surface ( $z_m$ ) and the range of laser focal positions with the sample (i.e., the depth of field, *d.o.f*) is given by:

$$z_m = \Delta \left[ m^2 \cdot \frac{NA^2(n^2 - 1)}{(1 - NA^2)} + n^2 \right]^{1/2} \quad (4.1)$$



$$d.o.f = \Delta \left[ \left[ \frac{NA^2(n^2 - 1)}{(1 - NA^2)} + n^2 \right]^{1/2} - n \right] \quad (4.2)$$

$$r_{\max} = \frac{fNA}{(1 - NA^2)^{1/2}} \quad (4.3)$$

where  $m$  is the normalized aperture radius,  $m=r_m/r_{\max}$ .  $m=1$  means the highest possible angle of incidence and  $m=0$  means the ray is normal to the sample surface;  $n=n_1/n_2$ ,  $n_1$  and  $n_2$  are the refractive indexes of the sample and the air;  $NA$  is the numerical aperture;  $\Delta$  is the nominal focal point position;  $f$  is the focal length of the lens.

From Equation 4.1, even for an ideal system the refraction means that rays at different distance from the center of the lens ( $m$ : 0~1) no longer come to focus at a single depth below the surface, so the axial resolution is degraded. A desirable objective lens to be used for depth scanning of stress fields induced by indentation or grinding/polishing is expected to have both high resolution and short depth of field ( $d.o.f$ ). The theoretical axial resolution for confocal microscope is defined in Equation 2.24 of Chapter 2 as  $1.5n_2\lambda / NA^2$  and higher axial resolution requires higher  $NA$ . However, according to Equation 4.2, higher  $NA$  will lead to longer  $d.o.f$ . The effect of  $NA$  on  $d.o.f$  and the theoretical axial resolution can be seen from Fig 4.10.

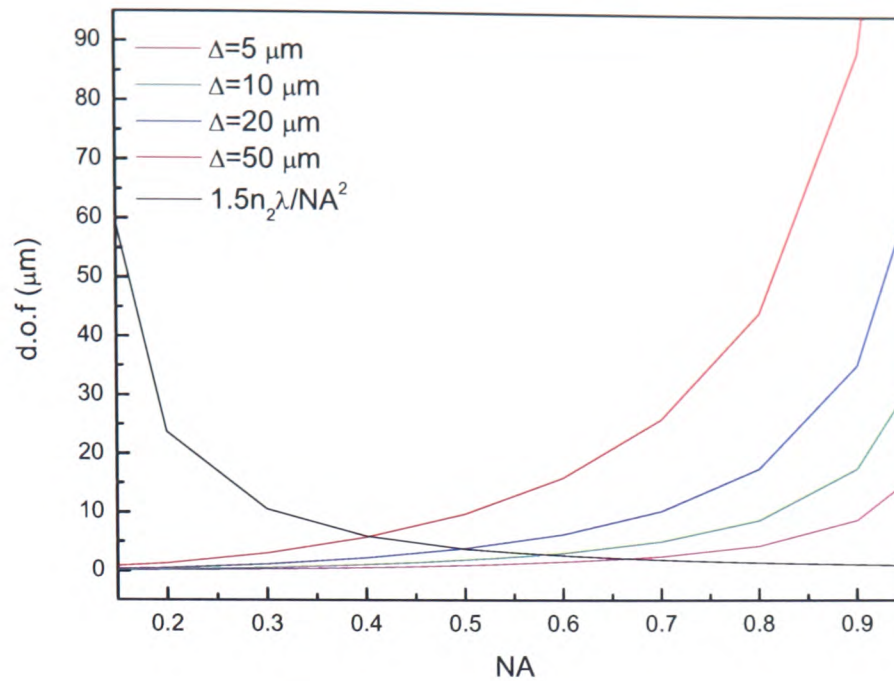


Fig 4.10 Dependence of depth of field and theoretical axial resolution on NA

The design of the Renishaw Raman microscope used in this work requires the use of a 100× lens with  $NA$  of 0.9 for the confocal setting, which means the  $d.o.f$  reaches about 18  $\mu\text{m}$  when the nominal focal depth is 10  $\mu\text{m}$  (Fig 4.10). Large  $d.o.f$  degrades the axial resolution: the signal collected at some depth actually comes from a long range around that position.

Decrease of  $d.o.f$  can be achieved by reducing  $n$  in Equation 4.2, i.e., to alleviate the refraction effect. The refraction effect can be minimized using an oil immersion objective with an oil of refractive index similar to that of the material tested (increasing  $n_2$ ). However, there is a disadvantage of oil immersion objectives which limits their application for investigating stresses in the depth direction of indented or ground materials: their short working distances, normally no more than 100  $\mu\text{m}$ . A

sapphire TEM sample was used in this work to test whether an oil immersion lens would improve the axial resolution. The oil immersion lens used was a 100× Leica lens with a  $NA$  range of 1.4-1.7 and working distance of 90  $\mu\text{m}$ . The  $NA$  could be adjusted using an iris within the lens. The oil had a refractive index of 1.51, which is close but still smaller than that of sapphire (1.8).

The measured axial resolutions with different iris opening conditions are listed in Table 4.6, with corresponding intensity-defocus distance curves shown in Fig 4.11. The best axial resolution on the incident surface is 5.4  $\mu\text{m}$ , which is worse than the 100× dry lens. However, the resolution measured on the bottom surface is much better, being around 10  $\mu\text{m}$ . The degradation of axial resolution inside the material is significantly reduced in the oil immersion lens.

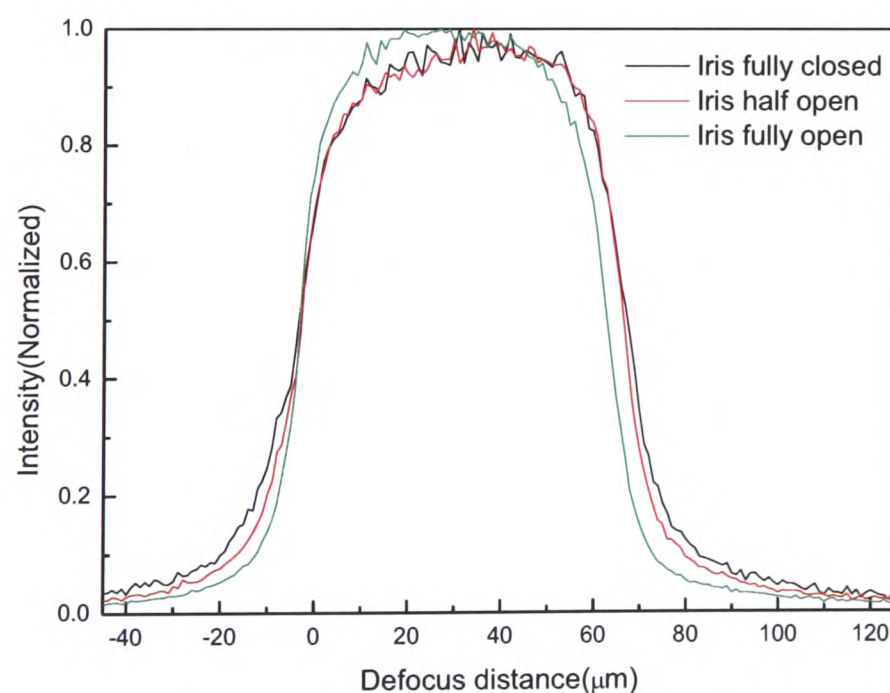


Fig 4.11 Intensity-defocus distance curves measured from a sapphire TEM sample using an oil immersion objective lens

Table 4.6 Measured axial resolution from a sapphire TEM sample using an oil immersion objective lens, with different iris opening conditions

| Iris Condition    | FWHM <sub>1</sub> (μm) | FWHM <sub>2</sub> (μm) |
|-------------------|------------------------|------------------------|
| Iris fully closed | 10.1                   | 13.8                   |
| Iris half open    | 7.5                    | 10.1                   |
| Iris fully open   | 5.4                    | 10.5                   |

The effect of refraction can also be reduced by using some special objective lens with a correction collar, which is designed to correct for the cover glass commonly used for biological specimen. Rotation of the correction collar separates or brings together some of the internal glass elements of the objective. The range of correction of the collar is usually equivalent to 0.11 to 0.22 mm of glass. A 40× Leica lens with a *NA* of 0.85 and a working distance of 240 μm was used to test the effect on axial resolution in sapphire. Results are shown in Fig 4.12 and Table 4.7. The best axial resolutions can be achieved on the top surface and bottom surface are 9.2 and 14.3 μm, respectively. Similar to the 100× oil immersion lens, resolution on the bottom surface is much improved over the standard 100× lens; however, the resolution on the incident surface is worse than both the 100× and the 50× lenses.



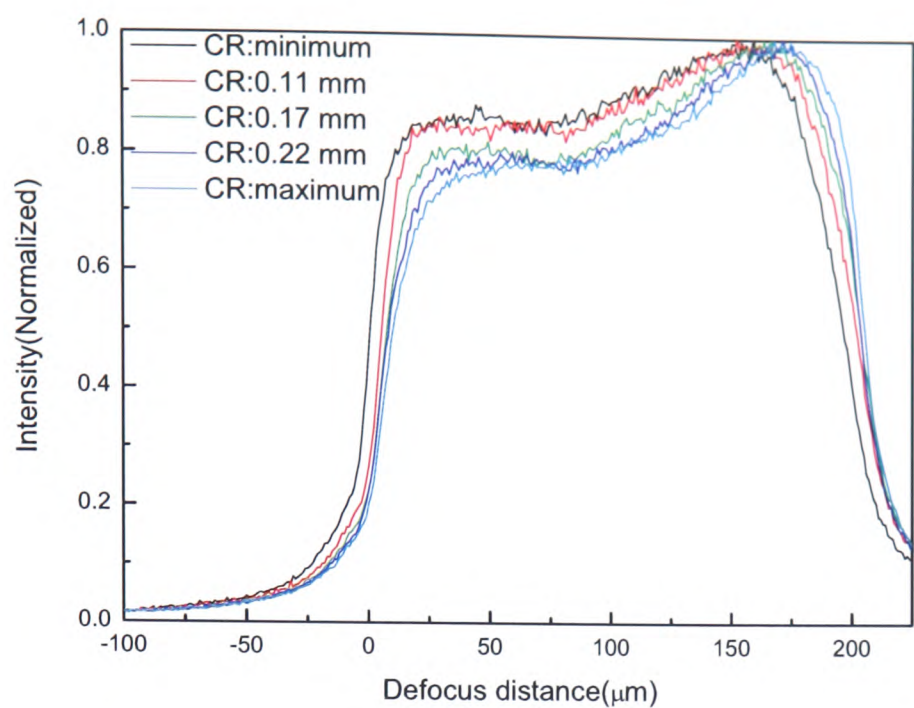


Fig 4.12 Intensity-defocus distance curves measured from a sapphire sample (400 μm) using a lens with correction collar, CR=correction

To summarize: theoretically, choosing an ideal objective lens can improve the axial resolution; experimentally, although substantial improvements have been found when focusing beneath the surface of transparent materials, such an ideal objective lens is not manufactured for our application. As a result, the 100× lens provided in Renishaw’s Raman microscope will be used throughout the work afterwards.

Table 4.7 Measured axial resolution from a sapphire sample (400 μm) using a lens with correction collar

| Correction of Collar (mm) | FWHM <sub>1</sub> (μm) | FWHM <sub>2</sub> (μm) |
|---------------------------|------------------------|------------------------|
| minimum                   | 9.2                    | 33.3                   |
| 0.11                      | 11.6                   | 32.3                   |
| 0.17                      | 13.4                   | 26.7                   |
| 0.22                      | 13.4                   | 18.2                   |
| maximum                   | 15                     | 14.3                   |

4.3 Probe Response Function

4.3.1 Convolution & Deconvolution

The depth resolution (axial resolution) of the optical microscope used for the fluorescence measurements is determined by the numerical aperture (*NA*) of the objective lens and the size of the collection aperture used [4]. When the laser is focused at a distance *z* below the surface of a transparent material such as sapphire, only the signal generated in a limited volume indicated in Fig 4.13 is collected. The total signal collected at depth *z* is actually the integral of the fluorescence from all depths within the material.

$$I(z_0) = \int_{-z_0}^{-z_0} g(z)dz$$

(4.4)

where *t* is the thickness of material, and *g(z)* is the depth resolution function of the lens, also called the probe response function (PRF).

*g(z)* describes the relative collection efficiency as a function of *z*, which is the

distance from the focal position. Normally, the focal plane is set to be on the top surface. Experimentally, the PRF can be obtained from the intensity-defocus distance curve, from which the FWHM was measured in section 4.2. For a transparent material like sapphire, the resolution function is differentiated from the intensity profile using a thick specimen [2, 4]:

$$g(z) = \frac{dI(z)}{dz} \quad (4.5)$$

Suppose the stress field is a function of depth only, the measured fluorescence line shift at depth  $z_0$  is a weighted average of the signals from all depths:

$$\overline{\Delta\nu(z_0)} = \frac{\int_{-z_0}^{-z_0} \Delta\nu(z_0 + z)g(z)dz}{\int_{-z_0}^{-z_0} g(z)dz} \quad (4.6)$$

If the depth resolution function  $g(z)$  is known, the real depth profile  $\Delta\nu(z_0)$  can in principle be deconvoluted from the experimental depth scan results using the above equation. Alternatively, an assumed  $\Delta\nu(z_0)$  (with adjustable parameters) can be convoluted with the known depth resolution function to produce a virtual average R-line shift curve,  $\overline{\Delta\nu(z_0)}$ . The adjustable parameters can be determined by the best fitting of the virtual curve to the experimentally observed R-line shift curve using a curve fitting method. The latter method is easier and better defined mathematically and will be used in analyzing stresses induced by indentation or grinding and polishing.

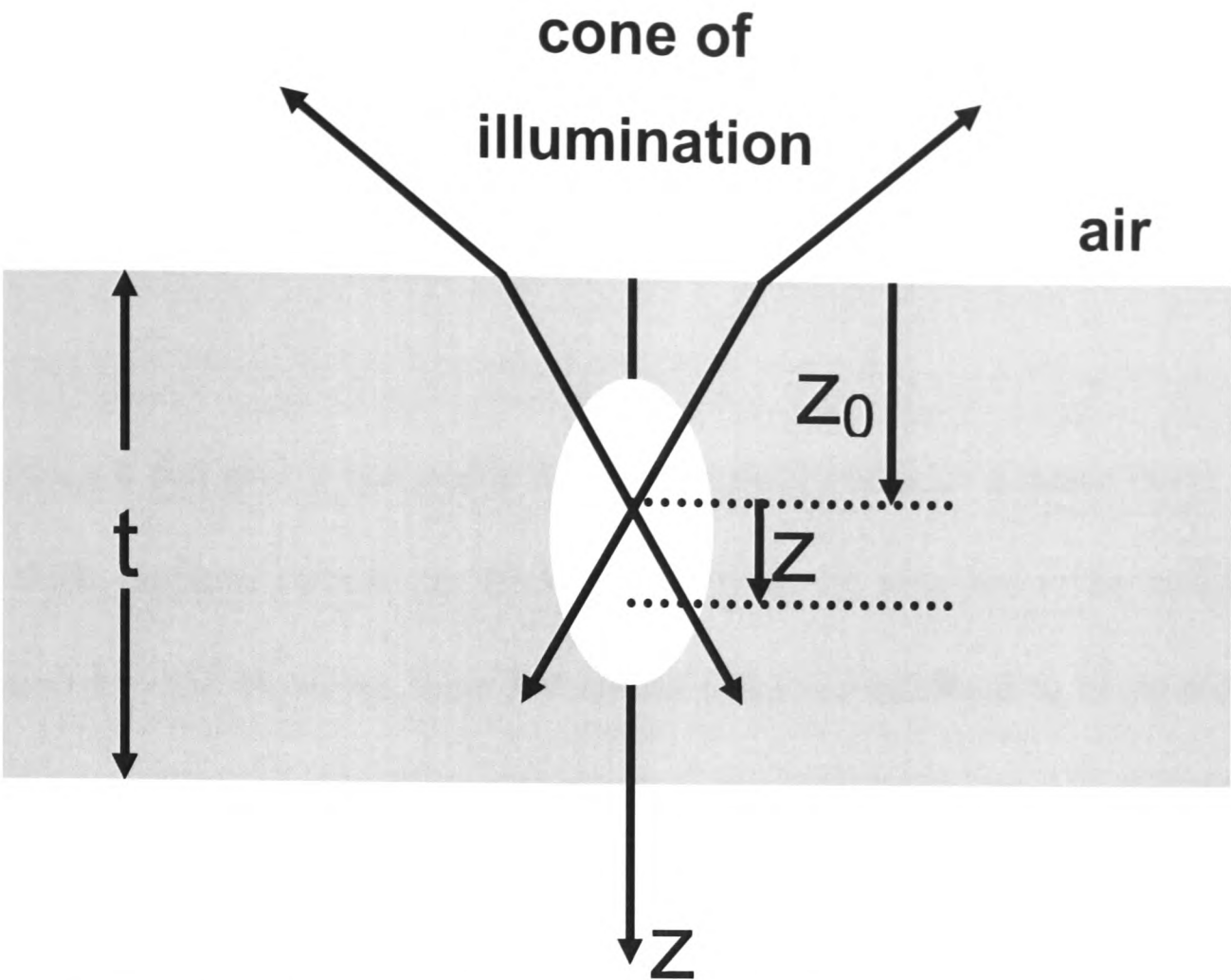


Fig 4.13 Illustration of the effect of the finite depth of field of the optical system (after [4])

4.3.2 Modeling of PRF

A mathematical description for PRF is required for the convolution in Equation 4.6. In section 2.5.4 of Chapter 2, previous work on PRF has been summarized. The basic model is proposed by Atkinson et al. [5]:

$$I(z_0) = \int_0^\infty \exp(-2\alpha z) \frac{p^2}{p^2 + (z - nz_0)^2} dz \tag{4.7}$$

where  $z_0$  is the distance away from the focal plane, which is set to be on the top surface of the material;  $p$  is half of the probe length, which should equal the experimentally measured FWHM;  $n$  is the refractive index of the material and  $\alpha$  is the absorption coefficient.

Equation 4.7 can be integrated analytically when  $\alpha$  is zero (for a completely transparent material like sapphire):

$$\frac{I(z_0)}{I(z_0)_{\max}} = \frac{1}{2} - \frac{1}{\pi} \arctan\left(-\frac{z_0}{p}\right) \quad (4.8)$$

Equation 4.8 can give a reasonable fit to the intensity-defocus distance curve for a very thick sapphire (where the thickness of it can be assumed to be infinity in Equation 4.7) [5]. However, there are several problems that need to be pointed out here.

1) The thickness of the sample cannot be assumed to be infinity for a sample that is thin relative to the depth below surface to which the probe extends. Taking the 100× lens that is commonly used for this research as an example, its working distance is 270  $\mu\text{m}$ . Considering the refraction effect ( $n \approx 1.80$ ), the laser can reach to about 480  $\mu\text{m}$  below surface. If a sample of thickness less than 480  $\mu\text{m}$  is used, as the case for the 400  $\mu\text{m}$ -thick sapphire and TEM sapphire used in section 4.2, the probe exits the bottom surface reducing the intensity. As a result, the thickness of sample has to be measured.

2) The actual focal position is not  $nz_0$  given in Equation 4.7. According to Equation 4.1, that is only true when  $m$  equals zero, i.e., incident ray normal to sample surface. However, that is an ideal condition and normally  $m$  is between 0 and 1. Hence, the actual focal position is deeper than  $nz_0$ . An effective refractive index,  $n'$

$(n' = \sqrt{m^2 NA^2 (n^2 - 1) / (1 - NA^2)} + n^2$ , see Equation 4.1), will be used to account for this. If  $NA=0.9$  and  $n=n_1/n_2=1.8$ ,  $n'$  is in the range of 1.8 to 3.6.

3) For the illuminated intensity defined in Equation 4.7, the integral is a function of half probe length,  $p$ :

$$\int_0^\infty \frac{p^2}{p^2 + z^2} dz = \frac{\pi p}{2} \quad (4.9)$$

As  $p$  goes into the fitting ( $P_f$  in Equation 4.13), it seems reasonable to use the form

$\frac{p}{p^2 + z^2}$  to replace  $\frac{p^2}{p^2 + z^2}$ , considering both  $p/(p^2 + z^2)$  and  $p^2/(p^2 + z^2)$  give a

Lorentzian shape and it is this Lorentzian shape that really matters:

$$\int_0^\infty \frac{p}{p^2 + z^2} dz = \frac{\pi}{2} \quad (4.10)$$

4) The broadening of the resolution function or the degradation of resolution with depth is not considered in Equation 4.7. In section 4.2, the effect of sample thickness on the resolution measured on the bottom surface has been found and it clearly shows the axial resolution dependence on depth. The broadening of the resolution function is discussed in Reference [6] and [7] and an almost linear relationship of broadening with depth can be assumed, at least for a limited depth.

$$p = p_0 + az \quad (4.11)$$

where  $p_0$  is the depth resolution on the top surface and  $a$  is the linear coefficient for  $p$  broadening with depth  $z$ .

5) For a transparent material like sapphire, the illuminated intensity comes from a long range of depth, and the intensity coming from each depth varies. This is mathematically expressed by a linear Fredholm integral [7], or by multiplying by another function,  $I'(z_0)$ , to resolution function.  $I'(z_0)$  can be assumed to have a Lorentzian shape [6, 7]:

$$I'(z_0) = (1-d) \frac{b^2}{b^2 + (z_0 - c)^2} + d \quad (4.12)$$

where  $b$ ,  $c$  and  $d$  are the width, center position and offset of the Lorentzian function.

6) There is an issue regarding the correct position of the zero depth. The laser is set to focus on the top surface of the material before a depth scan begins. This is by minimizing the laser spot size on the surface and the zero depth plane ( $z=0$ ) is set to be on the sample surface. However, it cannot be accurate to tell when the laser spot is minimal by eye. The actual zero position will deviate from the real sample surface by up to several micrometers. This offset has to be taken into account when doing the fitting.



Equation 4.7 is then modified according to the points made through above discussions and the fitting was carried out by the commercial software package, OriginPro (OriginLab, USA), using its nonlinear fitting module. The final expression of PRF used for fitting the experimentally measured intensity-defocus distance curve is:

$$I(z_0) = P_6 I'(z_0) \int_0^t \frac{P_1 + P_2(z_0 - P_8)}{([P_1 + P_2(z_0 - P_8)]^2 + [z - P_3(z_0 - P_8)]^2)} dz \quad (4.13)$$

$$I'(z_0) = (1 - P_7) \frac{P_4^2}{P_4^2 + (z_0 - P_5 - P_8)^2} + P_7 \quad (4.14)$$

where:  $t$  is the sample thickness,  $\mu\text{m}$ ;  $P_1$  is the depth resolution on the top surface,  $\mu\text{m}$ ;  $P_2$  is the linear coefficient of resolution broadening with depth, unitless;  $P_3$  is the effective refractive index, unitless;  $P_4$ ,  $P_5$  and  $P_7$  are the width ( $\mu\text{m}$ ), center position ( $\mu\text{m}$ ) and offset of the Lorentzian function (unitless) to account for illuminated intensity at different depth;  $P_6$  is the normalizing parameter for the measured intensity, unitless;  $P_8$  is the offset of zero depth position,  $\mu\text{m}$ .

The fitting for a 400- $\mu\text{m}$  thick sapphire is shown in Fig 4.14. Half of the probe length is 2.9  $\mu\text{m}$ , which is close to the FWHM measured from sapphire of the same thickness and Si. The linear coefficient of resolution broadening is 0.09, so even at 10  $\mu\text{m}$  below surface the axial resolution is still less than 4  $\mu\text{m}$ , which is reasonably good. The effective refractive index is 2.09, within the range based on Everall's ray-tracing method,  $n' = \sqrt{m^2 NA^2 (n^2 - 1) / (1 - NA^2) + n^2}$ .

Considering so many parameters in the fitting (also in the following models as well),



the sensitivity of these parameters is certainly a concern. The fitted parameters come from the best fit (they are reproducible) to the experimental results and it has been proved that less parameter would not generate a good fitting. The fitted parameters justify their physical meaning as they are all reasonable (e.g., probe length and effective refractive index as describe above), which is further validated in the following fitting for ruby and polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites: for example, the fitted absorption coefficient ( $P_9$  in Equation 4.15) increase in the more opaque materials; the parameters characterizing the scattering is higher in nanocomposite compared to in alumina ( $P_7/P_6$  in Equation 4.17); the zero depth position offset ( $P_8$  in Equation 4.13) is normally within  $\pm 2\mu\text{m}$ . It is difficult to say which parameter most or least determine the fitting, but the fact that the fitting parameters are quite reproducible (mathematically), together with their being of reasonable values (physically) further confirms their validity.

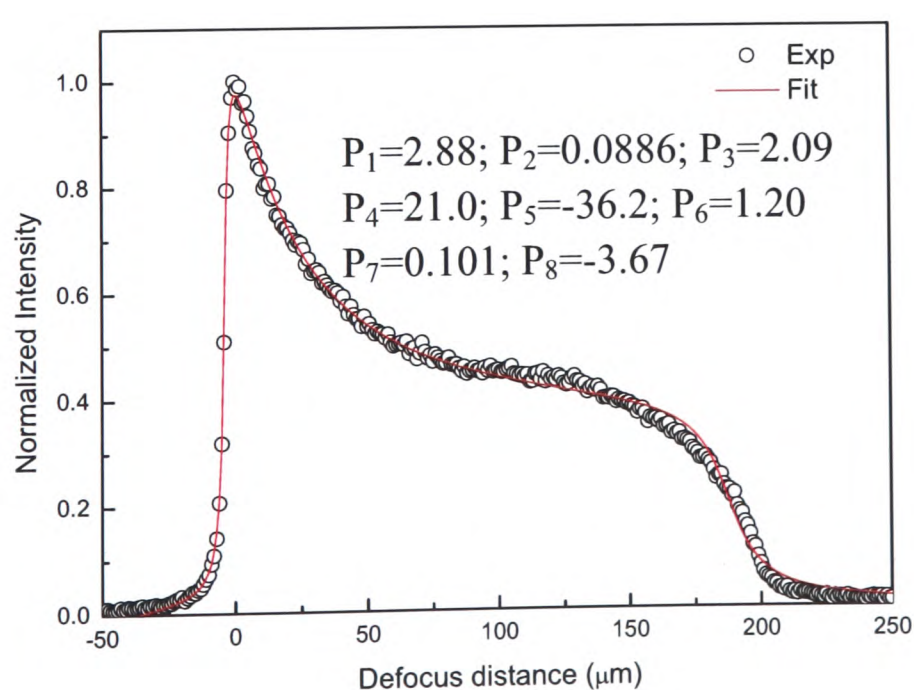


Fig 4.14 Fitting of the intensity-defocus distance curve for a 400-μm thick sapphire

In Equation 4.13, absorption is not taken into account as sapphire is very transparent and the absorption is negligible. Ruby is also transparent but its absorption is stronger than that of sapphire. By adding the absorption effect, the same model can be used to fit the intensity-defocus distance curve of ruby. The modified models are given in Equation 4.15. Except for the intensity normalizing factor  $P_6$  and zero depth offset parameter  $P_8$ , the same parameters as were found from sapphire were used as other parameters can be assumed to be instrument dependent. Two ruby samples were used: 500  $\mu\text{m}$ -thick (0001) ruby and 530- $\mu\text{m}$  thick (11 $\bar{2}$ 0) ruby. The fitting results are shown in Fig 4.15 and Fig 4.16.

$$I(z_0) = P_6 I'(z_0) \int_0^t \exp(-2P_9 z) \frac{P_1 + P_2(z_0 - P_8)}{([P_1 + P_2(z_0 - P_8)]^2 + [z - P_3(z_0 - P_8)]^2)} dz \quad (4.15)$$

$$I'(z_0) = \left( (1 - P_7) \frac{P_4^2}{P_4^2 + (z_0 - P_5 - P_8)^2} + P_7 \right) \quad (4.16)$$

where  $P_9$  is the absorption coefficient ( $\mu\text{m}^{-1}$ ) and the other parameters are the same as defined in Equation 4.13.

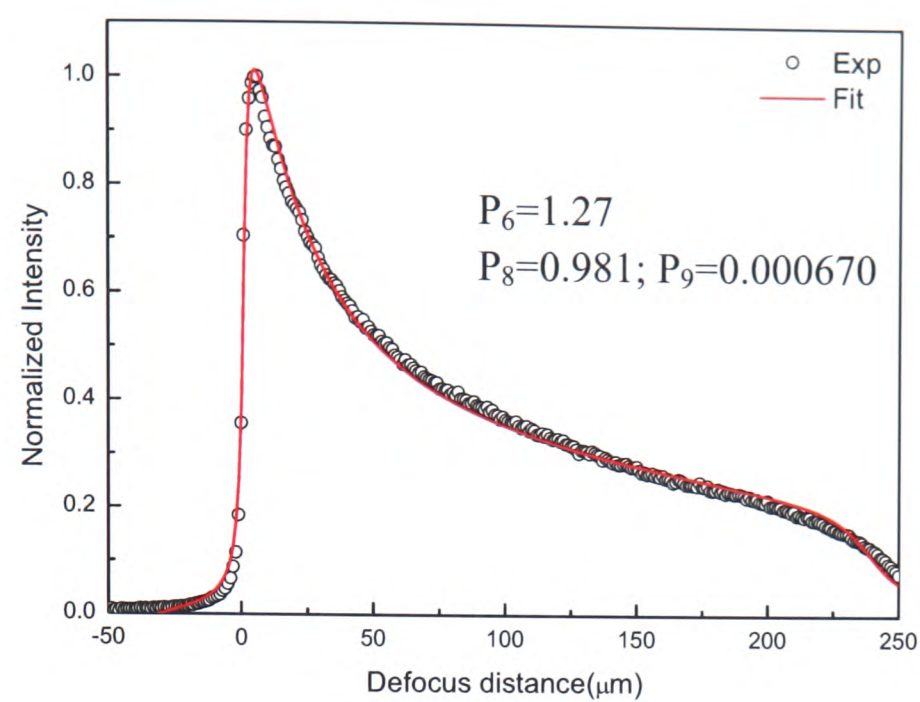


Fig 4.15 Fitting of the intensity-defocus distance curve for a 500- $\mu\text{m}$  thick (0001) ruby

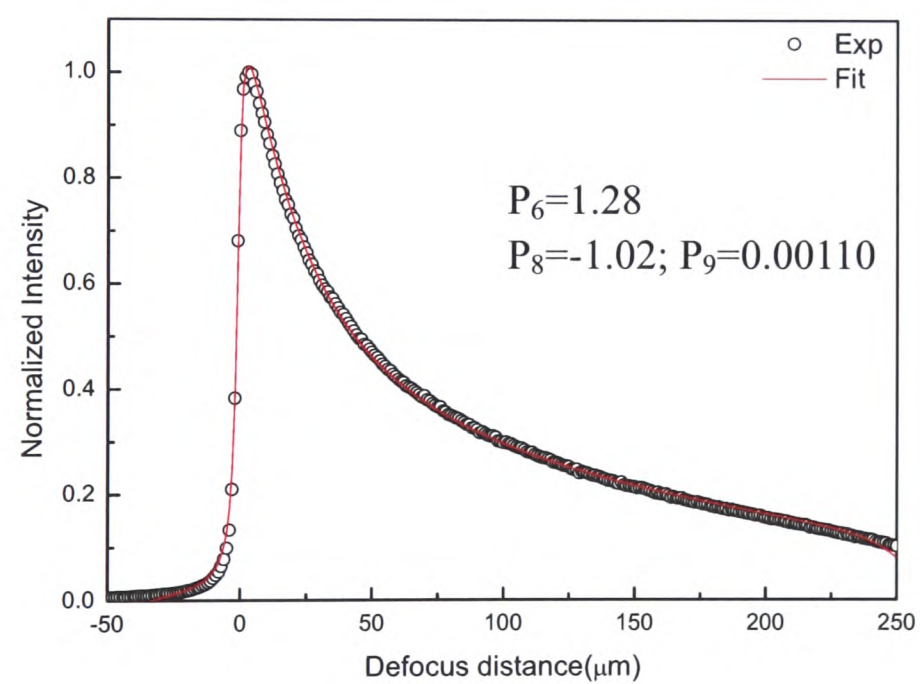


Fig 4.16 Fitting of the intensity-defocus distance curve for a 530- $\mu\text{m}$  thick  $(11\bar{2}0)$  ruby

Good fittings were achieved for both ruby samples, and the fact that essentially only one parameter (absorption coefficient) was varied to achieve this fit confirms the validity of the physical basis of the PRF model proposed above. The absorption

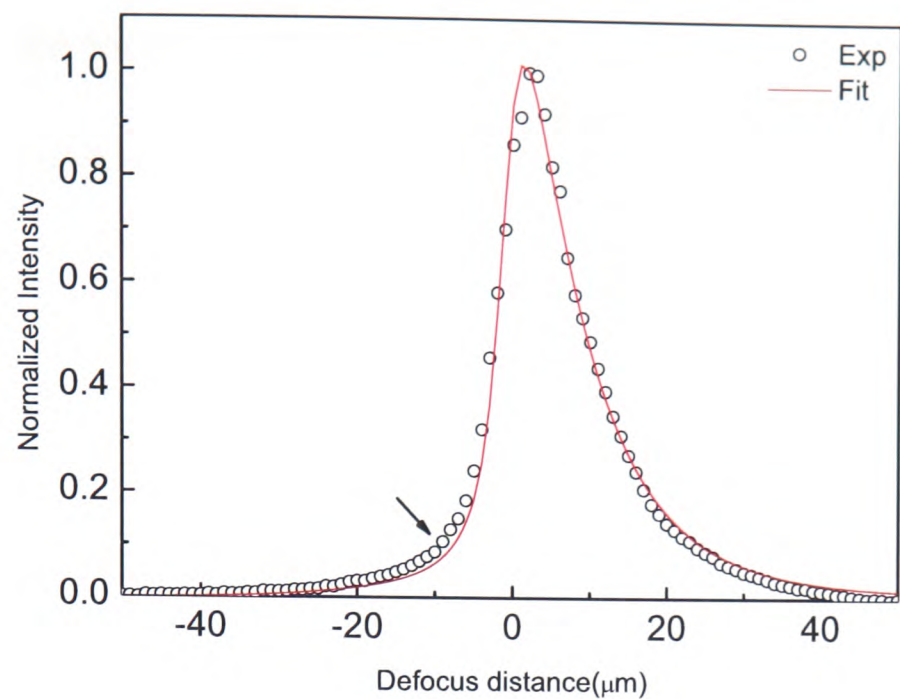
coefficients for both ruby samples are very small but they still affect the intensity variation against defocus distance. The absorption coefficients of sapphire and ruby are summarized in Table 4.8. The values are different for the two rubies because they have different chromium impurity concentration (higher in  $(1\bar{1}20)$  ruby).

Table 4.8 Fitting parameters of absorption coefficients for sapphire and ruby

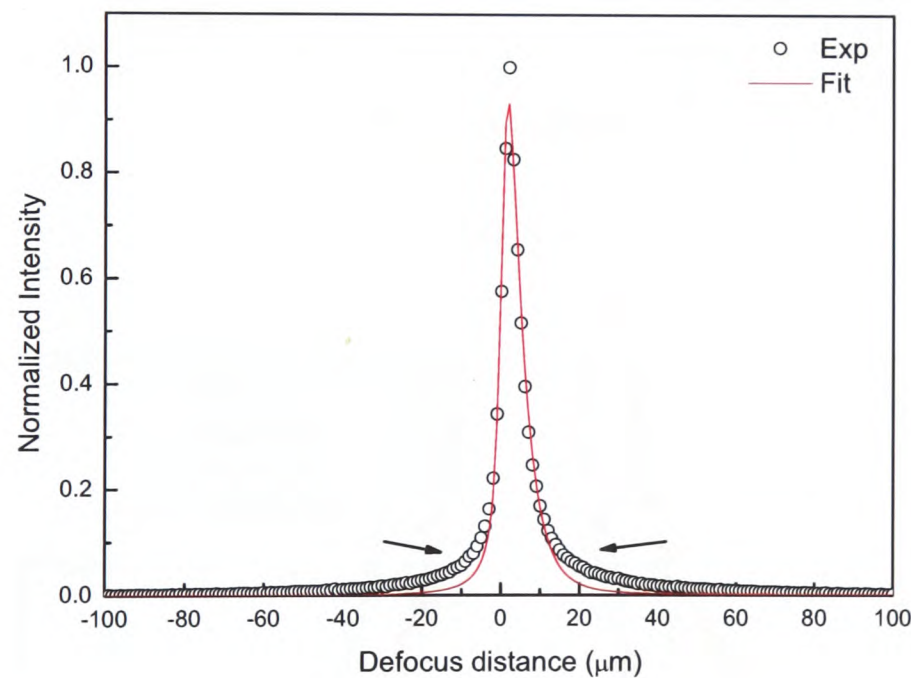
| Material            | Absorption Coefficient ( $\mu\text{m}^{-1}$ ) |
|---------------------|-----------------------------------------------|
| Sapphire            | 0                                             |
| (0001) Ruby         | 0.0007                                        |
| $(11\bar{2}0)$ Ruby | 0.001                                         |

In principle, the same PRF should be applied to translucent polycrystalline alumina and more opaque  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites. The main difference between transparent sapphire, ruby and these materials is the absorption coefficient. However, as seen from Fig 4.17, the fitting for alumina is not perfect and is even worse for the  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposite. It suggests some information is missing for modeling the PRF of alumina and nanocomposites.





(a)



(b)

Fig 4.17 Fitting of the intensity-defocus distance curves for (a) 5.6  $\mu\text{m}$   $\text{Al}_2\text{O}_3$  and (b) 2 vol.%  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposite using PRF in Equation 4.15. The arrows indicate the misfit.

By re-inspecting the intensity-defocus distance curves of alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites (shown in Fig 4.18), some interesting details can be observed. Compared to alumina, the intensities of nanocomposites decrease more quickly as the

probe goes below the surface, due to their larger light absorption. It is not true, however, when the focus is above surface, as shown in Fig 4.18 (b). On both sides, above or below surface, intensity of alumina converges to zero much slowly than nanocomposites.

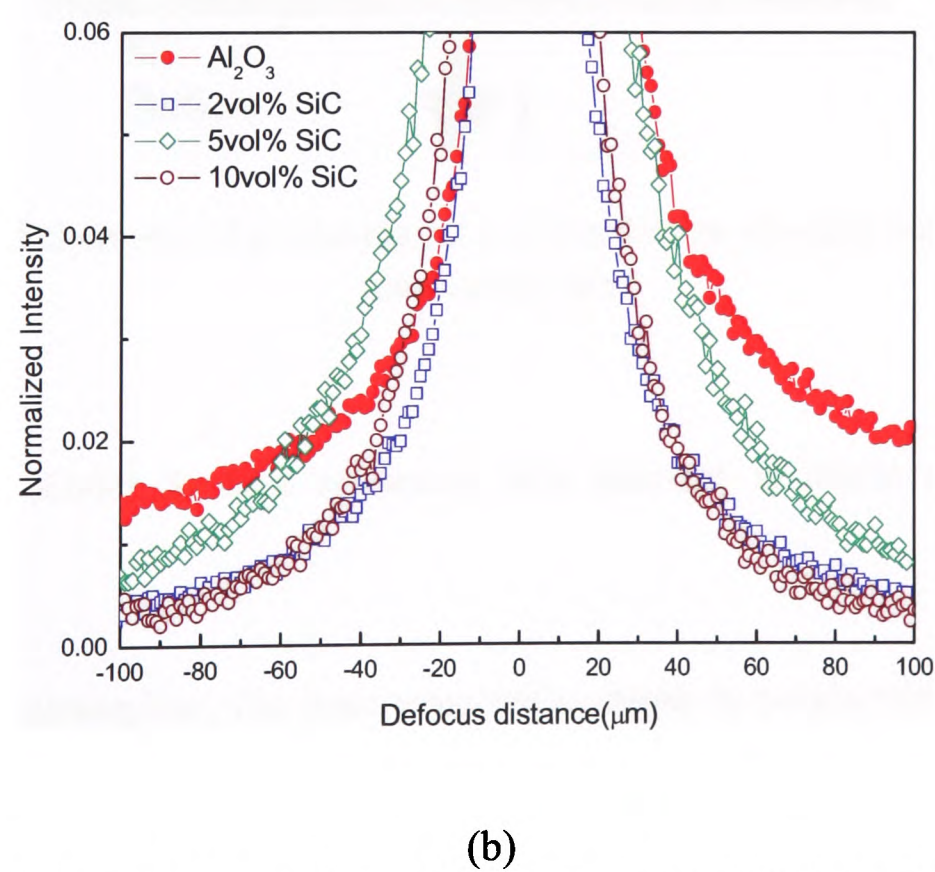
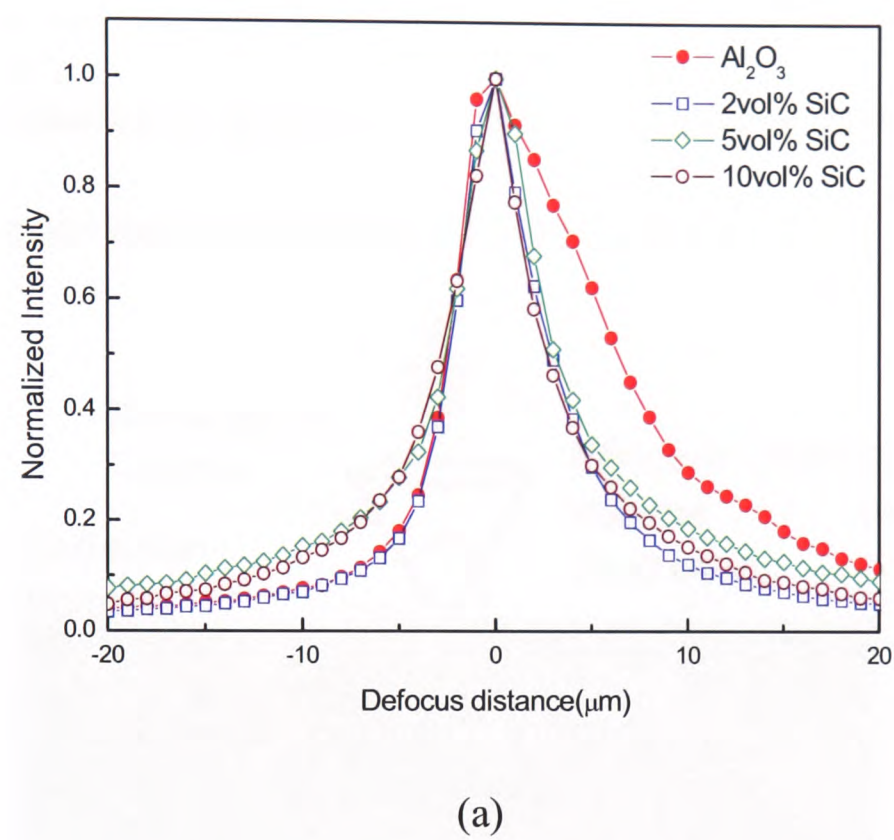


Fig 4.18 (a) Intensity-defocus distance curves of  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites; (b) is an enlargement of the tail part at distances from the surface greater than that shown in (a)



Apart from different absorption, the other main difference between single crystal sapphire or ruby and polycrystalline alumina or the nanocomposites that might cause the different behavior of intensity variation with depth is scattering [8]: scattering on the grain boundaries and pores, and scattering due to small SiC particles. The scattering makes the intensity profiles broader at the tail part: part of signal that Equation 4.15 assumes to have been absorbed is scattered back to the collection volume. A schematic shown of scattering is given in Fig 4.19.

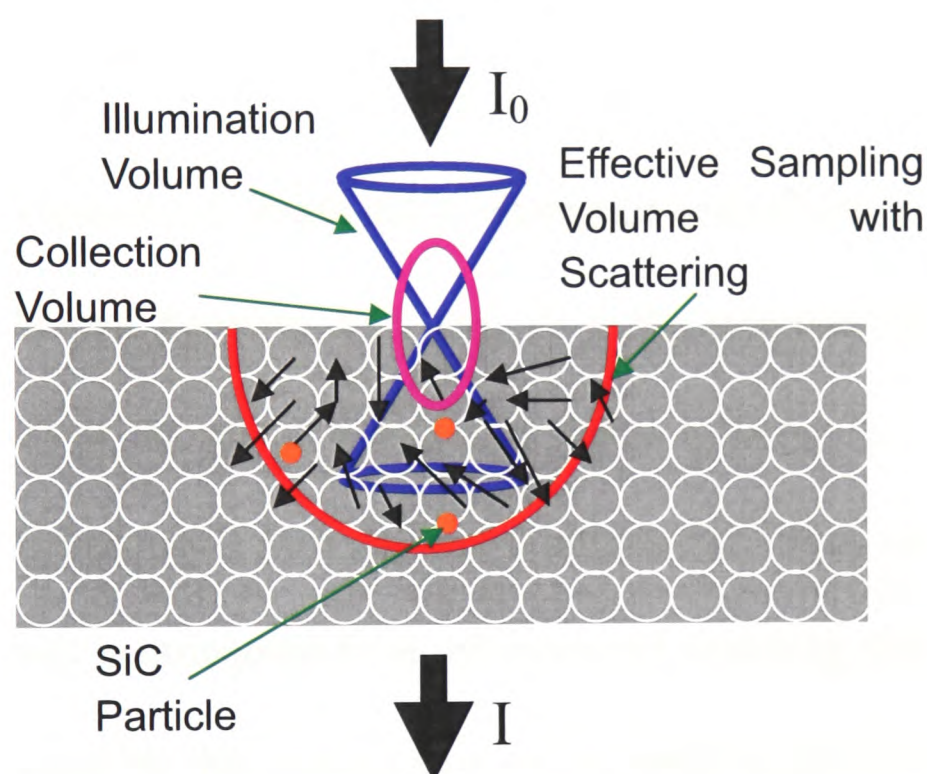


Fig 4.19 Schematic of scattering on polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites

The modified model to take scattering into account is based on the following assumptions:

- 1) Due to the absorption, the laser penetration depth in polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites is small compared to that in transparent materials. Hence the resolution broadening with depth is ignored. For the same reason, the difference of illuminated intensity at different depth is also ignored.

2) Assume the scattering and absorption follow the same light path and the scattering coefficient is the same as the absorption coefficient. This is actually not realistic but using two separate absorption coefficients would make the fitting unstable. If the absorption is strong enough (short absorption distance), the absorption (scattering) coefficient would not affect the shape of the curve much. The combined contribution from reflection and scattering to the collected intensity is described by Equation 4.17:

$$I(z_0) = \int_0^{\infty} \left[ P_6 \exp(-2P_4 z) \frac{P_1}{P_1^2 + (z - P_3(z_0 - P_5))^2} + P_7 \exp(-2P_4 z) \frac{P_2}{P_2^2 + (z - P_3(z_0 - P_5))^2} \right] dz \quad (4.17)$$

where  $P_1$  is the response half length due to absorption and reflection, which is known from sapphire through Equation 4.13,  $\mu\text{m}$ ;  $P_2$  is the response half length due to scattering,  $\mu\text{m}$ ;  $P_3$  is the effective refractive index, unitless;  $P_4$  is the absorption (scattering) coefficient,  $\mu\text{m}^{-1}$ ;  $P_5$  is the zero depth position offset,  $\mu\text{m}$ ;  $P_6$  and  $P_7$  are the relative intensity contributions from reflection and scattering, unitless. Clearly,  $P_4$  and  $P_7$  are connected but this connection is not currently understood so that the two parameters were fitted separately. Similar constraints apply to  $P_2$  and its connection with other parameters but it was also fitted separately.

In Chapter 5 and Chapter 6, Equation 4.17 will be convoluted with the stress function to predict the apparent stresses that are actually measured. Equation 4.17 will be cited as double-Lorentzian function in those chapters for simplicity. The fitting results for  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites are given in Fig 4.20, 4.21, 4.22 and 4.23. The fitted absorption coefficients are listed in Table 4.9. The absorption coefficients of



$\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites are greater than that of alumina, and the more SiC added, the greater the absorption coefficient.

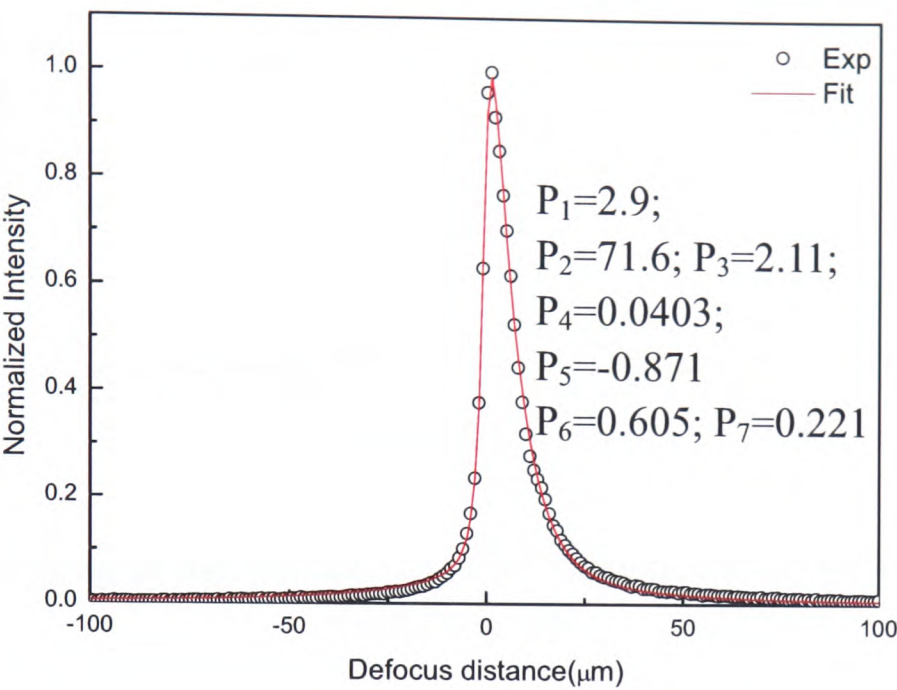


Fig 4.20 Fitting of the intensity-defocus distance curves for  $5.6\ \mu\text{m}\ \text{Al}_2\text{O}_3$

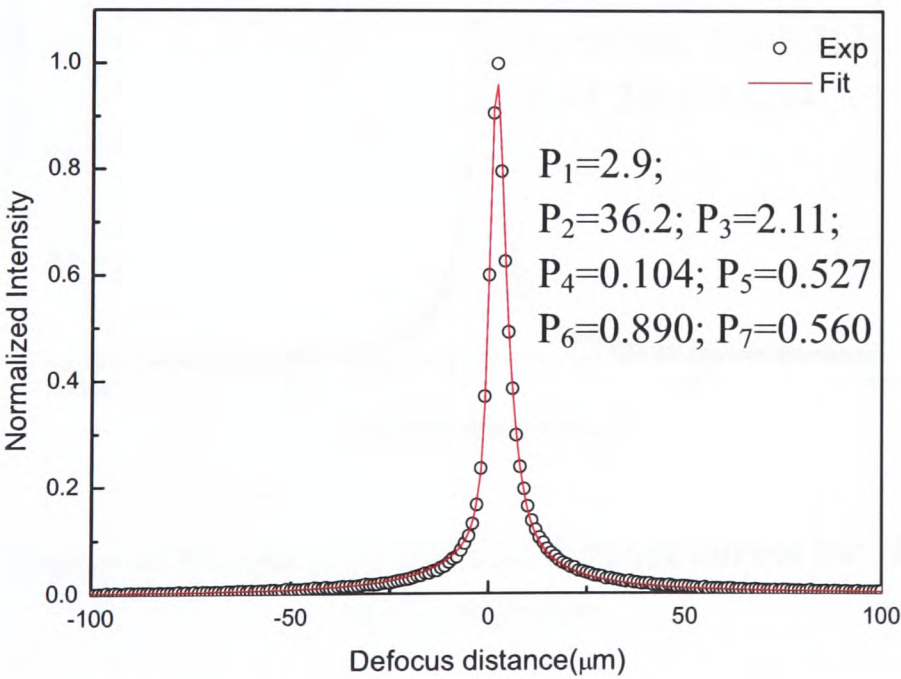


Fig 4.21 Fitting of the intensity-defocus distance curves for 2vol% SiC nanocomposite

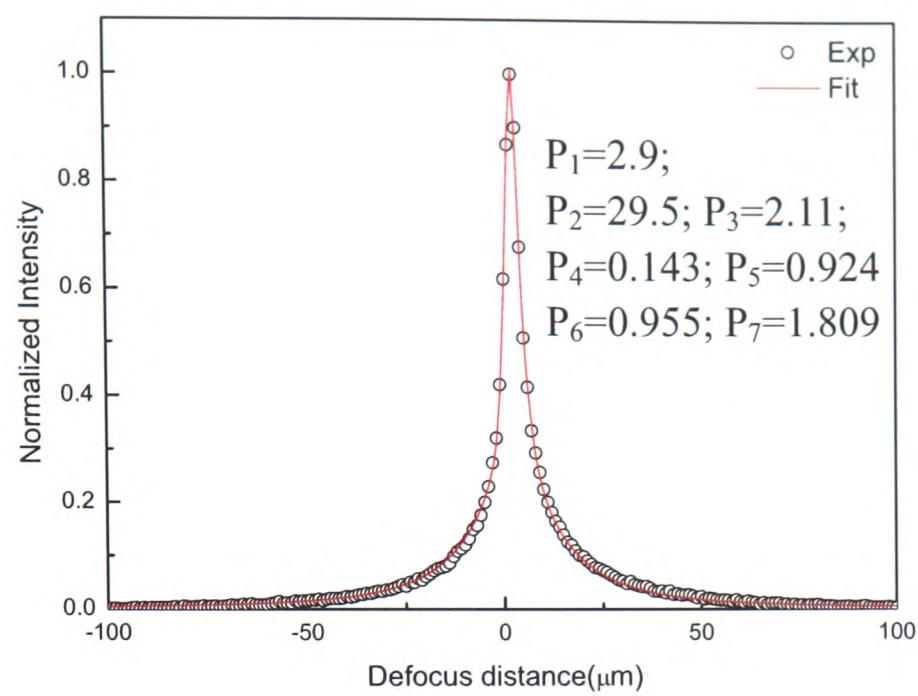


Fig 4.22 Fitting of the intensity-defocus distance curves for 5vol% SiC nanocomposite

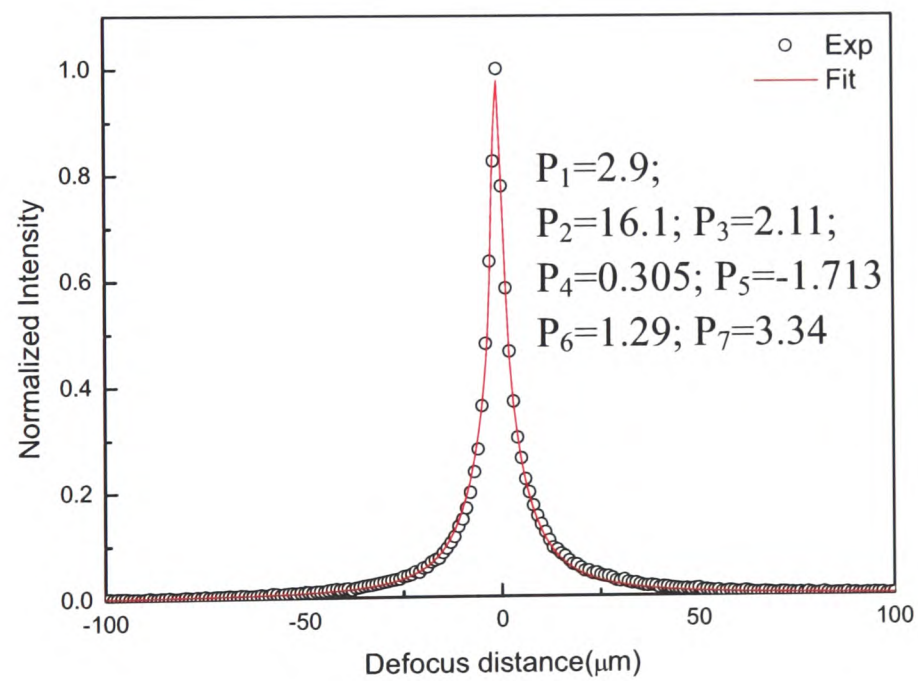


Fig 4.23 Fitting of the intensity-defocus distance curves for 10vol% SiC nanocomposite

Table 4.9 Fitted absorption coefficients for Al<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites

| Material                       | Absorption Coefficient (μm <sup>-1</sup> ) |
|--------------------------------|--------------------------------------------|
| Al <sub>2</sub> O <sub>3</sub> | 0.0403                                     |
| 2 vol.% SiC                    | 0.104                                      |
| 5 vol.% SiC                    | 0.143                                      |
| 10 vol.% SiC                   | 0.305                                      |

4.4 Summary

1) The axial resolution was measured from the FWHM of the intensity-defocus distance profiles. The best measured axial resolution of the Raman microscope used throughout the work is about 2 μm using 100× objective lens and confocal setup. The measured best lateral resolution is 1.5 μm.

2) Mathematical descriptions were proposed for the axial probe response in different materials, taking into account refraction, absorption, and scattering. They fit the experimental results very well. Useful information such as absorption coefficients were drawn from the fittings. The PRFs will be used to convolute with stress functions to predict sampling volume averaged stresses that are experimentally measured.

## References

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## Chapter 5

### Grinding and Polishing of Polycrystalline Alumina

#### 5.1 Introduction

Machining is normally the finishing procedure for fabrication of most structural ceramics like alumina. The introduction of near-surface damage, such as cracking, grain pullout or plastic deformation is almost unavoidable. As a result, the surface quality and the mechanical properties are affected by the machining process. The investigation into grinding and polishing of alumina ceramics became more interesting after researchers found that the addition of a small amount of sub-micron sized SiC significantly improves the surface quality (much fewer pullouts) and also improves the strength of pure alumina ceramics [1-5]. It is believed by some researchers that the difference in residual stresses between alumina and alumina/SiC nanocomposites is the strengthening mechanism behind it [5-7]. In this chapter, the residual stresses of polycrystalline alumina of four different grain sizes were measured by  $\text{Cr}^{3+}$  fluorescence microscopy, after 45  $\mu\text{m}$  grit diamond grinding and 3  $\mu\text{m}$  grit polishing. A model convoluting the probe resolution function (PRF) and the stress function was proposed to compare the experimental results with the theoretical prediction. In the following chapter, the residual stresses in alumina/SiC nanocomposites are investigated using the same method. Comparison of results from pure alumina and alumina/SiC nanocomposites can hopefully answer one question: is it true that the strengthening mechanism in  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites can be

attributed to surface residual stresses? In addition, Wu et al. have reported that the stressed layer is thicker in nanocomposites than that in alumina [8]. This will also be investigated in the next chapter.

## **5.2 Experimental Results**

The grinding and polish procedures have been introduced in Chapter 3. Depth scans through the ground regions and the pullout regions were carried out on four polycrystalline aluminas. As described in Chapter 4, the experimentally measured wavenumber shifts (hence the stress) at some distance from the surface are the averaged value over the sampling volume. Physically, they are the convoluted results from resolution function and the actual wavenumber shift at any point. The resolution function has already been determined in Chapter 4. A model predicting the actual stress distribution will be proposed in the following section, which includes some unknown parameters representing the thickness of the stressed layer and the magnitude of the residual stress. These parameters can be determined by fitting the convoluted prediction to the experimentally measured results.

### **5.2.1 Grinding**

The processing conditions of the four polycrystalline aluminas used for 45  $\mu\text{m}$  grit diamond grinding is listed in Table 3.1 of Chapter 3. The optical micrographs of the ground surface are shown in Fig 5.1. They are typical of ground polycrystalline alumina as intergranular fractures or grain dislodgements (dark regions in the optical



images) are clearly seen on the surfaces. The mean size of the pullouts and the pullout fraction are listed in Table 5.1.

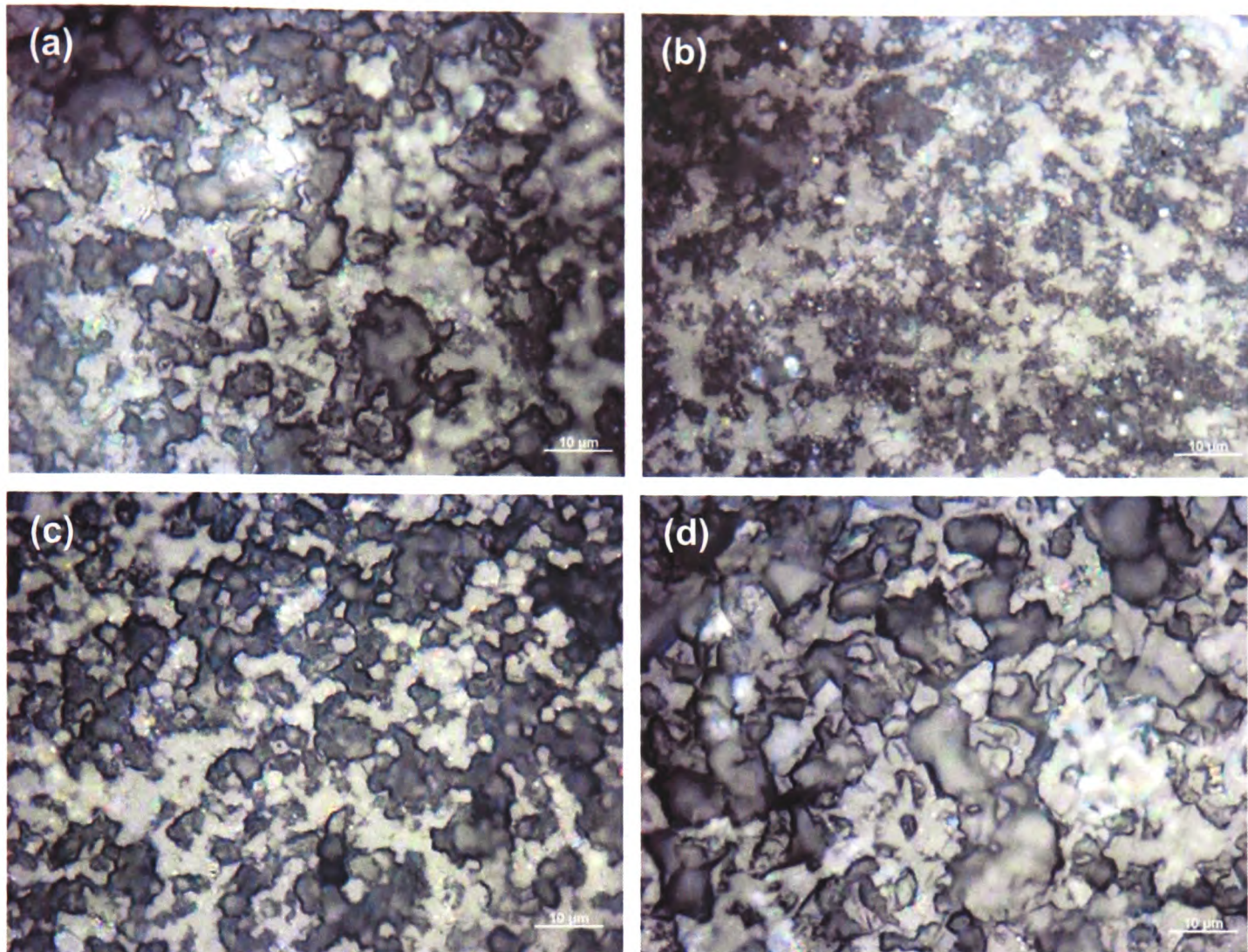


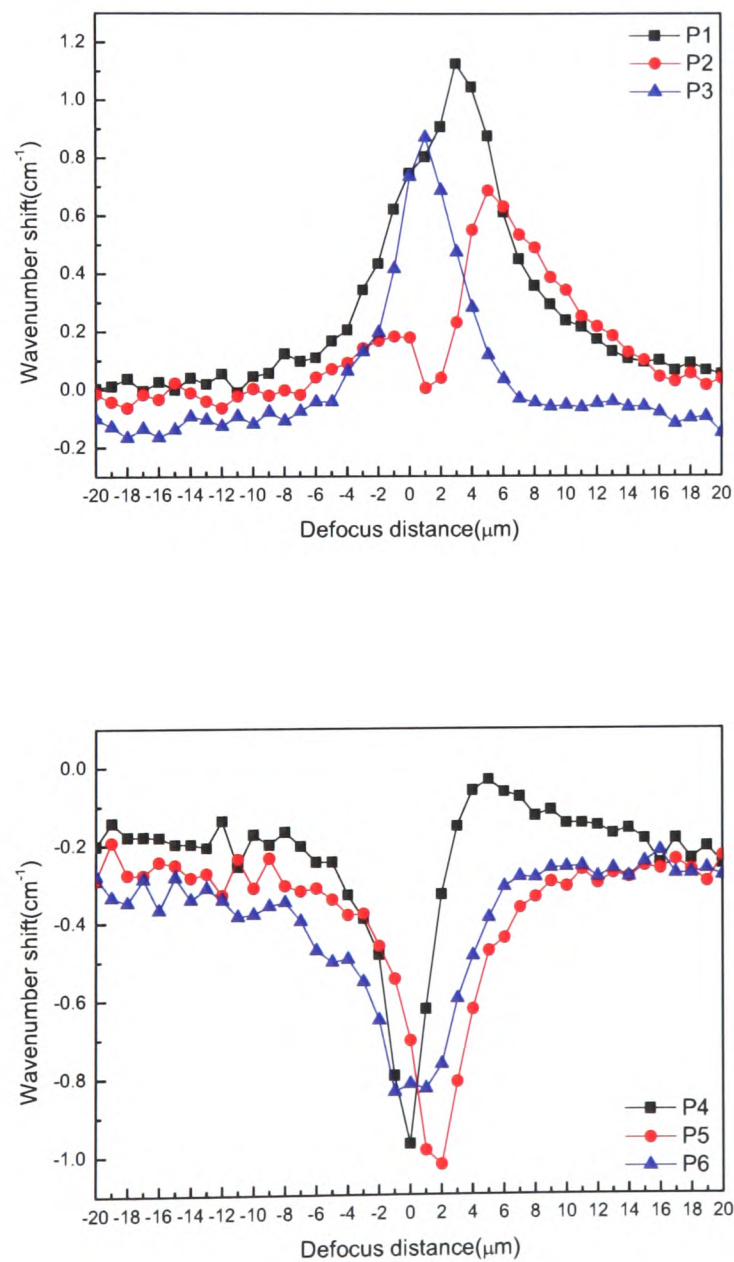
Fig 5.1 Optical micrographs of 45 grit diamond ground surfaces of polycrystalline aluminas. (a)  $G=2.4\text{ }\mu\text{m}$ ; (b)  $G=4.2\text{ }\mu\text{m}$ ; (c)  $G=5.6\text{ }\mu\text{m}$ ; (d)  $G=8.0\text{ }\mu\text{m}$ .  $G$  is the average grain size. Shiny regions and dark regions in the optical micrographs correspond to ground surfaces and pullouts.



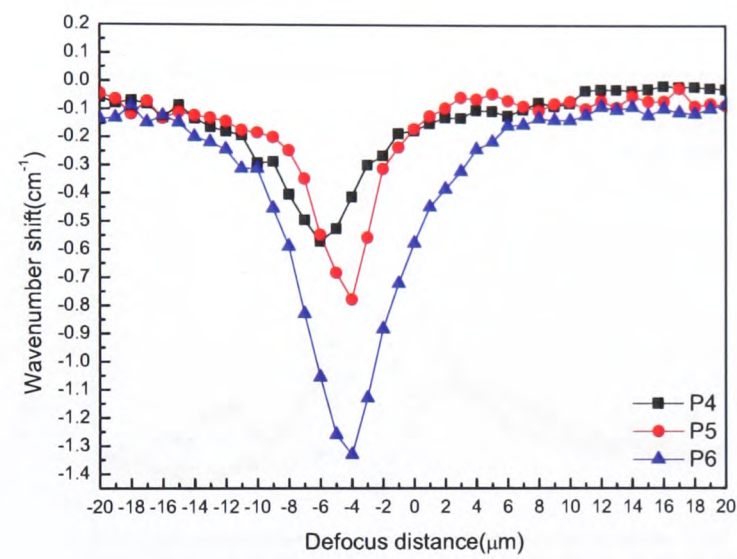
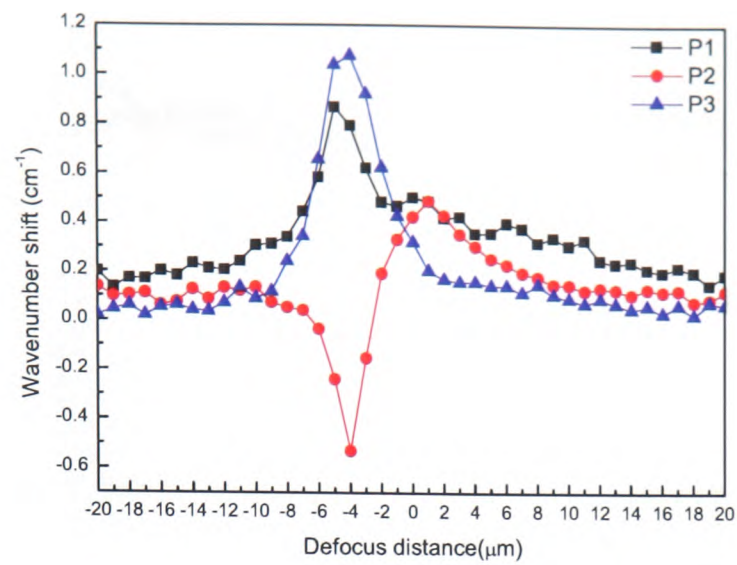
Table 5.1 List of the mean pullout size, area fraction of pullout and the mean ground island size of 45  $\mu\text{m}$  grit ground polycrystalline aluminas

| G                 | Mean Pullout Size, $p$ | Area Fraction of | Mean Ground Island Size, $^1s$ |
|-------------------|------------------------|------------------|--------------------------------|
| ( $\mu\text{m}$ ) | ( $\mu\text{m}$ )      | Pullout (%)      | ( $\mu\text{m}$ )              |
| 2.4               | 6.3                    | 54.3             | 5.3                            |
| 4.2               | 8.4                    | 50.9             | 8.1                            |
| 5.6               | 7.1                    | 58.2             | 5.1                            |
| 8.0               | 5.6                    | 51.8             | 5.2                            |

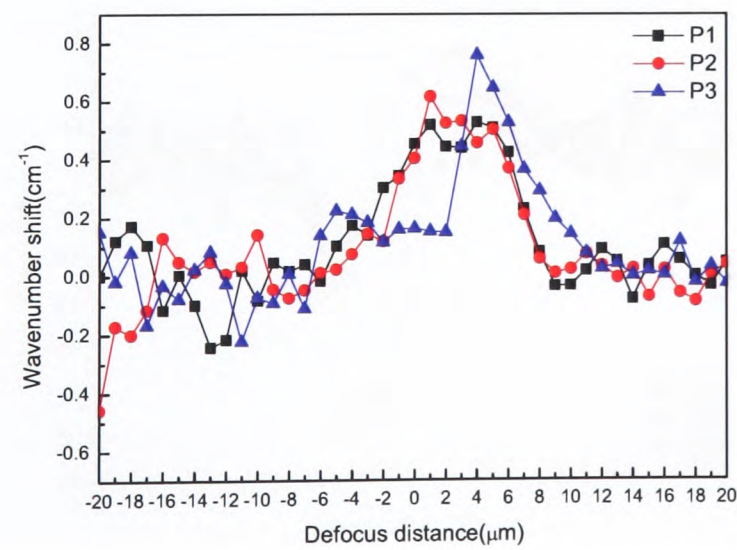
$^1s$  is calculated from  $p$  and the area fraction of pullout:  $pullout\%=p/(p+s)$

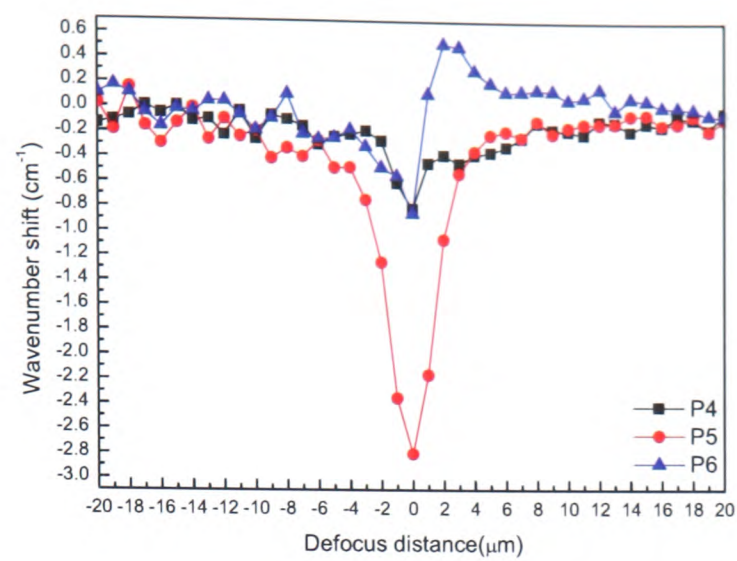


(a)

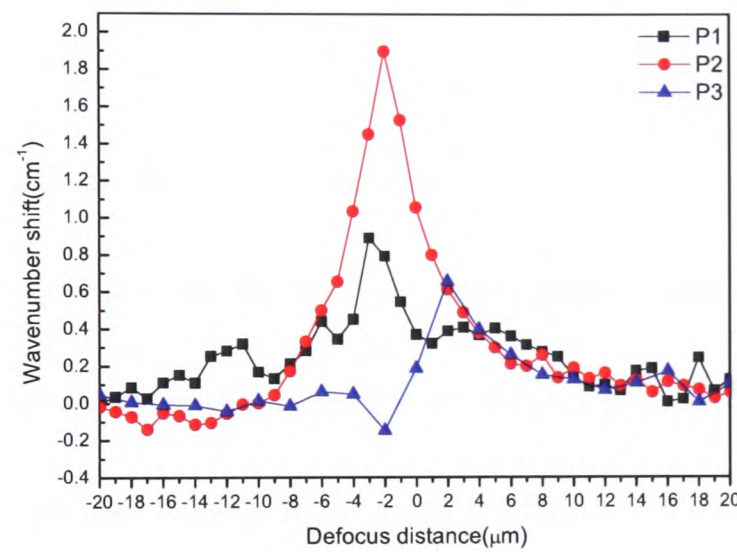


(b)





(c)



(d)

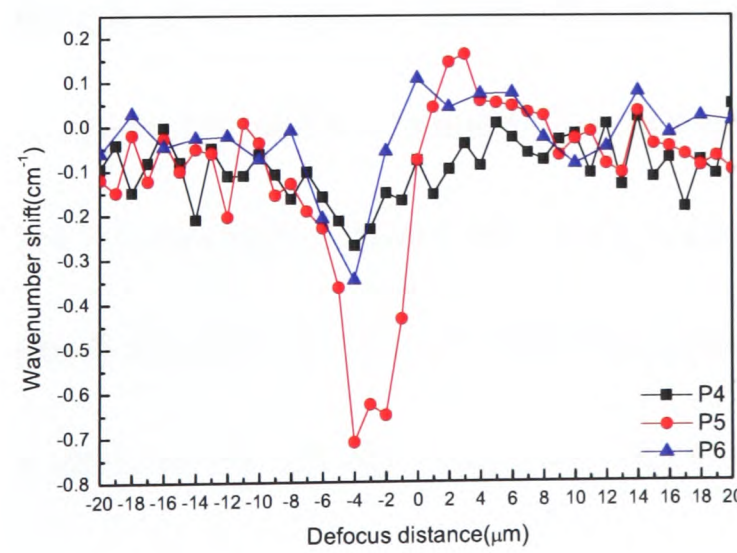


Fig 5.2 Depth profiles of four different grain-sized alumina, (a)  $G=2.4\text{ }\mu\text{m}$ ; (b)  $G=4.2\text{ }\mu\text{m}$ ; (c)  $G=5.6\text{ }\mu\text{m}$  (d)  $G=8.0\text{ }\mu\text{m}$ . P1, P2 and P3 are through ground regions and P4, P4 and P6 are through pullout regions.

The depth profiles of the four polycrystalline aluminas are shown in Fig 5.2, with the abscissa being the distance from surface (the laser was focused on surface at the beginning) and the ordinate being the R1 wavenumber shift relative to the reference position, i.e., the residual free position. A positive defocus distance means the laser is focused below the surface and negative distance means the laser is actually focusing above the surface. Although in some papers [9, 10], the R2 peak is chosen to characterize the wavenumber shift, as R2 peak shift has a better linearity to stress (piezo-spectroscopic effect), wavenumber shifts from the R1 peaks are chosen here as the R1 peak has a higher intensity compared with R2, and the complexity of the R lines due to the superimposition of different stress states also makes the weaker R2 position less reliable. This will be further illustrated in the following section. The reference position was determined from the averaged R1 positions (spectra collected at 20 random points) on the 1  $\mu\text{m}$  grit polished surface.

Three depth scans through ground regions (points P1, P2, and P3) and three depth scans through pullout regions (points P4, P5 and P6) were made on all four materials, which are shown on the left and right columns of Fig 5.2, respectively. Interestingly, a clear trend can be found from depth profiles on ground regions and pullout regions: a positive wavenumber shift on the ground region and negative shift on the pullout region. The magnitude of the shift was different case by case. Apparently, the surface residual stress on ground surfaces is dependent on the detail of the surface fracture and grain pullout in the local environment. The discrepancy of depth scan through P3



in Fig 5.2 (b) can be attributed to the low depth of field of the optical microscope: it was sometimes difficult to recognize pullout and ground region, although great care had been taken.

### 5.2.2 Polishing

For comparison, the same four different grain sized polycrystalline aluminas were polished with 3  $\mu\text{m}$  grit diamond and depth scans were made on the polished surfaces. The polished surfaces are much smoother compared with the 45  $\mu\text{m}$  grit ground surfaces, with much fewer pullouts (see Fig 5.3). The mean size of the pullouts and the pullout fraction are listed in Table 5.2.

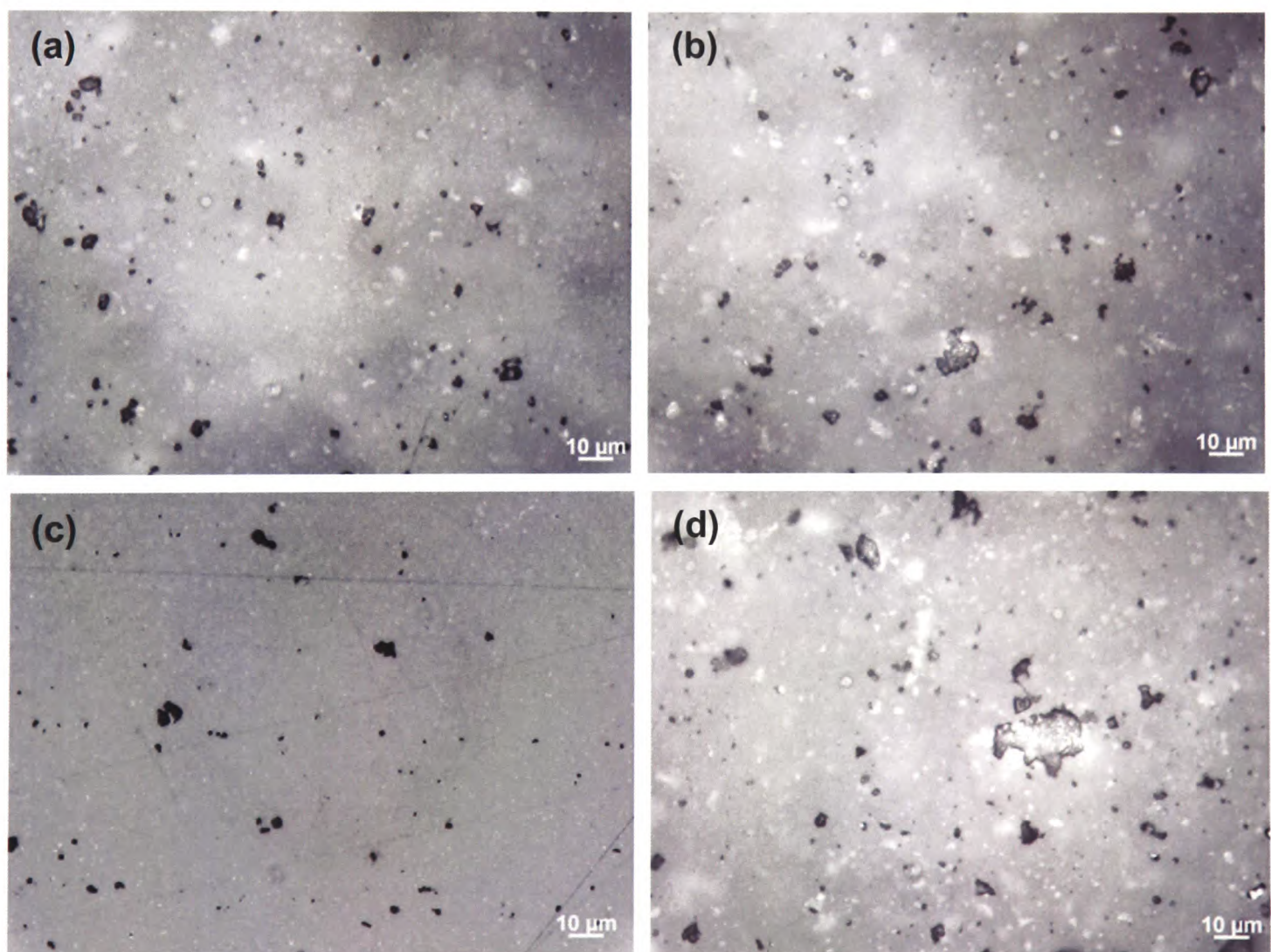
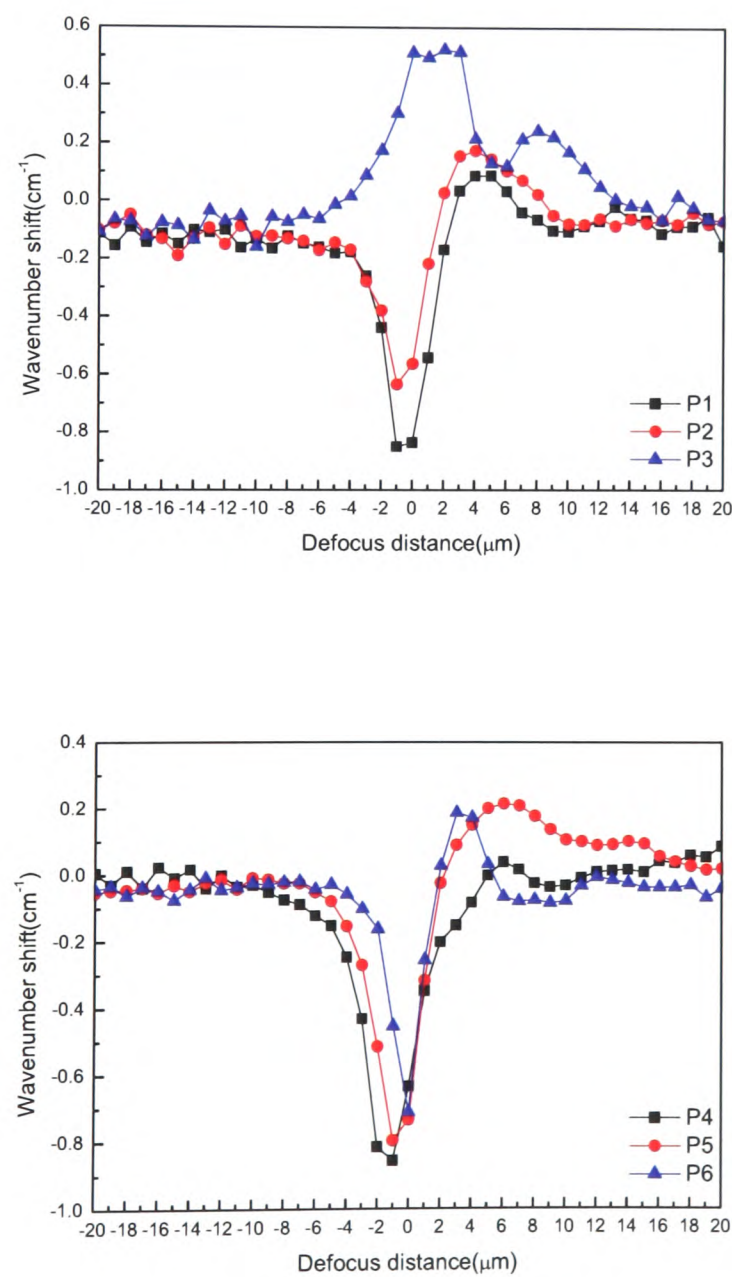


Fig 5.3 Optical micrographs of polished surfaces of polycrystalline alumina  
(a)  $G=2.4\ \mu\text{m}$ ; (b)  $G=4.2\ \mu\text{m}$ ; (c)  $G=5.6\ \mu\text{m}$ ; (d)  $G=8.0\ \mu\text{m}$

Table 5.2 List of the mean pullout size, area fraction of pullout and the mean ground island size of 3 μm grit polished polycrystalline aluminas

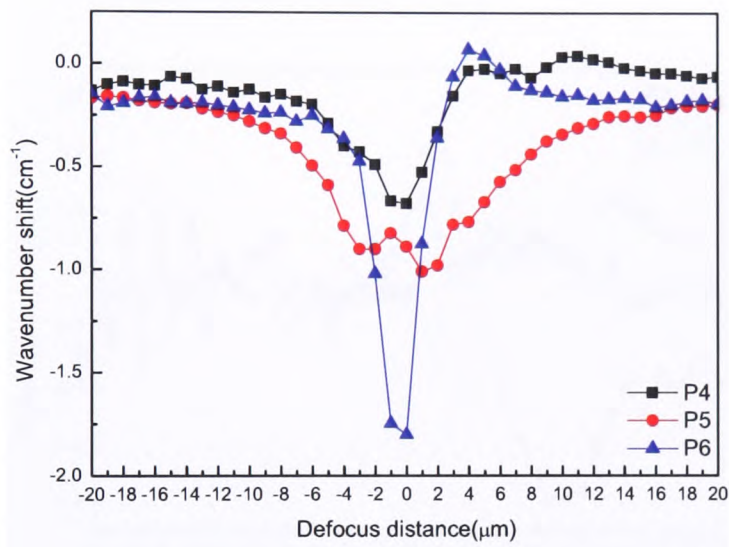
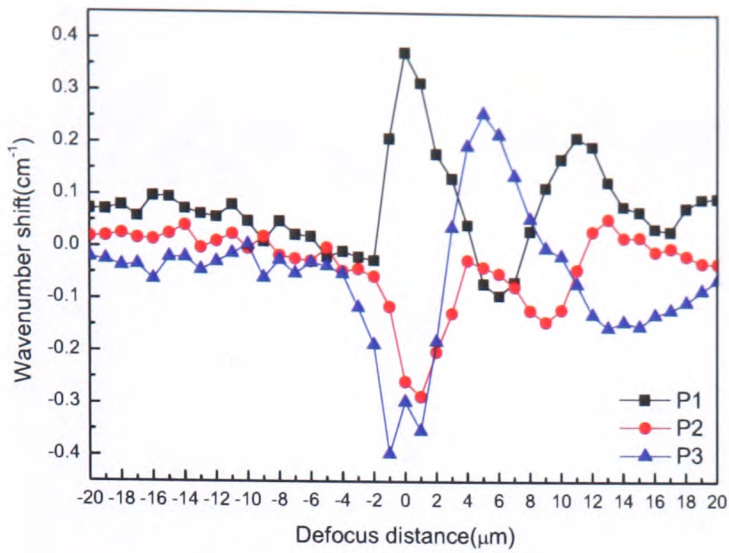
| G    | Mean Pullout Size, <i>p</i> | Area Fraction of | Mean Ground Island Size, <sup>1</sup> <i>s</i> |
|------|-----------------------------|------------------|------------------------------------------------|
| (μm) | (μm)                        | Pullout (%)      | (μm)                                           |
| 2.4  | 2.0                         | 1.72             | 114.5                                          |
| 4.2  | 2.4                         | 2.48             | 94.5                                           |
| 5.6  | 1.6                         | 0.86             | 184.6                                          |
| 8.0  | 2.3                         | 4.16             | 53.0                                           |

<sup>1</sup>*s* is calculated from *p* and the area fraction of pullout: *pullout*%=*p*/(*p*+*s*)

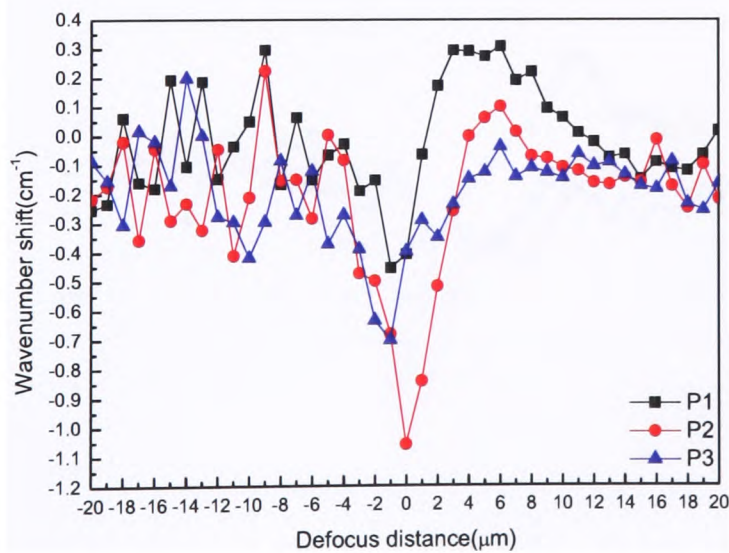


(a)

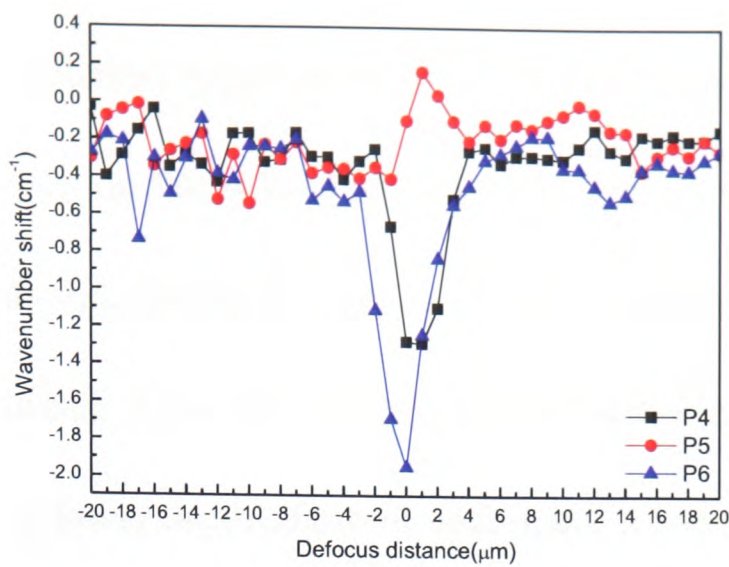




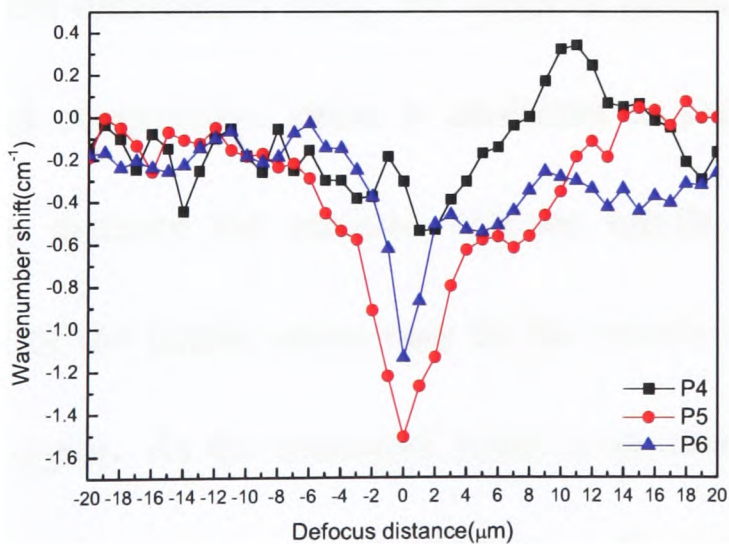
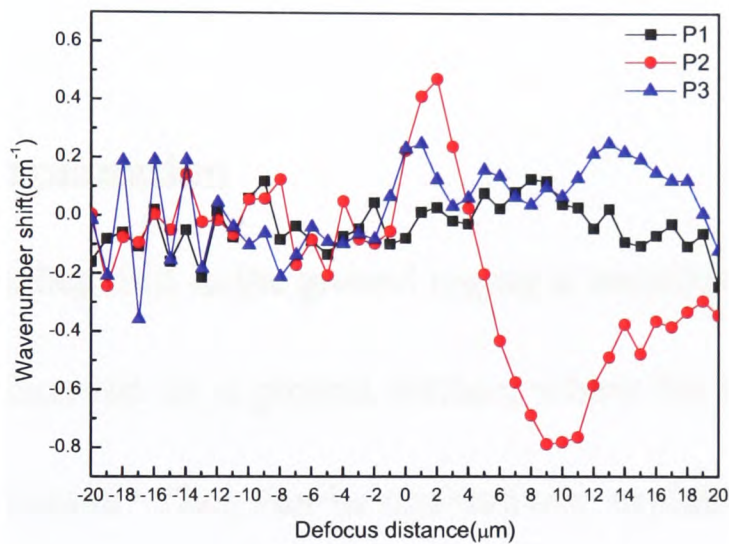
(b)







(c)



(d)

Fig 5.4 Depth profiles of four different grain-sized alumina, (a)  $G=2.4\text{ }\mu\text{m}$ ; (b)  $G=4.2\text{ }\mu\text{m}$ ; (c)  $G=5.6\text{ }\mu\text{m}$  (d)  $G=8.0\text{ }\mu\text{m}$ . P1, P2 and P3 are through ground regions and P4, P4 and P6 are through pullout regions.

The depth profiles for the four polycrystalline aluminas are given in Fig 5.4, where P1, P2 and P3 are on the polished region while P4, P5 and P6 are on the pullout region. The compressive stresses on the pullout regions were still observed, similar to those shown in Fig 5.2. Although the depth profiles from the polished regions indicate that stresses exist at the surface, their sign and magnitude are less consistent, which could be due to the fact that a lower depth of plastic deformation and/or the larger size of the ground island reduce the measurable stress. This will be explained further in the following sections.

### 5.3 Qualitative Explanation

The positive wavenumber shift in the ground region is surprising at first thought: why are tensile stresses observed on a ground surface, where the stresses are commonly assumed to be compressive? They can be qualitatively explained from the viewpoint of sampling volume, as illustrated in Fig 5.5 (a). In Wu's paper [8], a model was suggested for the stress distribution along the depth of ground material (see Fig 5.5 (b)). A large constant compressive stress is predicted to some distance below the surface; beyond this distance the stresses will be tensile, with a much lower magnitude. This part of the tensile stress may be the source of the observed tensile stress in the ground region. As the measured stress is an averaged value among the sampling volume, it is actually a matter of where the collected signal comes from. If most of the signal comes from the tensile region, the observed stress would be tensile; otherwise it would be compressive. The validity of this idea will be tested and

developed in the following sections.

It has to be pointed out now that the peak positions used in Fig 5.2 and Fig 5.4 are from fitting spectra with a mixed Gaussian and Lorentzian function, and only two peaks are defined: one for R1 and one for R2. However, it is found that near the surface, the spectra cannot be fitted well with only two peaks. The misfit on the tail of the R1 side (see Fig 5.15 (b)) indicates there exists at least an additional stress state. Similar bad fitting can be observed from spectra collected around the indent center in indentation experiments, which will be discussed further in Chapter 7. Before the curve fitting problem for spectra near the ground surface is explained, a dislocation model predicting the stress distribution on the ground material is introduced, which will also help to understand the curve fitting.

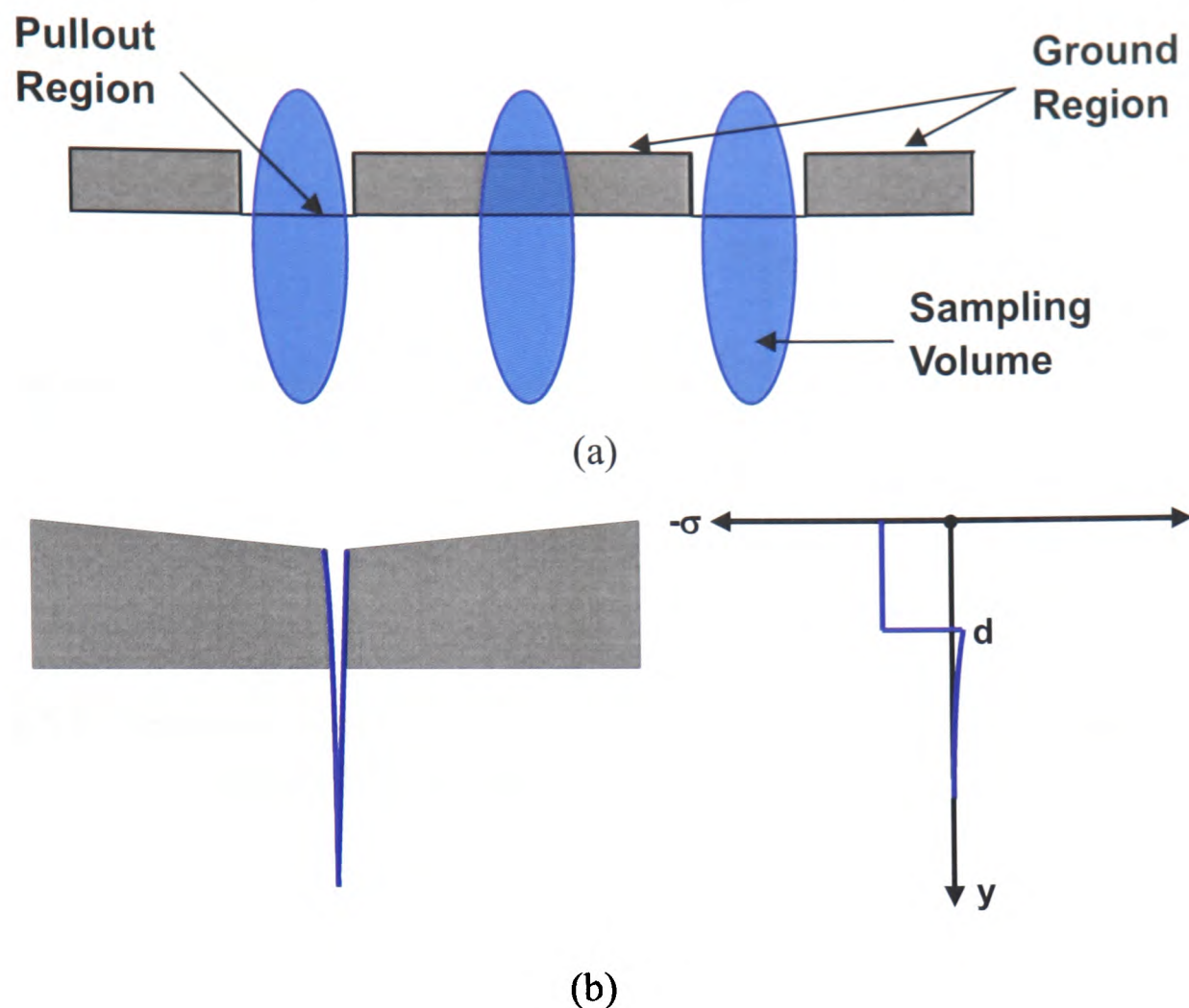


Fig 5.5 (a) Illustration of sampling volume for grinding and pullout. (b) Redrawn stress model predicted along depth direction on a ground material, proposed by Wu [8]. On the left a surface crack is shown; on the right,  $y$  is the coordinate of depth direction and  $d$  is the depth the stressed layer.

## 5.4 Stress Modeling

The displacement of material during grinding and polishing can be simulated by an array of edge dislocations, where extra material is inserted above the undeformed material. Since the stress field around an edge dislocation has been well established, the residual stress distribution on the ground surface can be predicted. The model starts from a single edge dislocation, with emphasis on satisfying the boundary conditions such as zero normal and shear force on the surface. The model then extends to an array of homogeneously distributed dislocations aligned along a plane at a fixed distance below the surface, and finally goes to an island model which takes into account the pullout, assuming the pullout region is dislocation free.



## 5.4.1 Single Dislocation

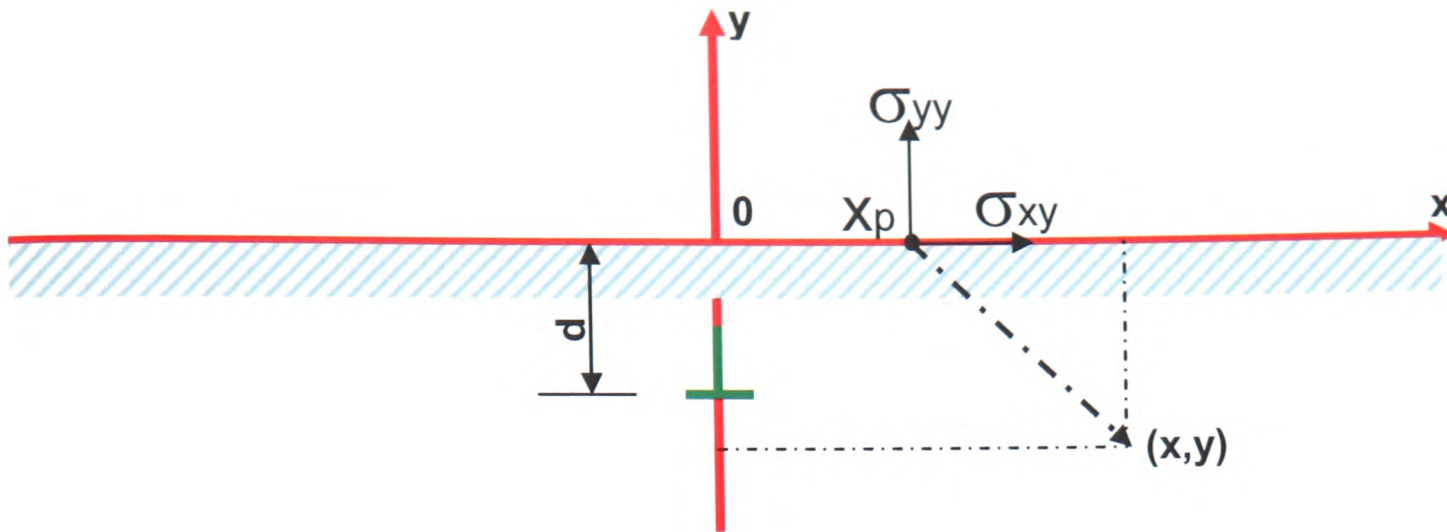


Fig 5.6 Schematic of normal stress ( $\sigma_{yy}$ ) and shear stress ( $\sigma_{xy}$ ) at point  $(x_p, 0)$  produced by an edge dislocation situated on  $(0, -d)$

Assume the surface of the ground material is on the x-z plane and normal to the y axis, as illustrated in Fig 5.6. An edge dislocation is situated at a distance  $d$  from the surface ( $x=0, y=-d$ ).  $d$  is the depth of the dislocation in the model. In reality, it gives a measure of the depth to which the plastic deformation has occurred during grinding.

The stress components around an edge dislocation in an infinite piece of material have already been established [11], with the form of:

$$\begin{aligned}
 \sigma_{xx} &= -D(y+d) \frac{3x^2 + (y+d)^2}{(x^2 + (y+d)^2)^2} \\
 \sigma_{yy} &= D(y+d) \frac{x^2 - (y+d)^2}{(x^2 + (y+d)^2)^2} \\
 \sigma_{xy} &= Dx \frac{x^2 - (y+d)^2}{(x^2 + (y+d)^2)^2} \\
 D &= \frac{Gb}{2\pi(1-\nu)}
 \end{aligned} \tag{5.1}$$

where  $D$  is a constant defined by  $G$ , shear modulus;  $b$ , the magnitude of the Burgers vector; and  $\nu$ , Poisson's ratio.

At any point on the surface ( $x=x_p$ ,  $y=0$ ), the stress components produced by the edge dislocation at  $(0, -d)$  are

$$\begin{aligned}\sigma_{xx}(x_p, 0) &= -\frac{Dd(3x_p^2 + d^2)}{(x_p^2 + d^2)^2} \\ \sigma_{yy}(x_p, 0) &= \frac{Dd(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \\ \sigma_{xy}(x_p, 0) &= \frac{Dx_p(x_p^2 - d^2)}{(x_p^2 + d^2)^2}\end{aligned}\tag{5.2}$$

According to Equation 5.2,  $\sigma_{yy}$  and  $\sigma_{xy}$  are not zero on the surface, which violates the boundary condition. The free surface (see Fig 5.7) can be created by cutting off the upper semi-infinite space above the surface and applying the surface tractions at the same time. To satisfy the boundary condition on the free surface, two virtual stresses,  $-\sigma_{yy}$  and  $-\sigma_{xy}$  are applied at the surface to counter-act  $\sigma_{yy}$  and  $\sigma_{xy}$ , as shown in Fig 5.7 and Fig 5.8. The stress field induced by them can be calculated assuming a concentrated force along a line of straight boundary, for which a known solution already exists [12]. The total stress components  $\sigma_{yy}$  and  $\sigma_{xy}$  will then be zero adding up contributions from the edge dislocation and the two concentrated forces, satisfying the boundary conditions. The stress field generated by an edge dislocation in a free surface condition is then established.



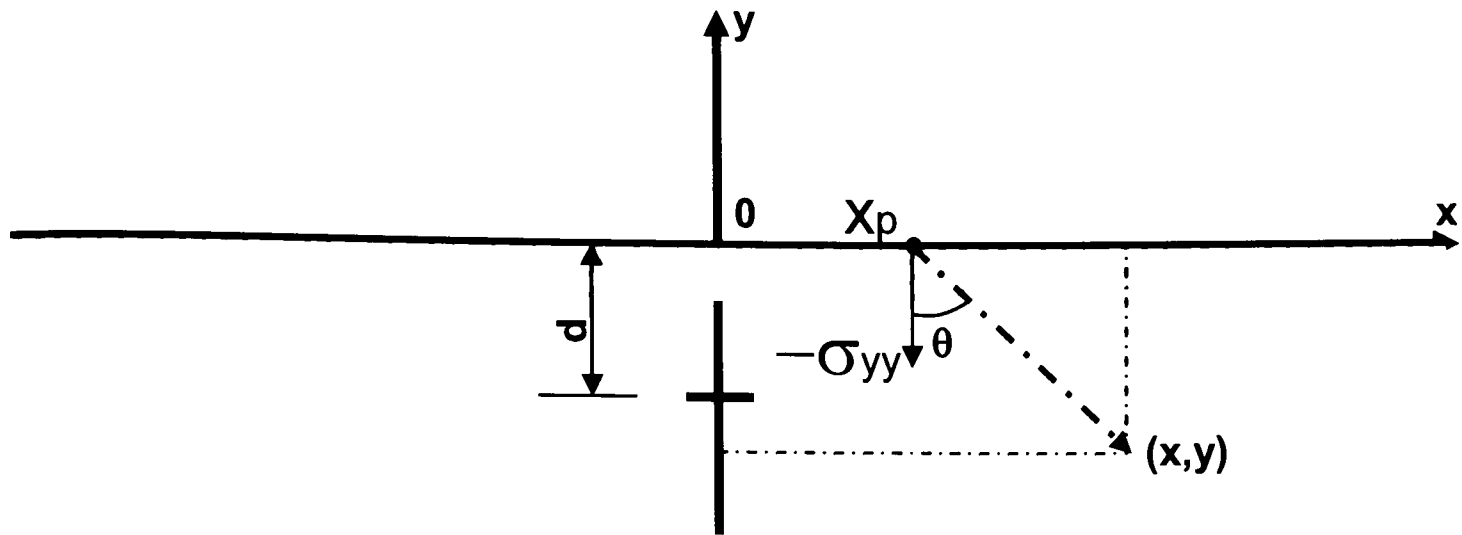


Fig 5.7 Schematic of an imagined normal force to balance  $\sigma_{yy}$  in Fig 5.6

The stress components produced by the normal surface traction  $-\sigma_{yy}$  at any point below the surface  $(x, y)$  are as follows:

$$\sin \theta = \frac{x - x_p}{\sqrt{(x - x_p)^2 + y^2}}; \cos \theta = \frac{y}{\sqrt{(x - x_p)^2 + y^2}}$$

$$\sigma_{xx} = \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dd(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin^2 \theta \cos^2 \theta dx_p$$

$$\sigma_{yy} = \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dd(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \cos^4 \theta dx_p$$

$$\sigma_{xy} = \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dd(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin \theta \cos^3 \theta dx_p$$
(5.3)

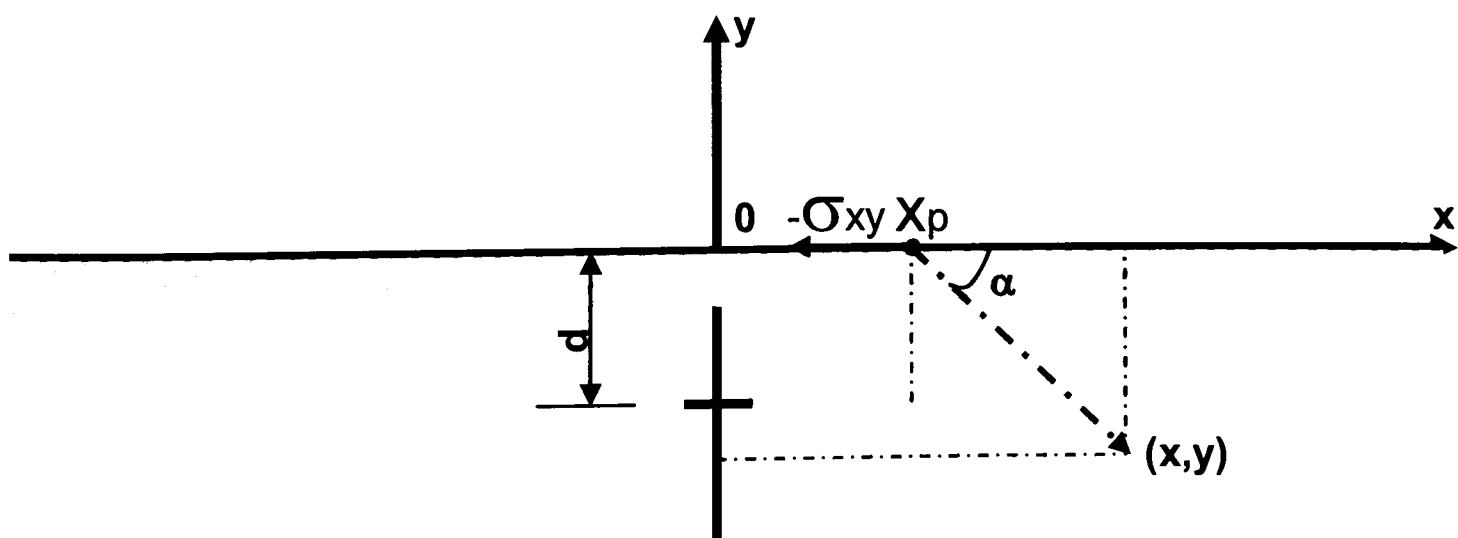


Fig 5.8 Schematic of an imagined shear force to balance  $\sigma_{xy}$  in Fig 5.6

Similarly, the stress components produced at (x,y) by  $-\sigma_{xy}$  are as follows:

$$\begin{aligned}
 \sin \alpha &= \frac{y}{\sqrt{(x-x_p)^2 + y^2}}; \cos \alpha = \frac{x-x_p}{\sqrt{(x-x_p)^2 + y^2}} \\
 \sigma_{xx} &= \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dx_p(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin \alpha \cos^3 \alpha dx_p \\
 \sigma_{yy} &= \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dx_p(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \cos \alpha \sin^3 \alpha dx_p \\
 \sigma_{xy} &= \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dx_p(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin^2 \alpha \cos^2 \alpha dx_p
 \end{aligned} \tag{5.4}$$

Adding up the same components from Equations 5.1, 5.3 and 5.4,

$$\begin{aligned}
 \sigma_{xx} = f(x, y) &= \frac{-D(y+d)[3x^2 + (y+d)^2]}{[x^2 + (y+d)^2]^2} + \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dd(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin^2 \theta \cos^2 \theta dx_p \\
 &+ \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dx_p(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin \alpha \cos^3 \alpha dx_p \\
 &= \frac{-D(y+d)[3x^2 + (y+d)^2]}{[x^2 + (y+d)^2]^2} + \frac{-Dd[x^4 - (d-y)(d^3 - (d^2 + 6x^2)y - dy^2 + y^3)]}{[x^2 + (d-y)^2]^3} \\
 &+ \frac{D[-4dx^2(d^2 + x^2) + 3(d^2 + x^2)^2 y - 2d(5d^2 + 3x^2)y^2 + 4(3d^2 + x^2)y^3 - 6dy^4 + y^5]}{[x^2 + (d-y)^2]^3} \\
 \sigma_{yy} = g(x, y) &= \frac{D(y+d)[x^2 - (y+d)^2]}{[x^2 + (y+d)^2]^2} + \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dd(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \cos^4 \theta dx_p \\
 &+ \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dx_p(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \cos \alpha \sin^3 \alpha dx_p \\
 &= \frac{D(y+d)[x^2 - (y+d)^2]}{[x^2 + (y+d)^2]^2} + \frac{Dd[-x^4 + (d-y)(d^3 - 5d^2 y + 6x^2 y + 7dy^2 - 3y^3)]}{[x^2 + (d-y)^2]^3} \\
 &+ \frac{-Dy[d^4 - 6d^2 x^2 + x^4 - 2d^3 y - y^4 + 2d(3x^2 y + y^3)]}{[x^2 + (d-y)^2]^3}
 \end{aligned}$$

$$\begin{aligned}
\sigma_{xy} = h(x, y) &= \frac{Dx[x^2 - (y + d)^2]}{[x^2 + (y + d)^2]^2} + \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dd(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin \theta \cos^3 \theta dx_p \\
&+ \int_{-\infty}^{+\infty} \frac{-2}{\pi y} \left[ -\frac{Dx_p(x_p^2 - d^2)}{(x_p^2 + d^2)^2} \right] \sin^2 \alpha \cos^2 \alpha dx_p \\
&= \frac{Dx[x^2 - (y + d)^2]}{[x^2 + (y + d)^2]^2} + \frac{-2Ddx[x^2 - 3(d - y)^2]y}{[x^2 + (d - y)^2]^3} \\
&+ \frac{Dx(d^4 - x^4 + 2d^3y - 2dx^2y - 6d^2y^2 + 2dy^3 + y^4)}{[x^2 + (d - y)^2]^3}
\end{aligned} \tag{5.5}$$

By replacing  $x=x$ ,  $y=0$  in Equation 5.5,  $\sigma_{yy}=0$ ,  $\sigma_{xy}=0$ . The boundary conditions are thus satisfied.

#### 5.4.2 Multiple Dislocations

If there is no pullout, the plastic deformation on the ground layer can be simulated by an array of continuously distributed dislocations aligned on the same distance,  $d$  from the surface, as shown in Fig 5.9. The introduction of the virtual dislocation layer aims to simulate the deformation in the grinding condition and to use the well-established stress field generated by the edge dislocation. A parallel set of edge dislocation is a simplification of the real deformation: the magnitude of displacement is higher on the surface and becoming less downwards. However, assuming the dislocations are on the same depth makes the modeling easier.

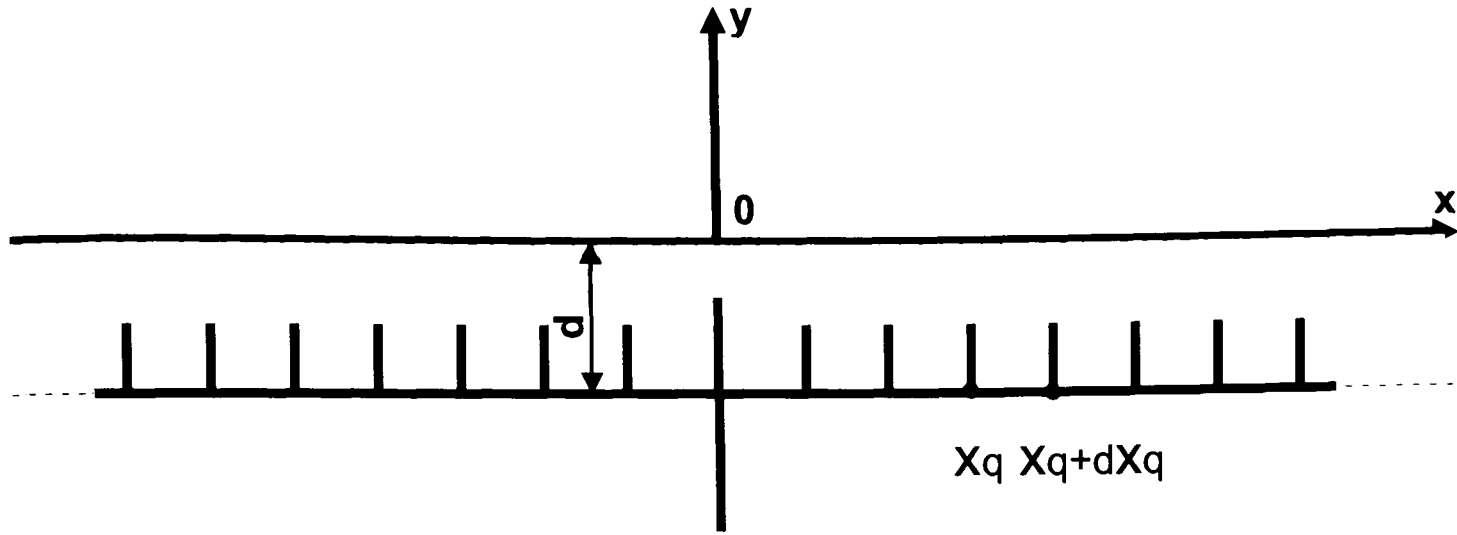


Fig 5.9 Schematic of an array of continuously distributed dislocations situated on  $y=-d$

At any position  $(x, y)$ , the stress components  $\sigma_{xx}$ ,  $\sigma_{yy}$  and  $\sigma_{xy}$  generated by a dislocation situated at  $x=0$ ,  $y=-d$  are described by Equation 5.5, except that  $D$  in

Equation 5.1 becomes  $D'dx_q = \frac{Gb'}{2\pi(1-\nu)}dx_q$ , where  $b'$  is the continuously

distributed Burgers vector per unit length in the  $x$  direction.  $b'$  determines the density of the continuously distributed dislocations and it equals to the stress-free mismatch in strain between the surface layer and the bulk. Take  $\sigma_{xx}$  as an example, the contribution,  $d\sigma_{xx}$ , of the dislocations in distance  $dx_q$  situated at  $(0, -d)$  to  $\sigma_{xx}$  at a point  $(x, y)$  is:

$$\begin{aligned}
 d\sigma_{xx} = f'(x, y) &= dx_q \frac{-D'(y+d)[3x^2 + (y+d)^2]}{[x^2 + (y+d)^2]^2} \\
 &+ dx_q \frac{-D'd[x^4 - (d-y)(d^3 - (d^2 + 6x^2)y - dy^2 + y^3)]}{[x^2 + (d-y)^2]^3} \\
 &+ dx_q \frac{D'[-4dx^2(d^2 + x^2) + 3(d^2 + x^2)^2y - 2d(5d^2 + 3x^2)y^2 + 4(3d^2 + x^2)y^3 - 6dy^4 + y^5]}{[x^2 + (d-y)^2]^3} \\
 &= f(x, y)dx_q
 \end{aligned}$$

(5.6)

Note that  $f(x, y)$  has the same form as in Equation 5.5 but the dimension is different.

Similarly, the contribution of the dislocations over distance  $dx_q$  situated at  $(x_q, -d)$  leads to a stress component at  $(x, y)$  of  $d\sigma_{xx} = f'(x - x_q, y) = f(x - x_q, y)dx_q$ . The contributions from all the dislocations on the array into the  $\sigma_{xx}$  component can be integrated as:

$$\sigma_{xx} = \int_{-\infty}^{+\infty} f'(x - x_q, y) = \int_{-\infty}^{+\infty} f(x - x_q, y)dx_q \quad (5.7)$$

$$\text{Similarly, } \sigma_{yy} = \int_{-\infty}^{+\infty} g(x - x_q, y)dx_q; \sigma_{xy} = \int_{-\infty}^{+\infty} h(x - x_q, y)dx_q$$

The integration was carried out using a commercial software package, Mathematica (Wolfram Research, USA). If  $D'$  and  $d$  are known, a three dimensional view of the stress components on x and y axes can be generated.

For the sake of illustration, results assuming  $D'=1$  GPa and  $d=5$   $\mu\text{m}$ ,  $\sigma_{xx}, \sigma_{yy}$  and  $\sigma_{xy}$  are shown in Fig 5.10.  $\sigma_{yy}$  and  $\sigma_{xy}$  are zero everywhere;  $\sigma_{xx}$  is negative and constant ( $\sigma_{xx}=-12.566$  GPa) when the depth is within the dislocation array ( $y>-d$ ) and is zero when  $y<-d$ . This is exactly what should be expected from the model.

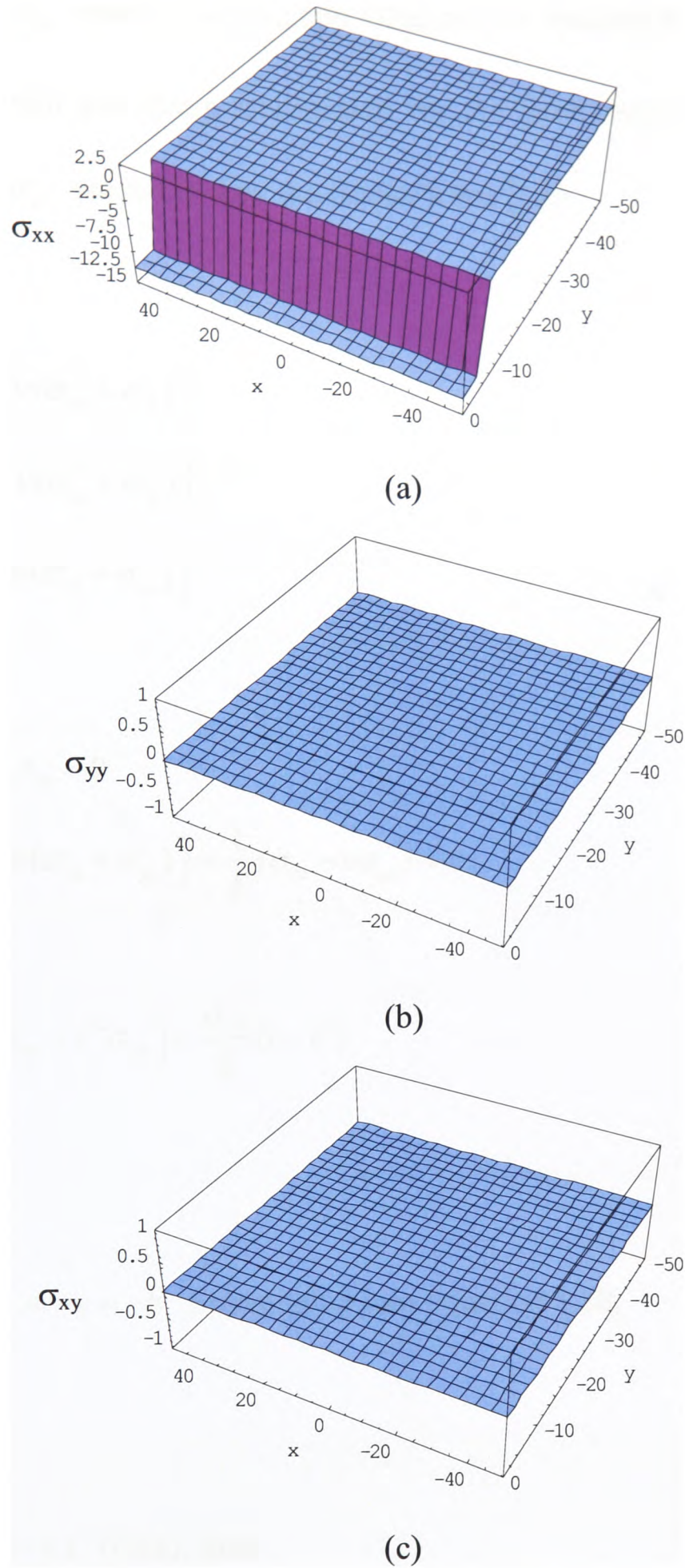


Fig 5.10 Stress components that are produced by the dislocation array shown in Fig 5.9. (a)  $\sigma_{xx}$ ; (b)  $\sigma_{yy}$ ; (c)  $\sigma_{xy}$ . The units of  $\sigma_{ij}$  are GPa and  $x$  and  $y$  are in  $\mu\text{m}$ .



The value of  $\sigma_{xx}$  when  $y \gg d$  (-12.566 GPa) can be checked by using basic elasticity theory. The strain and stress relationship for the three normal stresses components

$\sigma_{xx}, \sigma_{yy}$  and  $\sigma_{zz}$  are defined by the Hooke's law [12]:

$$\begin{aligned}\varepsilon_{xx} &= \frac{1}{E} [\sigma_{xx} - \nu(\sigma_{yy} + \sigma_{zz})] \\ \varepsilon_{yy} &= \frac{1}{E} [\sigma_{yy} - \nu(\sigma_{xx} + \sigma_{zz})] \\ \varepsilon_{zz} &= \frac{1}{E} [\sigma_{zz} - \nu(\sigma_{xx} + \sigma_{yy})]\end{aligned}\tag{5.8}$$

Here,  $\sigma_{yy} = 0; \varepsilon_{zz} = 0$

$$\begin{aligned}\varepsilon_{zz} &= \frac{1}{E} [\sigma_{zz} - \nu(\sigma_{xx} + \sigma_{yy})] = \frac{1}{E} (\sigma_{zz} - \nu\sigma_{xx}) = 0 \\ \sigma_{zz} &= \nu\sigma_{xx}\end{aligned}\tag{5.9}$$

$$\begin{aligned}\text{So, } \varepsilon_{xx} &= \frac{1}{E} [\sigma_{xx} - \nu^2\sigma_{xx}] = \frac{\sigma_{xx}}{E} (1 - \nu^2) \\ \sigma_{xx} &= \frac{E\varepsilon_{xx}}{1 - \nu^2}\end{aligned}\tag{5.10}$$

In this case,  $\varepsilon_{xx} = -\varepsilon = -b'$ . Substituting into Equation 5.10,

$$\sigma_{xx} = -\frac{Eb'}{1 - \nu^2}\tag{5.11}$$

If  $D' = \frac{Gb'}{2\pi(1 - \nu)} = 1$  (GPa), then

$$b' = \frac{2\pi(1 - \nu)}{G}\tag{5.12}$$

$G$  can be related to  $E$  by

$$G = \frac{E}{2(1 + \nu)}\tag{5.13}$$

From Equations 5.11, 5.12 and 5.13,

$\sigma_{xx} = -4\pi = -12.566$  (GPa), which is the same as shown in Fig 5.10. It again proves that validity of the above model.

### 5.4.3 Island Model

In section 5.4.2, the dislocation array is assumed to be continuous. However, it is not true for the case of the real grinding condition as pullouts appear and will break the virtual dislocation array. To simplify the problem, several assumptions are made:

- 1) All the pullouts and islands of ground region have constant sizes,  $p$  and  $s$ , respectively;
- 2) There must be one and only one pullout between two islands of ground region;
- 3) The origin of the coordinates is situated in the middle of an island of ground region.

Fig 5.11 illustrates the distribution of islands of ground regions and pullout regions that are assumed in this model.

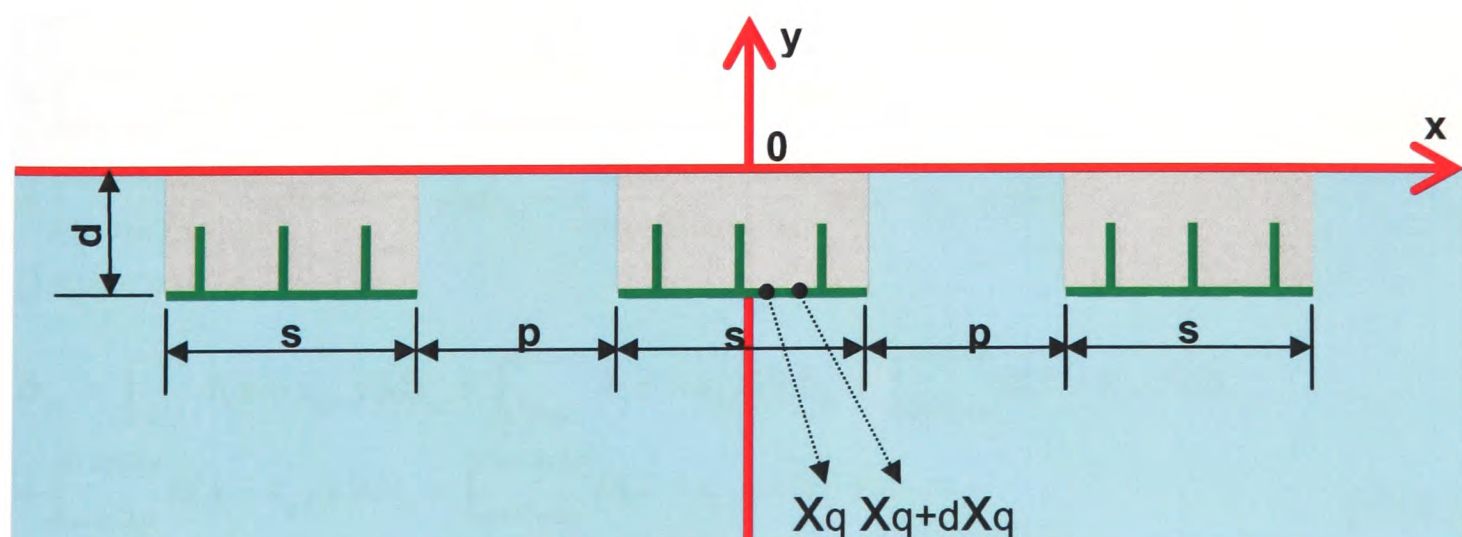


Fig 5.11 Schematic of islands of ground regions (average size:  $s$ ) and pullouts (average size:  $p$ ).

The integration process is similar to that in section 5.4.2, with the only difference being the integral interval. Again taking  $\sigma_{xx}$  as an example, at any position ( $x=x$ ,  $y=y$ ),  $d\sigma_{xx}$ , which is generated by dislocations situated at  $x=0$ ,  $y=-d$  is defined by

Equation 5.6:  $d\sigma_{xx} = f'(x, y) = f(x, y)dx_q$ .

A dislocation element  $dx_q$  situated at  $x=x_q$ ,  $y=-d$  leads to a stress component of  $d\sigma_{xx} = f'(x - x_q, y) = f(x - x_q, y)dx_q$ . The contributions from all the dislocations on the array into the  $\sigma_{xx}$  component can be integrated as:

$$\begin{aligned} \sigma_{xx} = & \int_{-s/2}^{+s/2} f(x - x_q, y)dx_q + \int_{s/2+p}^{3s/2+p} f(x - x_q, y)dx_q + \int_{-3s/2+p}^{-s/2+p} f(x - x_q, y)dx_q \\ & + \int_{3s/2+2p}^{5s/2+2p} f(x - x_q, y)dx_q + \int_{-5s/2+2p}^{-3s/2+2p} f(x - x_q, y)dx_q + \dots \\ & + \int_{(2n-3)s/2+(n-1)p}^{(2n-1)s/2+(n-1)p} f(x - x_q, y)dx_q + \int_{[(2n-1)s/2+(n-1)p]}^{[(2n-3)s/2+(n-1)p]} f(x - x_q, y)dx_q \\ & (1 \leq n < \infty) \end{aligned} \quad (5.14 \text{ a})$$

Similarly,

$$\begin{aligned} \sigma_{yy} = & \int_{-s/2}^{+s/2} g(x - x_q, y)dx_q + \int_{s/2+p}^{3s/2+p} g(x - x_q, y)dx_q + \int_{-3s/2+p}^{-s/2+p} g(x - x_q, y)dx_q \\ & + \int_{3s/2+2p}^{5s/2+2p} g(x - x_q, y)dx_q + \int_{-5s/2+2p}^{-3s/2+2p} g(x - x_q, y)dx_q + \dots \\ & + \int_{(2n-3)s/2+(n-1)p}^{(2n-1)s/2+(n-1)p} g(x - x_q, y)dx_q + \int_{[(2n-1)s/2+(n-1)p]}^{[(2n-3)s/2+(n-1)p]} g(x - x_q, y)dx_q \\ & (1 \leq n < \infty) \end{aligned} \quad (5.14 \text{ b})$$

$$\begin{aligned} \sigma_{xy} = & \int_{-s/2}^{+s/2} h(x - x_q, y)dx_q + \int_{s/2+p}^{3s/2+p} h(x - x_q, y)dx_q + \int_{-3s/2+p}^{-s/2+p} h(x - x_q, y)dx_q \\ & + \int_{3s/2+2p}^{5s/2+2p} h(x - x_q, y)dx_q + \int_{-5s/2+2p}^{-3s/2+2p} h(x - x_q, y)dx_q + \dots \\ & + \int_{(2n-3)s/2+(n-1)p}^{(2n-1)s/2+(n-1)p} h(x - x_q, y)dx_q + \int_{[(2n-1)s/2+(n-1)p]}^{[(2n-3)s/2+(n-1)p]} h(x - x_q, y)dx_q \\ & (1 \leq n < \infty) \end{aligned} \quad (5.14 \text{ c})$$



Assuming  $D'=1$  GPa,  $d=5$   $\mu\text{m}$ ,  $s=16$   $\mu\text{m}$ ,  $p=8$   $\mu\text{m}$ , the stress components predicted from the island model are shown in Fig 5.12.

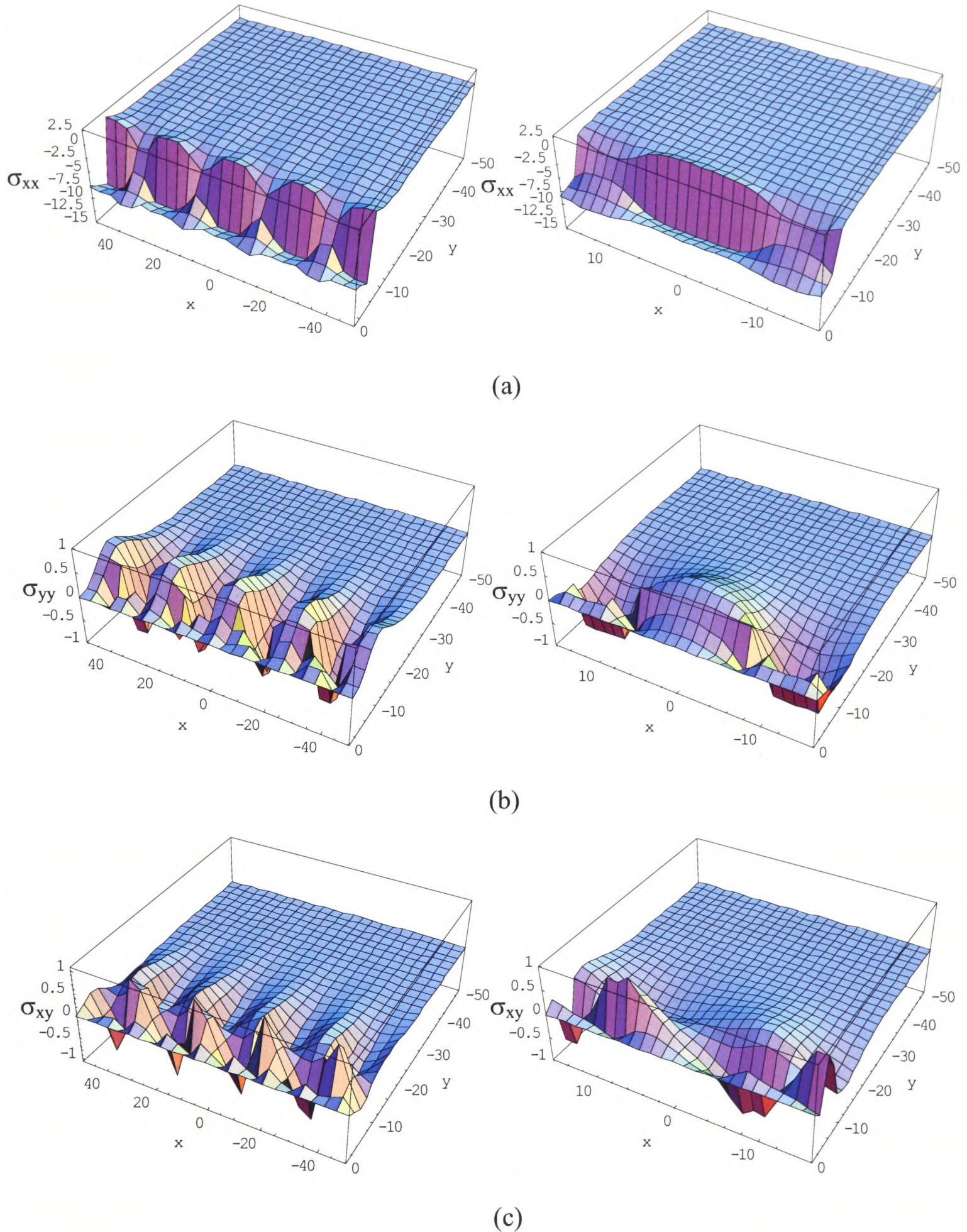


Fig 5.12 Stress components that are produced by the dislocation array shown in Fig 5.11. (a)  $\sigma_{xx}$ ; (b)  $\sigma_{yy}$ ; (c)  $\sigma_{xy}$ . The plots on the right expands the x scale of the plots on the left. The units of the stresses are GPa and x and y are in  $\mu\text{m}$ .

The magnitude of the stress components is dependent on the value of  $D'$ ,  $d$ ,  $s$  and  $p$ . As a result, what is shown in Fig 5.12 just gives an example of how  $\sigma_{xx}$ ,  $\sigma_{yy}$  and  $\sigma_{xy}$  are distributed across the surface ( $x$ ) and along the depth ( $y$ ) of the material. Seen from Fig 5.12, all three stress components  $\sigma_{xx}$ ,  $\sigma_{yy}$ , and  $\sigma_{xy}$  approach zero far below the surface. Note that  $\sigma_{xx}$  and  $\sigma_{yy}$  are strongly tensile for some distance below the ground regions. This is important to explain for the cracking phenomenon: tensile  $\sigma_{xx}$  will lead to crack growth if it is beyond the fracture strength of the material, and also weaken the material; tensile  $\sigma_{yy}$  will lead to spalling off and grain pullout. These tensile stresses are not observed before the pullouts are introduced into the model (see Fig 5.10). The existing pullouts actually lead to further pullouts and further weakening of the material.

It has to be pointed out that the predicted stress components above material ( $y > 0$ ) are not zero, which is physically meaningless. However, when the stress is convoluted with the resolution function to predict the theoretical average stress, the integral interval will be within the material only ( $y \leq 0$ ), so the non-negative stress above surface will not affect the validity of the convolution results.

Only the normal stress components contribute to the peak shift in the fluorescence microscopy [13], and after averaging for crystallographic anisotropy, this is proportional to the total normal stress, which is  $(1 + \nu)(\sigma_{xx} + \sigma_{yy})$  in plane strain [11]:

$$\begin{aligned}\sigma_{zz} &= \nu(\sigma_{xx} + \sigma_{yy}) \\ \sigma_{xx} + \sigma_{yy} + \sigma_{zz} &= (1 + \nu)(\sigma_{xx} + \sigma_{yy})\end{aligned}\tag{5.15}$$



where  $\nu=0.24$  is used for alumina materials [14].

Again taking  $D'=1$  GPa,  $d=5$   $\mu\text{m}$ ,  $s=16$   $\mu\text{m}$  and  $p=8$   $\mu\text{m}$ , the total normal stress is shown in Fig 5.13.

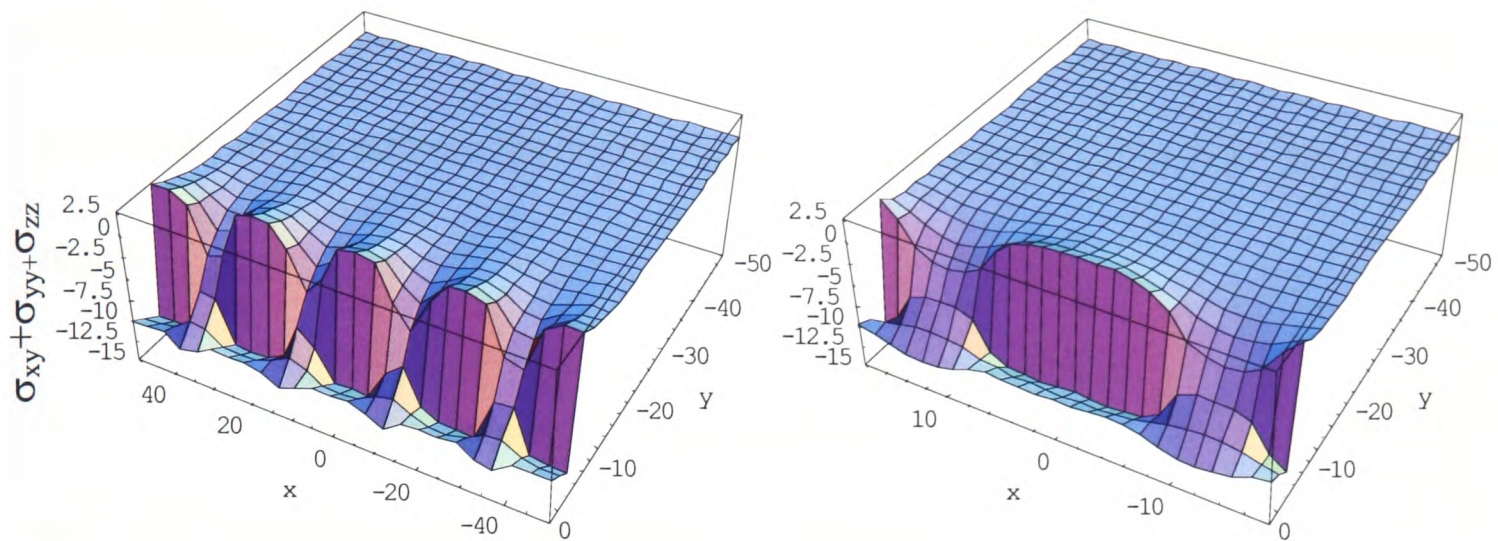


Fig 5.13 The total normal stress. The plot on the right expands the x scale of the plot on the left. The units of the stresses are GPa and x and y are in  $\mu\text{m}$ .

In the real situation, even for a seemingly depth-only scan, the signal collected is from a finite volume rather than just on a vertical line, and the convolution is actually a three dimensional issue. Taking into account the lateral contribution would make the calculation too time consuming. To simplify, two assumptions are made for the stress functions.

1) Only the stresses along the depth direction will be considered in the following convolution, and they are then convoluted with the resolution function on the depth direction only, which is known from Chapter 4.

$$\sigma(x, y) = \sigma(x_0, y) = (1 + \nu)(\sigma_{xx} + \sigma_{yy}) \Big|_{x=x_0, y=y} \quad (5.16)$$

$x_0$  is the position where the depth scan is made.



This is a reasonable simplification as the lateral resolution is much better than the axial resolution (see Chapter 4).

2) The depth scan through the ground regions and pullout regions are made in their centers. Fig 5.12 shows that the variation of the stress components  $\sigma_{ij}$  with  $x$  is slow at the center. This also confirms that ignoring the lateral resolution function will have little effect on the stresses. Referring to the simple geometry in Fig 5.11, for a ground region,

$$\begin{aligned}\sigma(x, y) &= \sigma(0, y) = \sigma((s + p), y) = \sigma(-(s + p), y) \\ &= \sigma(2(s + p), y) = \sigma(-2(s + p), y) = \dots\end{aligned}\tag{5.17}$$

and  $\sigma(0, y)$  will be used for all future calculations.

For pullout regions,

$$\begin{aligned}\sigma(x, y) &= \sigma\left(\frac{s + p}{2}, y\right) = \sigma\left(-\frac{s + p}{2}, y\right) \\ &= \sigma\left(\frac{3(s + p)}{2}, y\right) = \sigma\left(-\frac{3(s + p)}{2}, y\right) = \dots\end{aligned}\tag{5.18}$$

and  $\sigma\left(\frac{s + p}{2}, y\right)$  will be used for all future calculations.

Again assuming  $D'=1$  GPa,  $d=5$   $\mu\text{m}$ ,  $s=16$   $\mu\text{m}$  and  $p=8$   $\mu\text{m}$ , the total normal stress distributions through the ground region and pullout region are given in Fig 5.14 (a) and (b). A sharp change of stress at the depth of the dislocation array ( $y=-d$ ) is observed, which is also seen in Fig 5.10 (a).

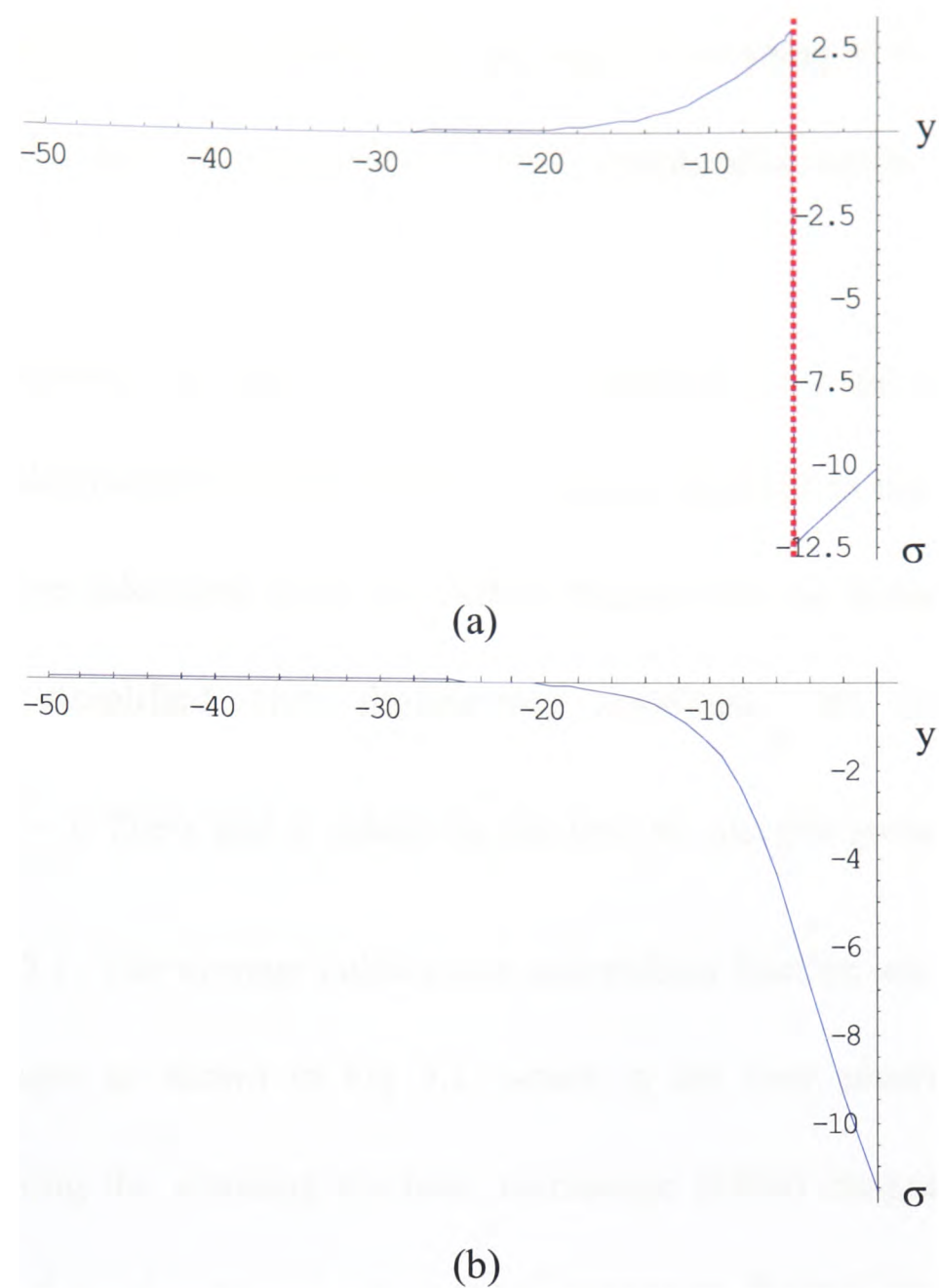


Fig 5.14 Total normal stress distributions along the depth direction. (a) through the center of the ground region, (b) through the center of the pullout region. Unit of stresses are in GPa and  $y$  is in  $\mu\text{m}$ .

5.5 Curve Fitting and Convolution

Since the resolution function is already known, and the stress function has also been developed based on the dislocation model, this modeling is now ready for the convolution step. The details of the resolution function will be further discussed later in this section as some meaningful values of  $D'$ ,  $d$ ,  $s$  and  $p$  will help to quantify the problem. Some details of the experimental results need to be considered before being

fitted by the prediction. At the same time, the physical meaning of the convolution is further discussed in the following analysis of the experimental results.

In the stress function, there are four parameters, namely  $D'$ ,  $d$ ,  $s$  and  $p$ .  $s$  and  $p$  can be experimentally determined to some extent as  $p$  can be regarded as the average pullout size and  $s$  can be calculated from the pullout fraction (as the dislocation model is based on a simplified two dimensional condition, the pullout fraction

$$\text{pullout}\% = \frac{p}{p + s}).$$

The  $s$  and  $p$  values for the four 45  $\mu\text{m}$  grit ground aluminas are

given in Table 5.1. The average pullout size and pullout fraction are measured from the optical images as shown in Fig 5.1, which is not very accurate and can be improved by using the scanning electron microscope (SEM) images. However, for each depth scan through a ground region or pullout region, the real sizes of the ground island and pullout are actually varying case by case depending on the location, and the  $s$  and  $p$  listed in Table 5.1 are just an averaged value. A rough estimate of  $s$  and  $p$  via optical images will suffice for this requirement.

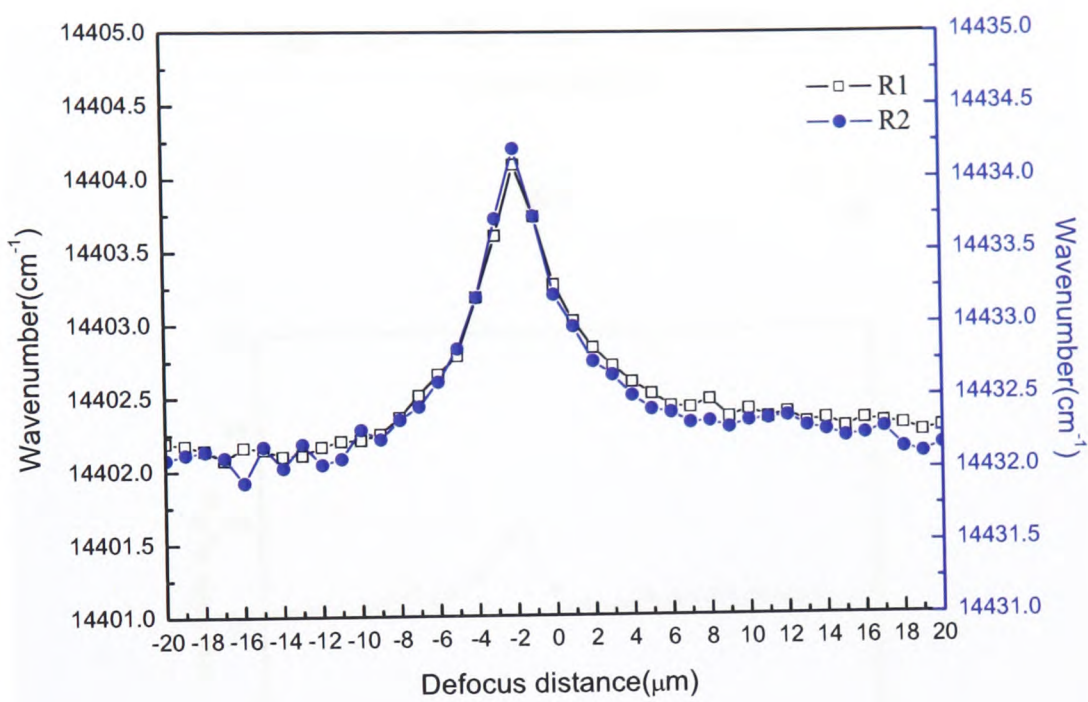
$D'$  and  $d$  are to be determined, which can be obtained from the best fit to the experimental results. Considering the complexity of the fitting, values of  $D'$  and  $d$  were in practice obtained by a trial and error method to make the prediction and experimental result a good fit.

### 5.5.1 Curve Fitting

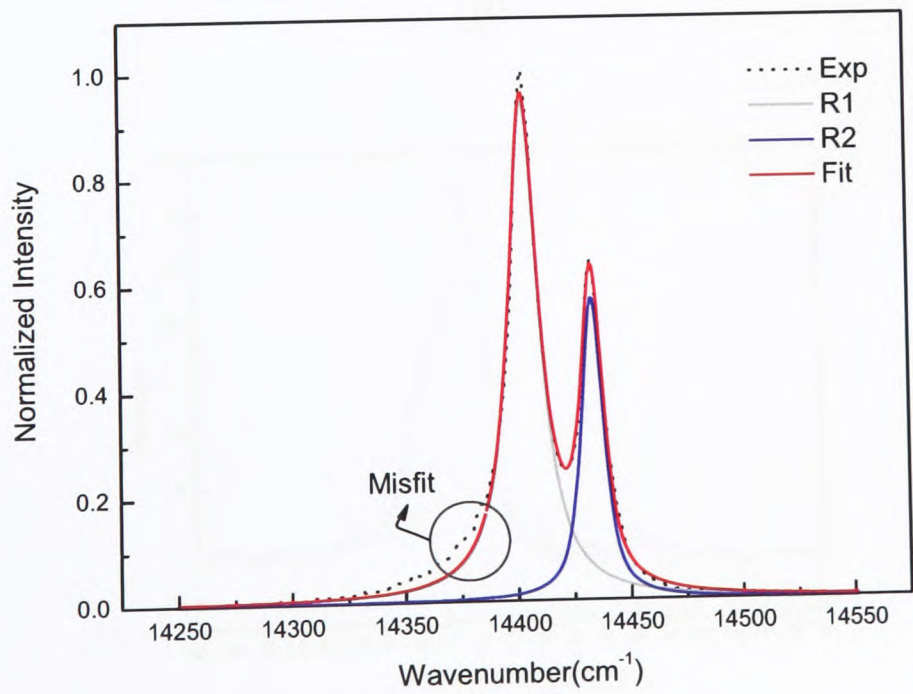
It has already been mentioned in section 5.3 that the experimentally measured wavenumber shift is from the R1 peak position using a mixed Gaussian and Lorentzian function to fit the spectra with two peaks (R1, R2, see Fig 5.15 (a)). It works well when the spectrum is collected by focusing far away from the surface, i.e., a very good fitting can be obtained. However, when focusing near the ground surface (and not in the pullout region) the spectra cannot be fitted satisfactorily. A typical example is shown in Fig 5.15 (b), where an obvious misfit on the R1 side between the fitted curve and the original spectrum can be seen. Mathematically, the fitting of the same spectrum can be improved by adding another set of R1 and R2 peaks, as seen in Fig 5.16 (a). Physically, the extra doublet means another quite different stress state within the sampling volume. The peak shift characterizes the magnitude of the stress, while the width of the peak can be related to the complexity (variation) of the stress states around the mean value. The peak width would broaden if there were more than one stress state existing within the sampling volume as the measured spectra are actually representing the superimposition of different states.

Separation of different stress states, or in this case different sets of the R line doublet, is possible only when they are quite different to each other. Seen from Fig 5.16(a), two sets of R lines can be separated mathematically from the original spectrum by appropriate curve fitting and the one having the bigger negative shift is also broader than the other one. The broad peak indicates larger stress around the mean stress for

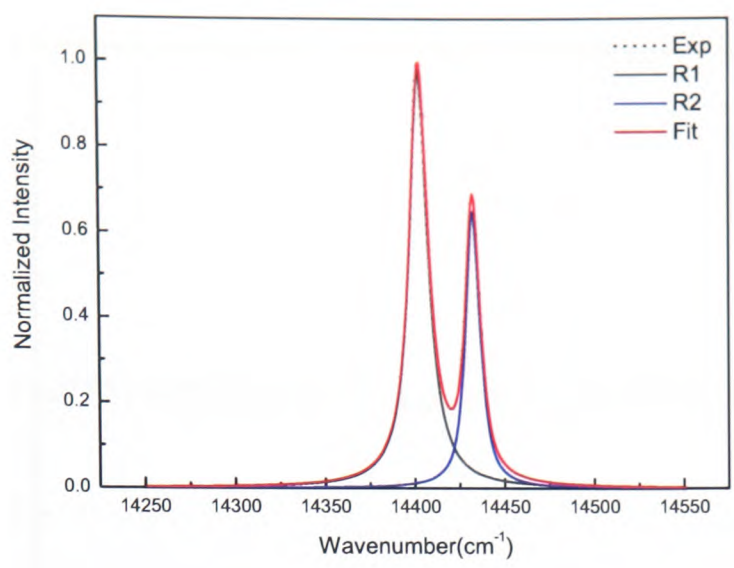
this pair of peaks. As will be discussed below, it is argued here that the broad, second pair of peaks is physically meaningful and corresponds to the plastically deformed surface region, where dislocations and twins will cause a range of stresses that leads to broadening, even if the macro stresses are uniform.



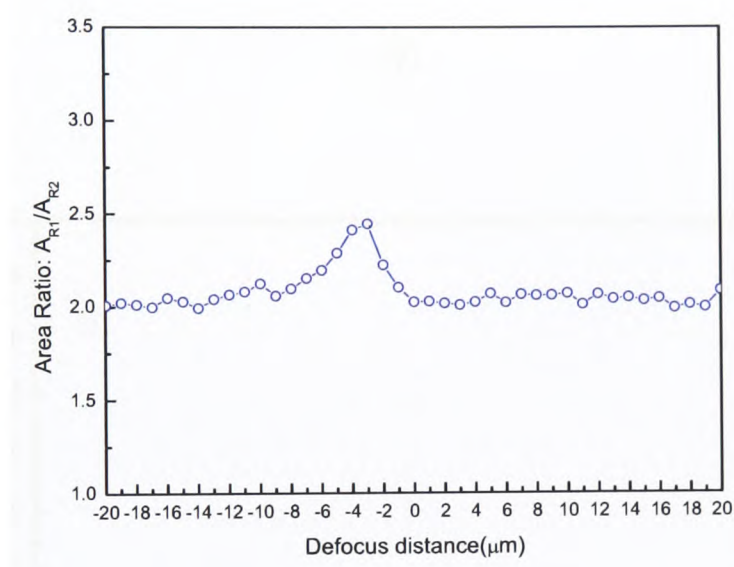
(a)



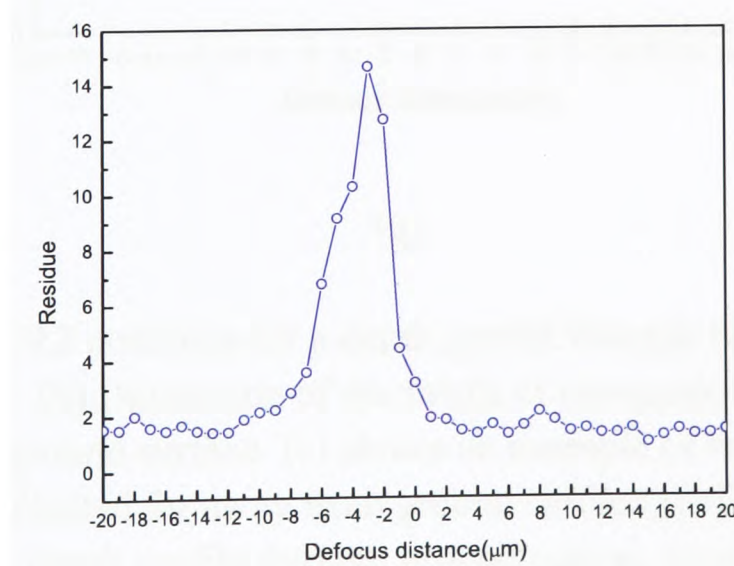
(b)



(c)

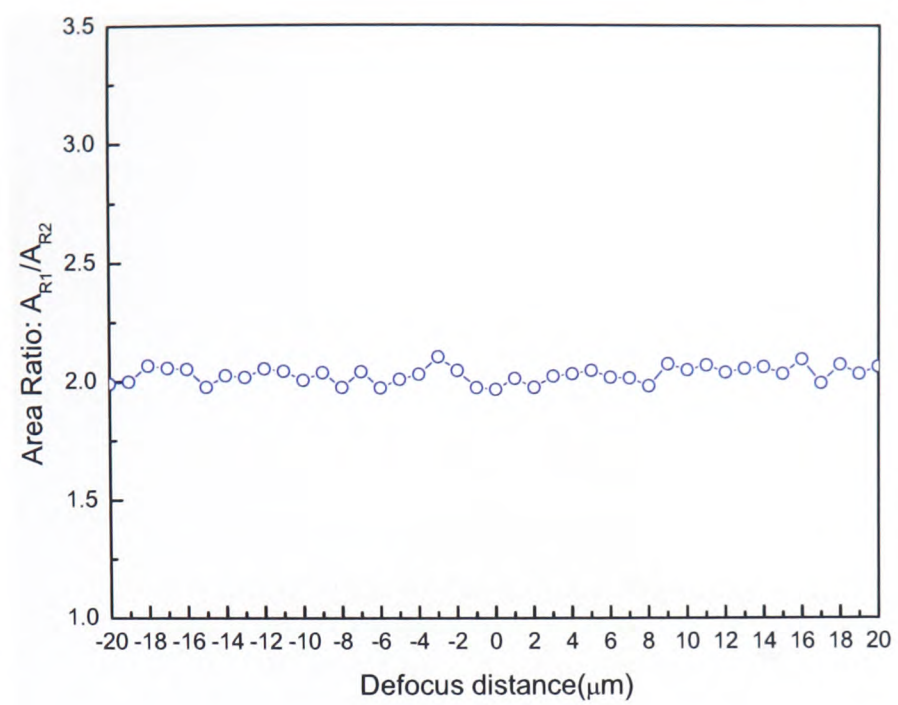


(d)

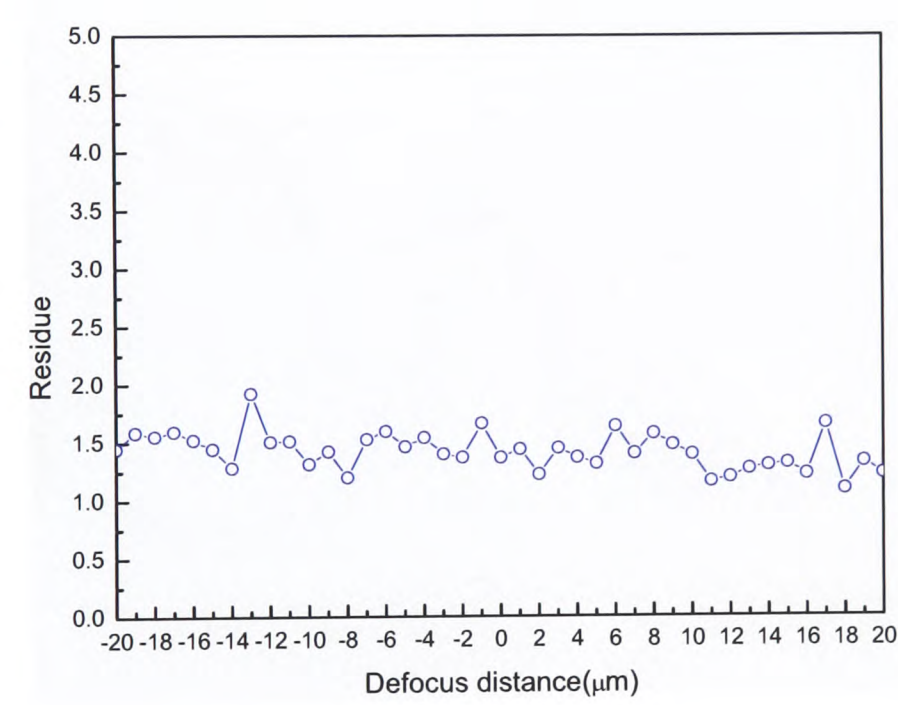


(e)



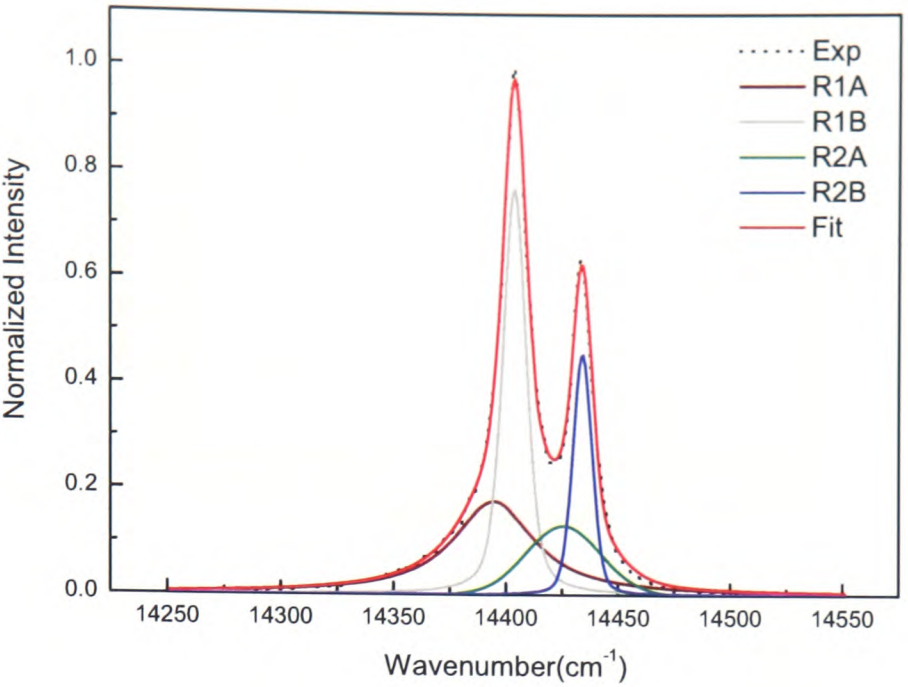


(f)

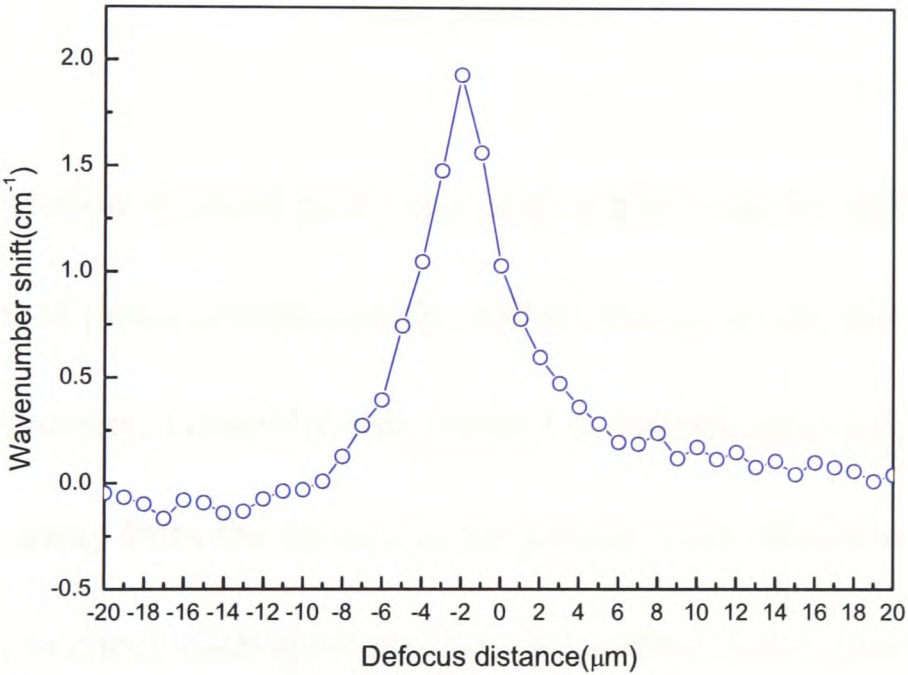


(g)

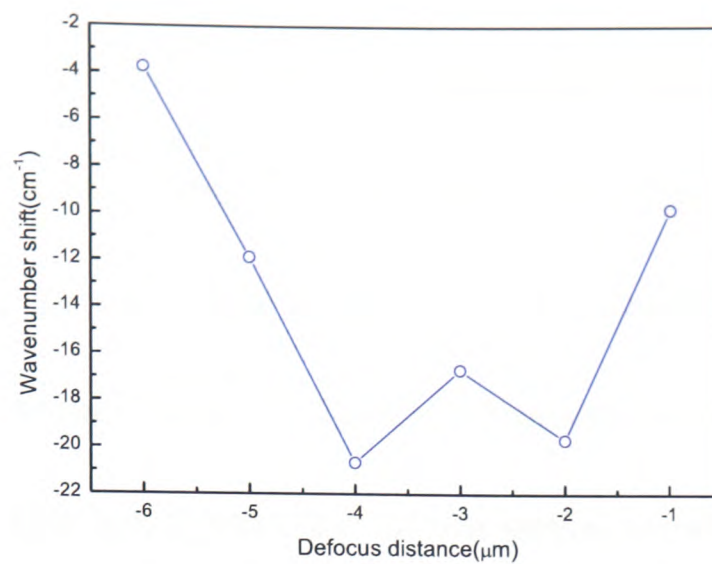
Fig 5.15 (a) R1 and R2 positions for a depth profile through the ground region, from two-peak fittings; (b) Illustration of the misfit of two-peak fitting to a spectrum collected from near ground surface. (c) shows an example of two-peak fitting working well for spectra collected far away from ground surface;(d) and (e): area ratio and fitting residue for a depth profile through ground region, from two-peak fittings; (f) and (g): area ratio and fitting residue for a depth profile through pullout region, from two-peak fittings.



(a)



(b)



(c)

Fig 5.16 (a) Improvement of curve fitting by using four-peak fitting. The spectrum is the same as in Fig 5.15 (b);

(b) and (c) are R1 peak positions after separating broad peaks (indicated by subscript A) and narrow peaks (indicated by subscript B): (b) narrow peak positions, (c) broad peak positions

Meaningful separation of broad peaks and narrow peaks can be made only for spectra in which one set of peaks is significantly shifted relative to the other and the two sets are of similar intensity. Generally this applied to spectra that were collected within several microns away from the surface in the present work. However, it is tedious and time consuming to check each spectrum near the surface to see whether a good fitting is possible. By using one set of peaks only (R1 and R2, called two-peak fitting from here onwards), some useful information can be drawn to give hints on when to use two sets of R-lines (called four-peak fitting afterwards) and when only one set is good enough. It is found that for those spectra when two-peak fitting fails, the area ratios of R1 to R2 peaks (see Fig 5.15 (d)) are higher than average, while the ratios for those spectra collected far away from the surface are nearly constant or fluctuate very

slightly around an average value. The fitting residue plot (Fig 5.15 (e)) also reflects of the goodness of fitting. Both R1/R2 area ratio and fitting residue can be easily generated by the two-peak fitting. They are used as the criteria to determine when to use two-peak fitting or four-peak fitting. The procedure of four-peak fitting has already been explained in Chapter 3. Typical R1 positions from depth scans through the ground regions, both broad peaks and narrow peaks, are shown in Fig 5.16 (b) and (c). The narrow peaks are reflecting tensile stresses, while the broad peaks are reflecting strongly compressive stresses.

### 5.5.2 Convolution

The stresses are converted to wavenumber shifts to fit the experimentally measured results:

$$\begin{aligned} \Delta \nu &= \frac{\Pi_{11} + \Pi_{22} + \Pi_{33}}{3} (\sigma_{xx} + \sigma_{yy} + \sigma_{zz}) = \\ &\frac{\Pi_{11} + \Pi_{22} + \Pi_{33}}{3} (1 + \nu) (\sigma_{xx} + \sigma_{yy}) = 3.14 (\sigma_{xx} + \sigma_{yy}) \end{aligned} \quad (5.19)$$

where  $\Pi_{11}=2.56$ ,  $\Pi_{22}=3.50$  and  $\Pi_{33}=1.53 \text{ cm}^{-1} \text{ GPa}^{-1}$  were used [15].

From the analysis in section 5.5.1, it seems reasonable to use the depth of the dislocation array corresponding to the plastically deformed surface layer to divide these two regions of different stress state and correlate the broad peaks and narrow peaks with these two stress states. Mathematically, it is just a matter of the integral interval. For the broad peak results, the convolution is of the form:

$$\overline{\Delta\nu(y_0)} = \frac{\int_{-d}^0 \Delta\nu(0, y) g(y, y_0) dy}{\int_{-d}^0 g(y, y_0) dy} \quad (5.20)$$

where  $y_0$  is the distance away from the surface. As the laser is set to focus on the sample surface before each depth scan starts, so  $y_0$  is also the defocus distance (refer to Chapter 3);  $\Delta\nu(y_0)$  is the predicted wavenumber shift;  $g(y, y_0)$  is the (axial) resolution function; and  $\overline{\Delta\nu(y_0)}$  is the measured (average) wavenumber shift.

For the narrow peak results, the integral interval for the convolution is below the dislocation array:

$$\overline{\Delta\nu(y_0)} = \frac{\int_{-t}^d \Delta\nu(0, y) g(y, y_0) dy}{\int_{-t}^d g(y, y_0) dy} \quad (5.21)$$

where  $t$  is the thickness of the material.

From the viewpoint of curve fitting, the R1 positions for the narrow peak are well defined and can be reliably repeated; the ill-defined R1 positions for the broad peak are less reliable and vary between different fitting runs. Further away from the surface, the signal/noise ratios of the spectra decrease and it also makes the determination of peak positions less accurate. When fitting the broad peaks, the convoluted results are adjusted (varying  $D'$  and  $d$ ) to fit the average R1 positions from spectra collected very close to surface, normally where R1/R2 area ratio and residue are bigger. Fortunately, the shifts of the broad peaks from the zero stress position are very large, so the percentage error in the results remains small.

Note that the four-peak fitting is only necessary for depth scans through the ground region. For most of the spectra that were collected from depth scans through the pullout region, two-peak fitting can work very well (see Fig 5.15 (f) and (g)). It is understandable because pullout removes the plastically deformed layer from the surface. This also verifies that it is reasonable to assume the pullout region is dislocation free in the model. For the pullout results, therefore, the convolution is of the form:

$$\overline{\Delta \nu(y_0)} = \frac{\int_{-l}^0 \Delta \nu\left(\frac{s+p}{2}, y\right) g(y, y_0) dy}{\int_{-l}^0 g(y, y_0) dy} \quad (5.22)$$

Since the sign of  $y$  (distance away from distance) was defined differently in the experiments (below surface,  $y > 0$ ) and in the dislocation model (below surface,  $y < 0$ ), the experimental results were flipped during the curve fitting. The zero positions of the experimental results are also offset to match with the predicted results. The offset is due to the difficulty in focusing the laser exactly on the surface to set zero position, which was also considered when modeling the resolution function in Chapter 4.

### 5.5.3 Resolution Function

In the axial resolution function that is developed in Chapter 4, two parameters are of most importance: the width  $w$ , and the absorption coefficient  $\alpha$ . The simplified form of the resolution function can be written as:

$$g(y, y_0) = e^{-2\alpha} \frac{w}{w^2 + (y - ny_0)^2} \quad (5.23)$$



where  $n$  is the refractive index.

$w$  depends on the instrument, mainly relying on the lens, confocal optics and the laser alignment, and is also affected by vibration and temperature. Each time before the depth scan was carried out, a depth scan through a sapphire sample was used to determine  $w$ , by the method described in Chapter 4. The absorption coefficient,  $\alpha$ , was determined from a depth scan of a 1  $\mu\text{m}$  grit polished alumina using the double-Lorentzian function also developed in Chapter 4. The introduction of the second Lorentzian function is mainly to compensate for the scattering effect due to the grain boundaries. For simplicity, the simplified form of resolution function (Equation 5.23) was used in the convolution. As will be shown later, ignoring the second Lorentzian function will not generate a significant effect on the convolution results for interesting regions.

## 5.6 Fitting Results and Discussion

### 5.6.1 Grinding

Fig 5.17 shows the best fit of the convolution to the experimental results for 45 diamond grit ground 8.0  $\mu\text{m}$  sized alumina, more specifically depth scans through P2 and P5 in Fig 5.2 (d). With the known  $s$  and  $p$  values measured from the optical micrographs, the  $D'$  and  $d$  values given in Table 5.4 were determined based on the fitting for the narrow and broad peaks from the ground regions. The same four parameters were inspected on the peak positions from the pullout regions.

Considering the simplicity of the dislocation model and the many assumptions that were made, the fitting is reasonably good, and the model successfully predicts how the peak positions vary on the depth direction (the shape), for both ground regions and pullout regions. As seen in Fig 5.17 (a), the fitting below the surface ( $y \leq 0$ ) is good for the ground region. The fitting above the surface ( $y > 0$ ) is not very satisfactory but it is less important as it is the stresses within material that are of interest. The fitting problem above the surface could be due to the tails in the resolution function: when the focus is above the surface, the main contribution to the collected signal comes from the tail of the probe response curve, which was actually not fitted very well using the PRF proposed in this work (demonstrated in Fig 5.18). Lipkin and Clarke also addressed the similar problem and they referred it as the “probe channeling” effect [16].

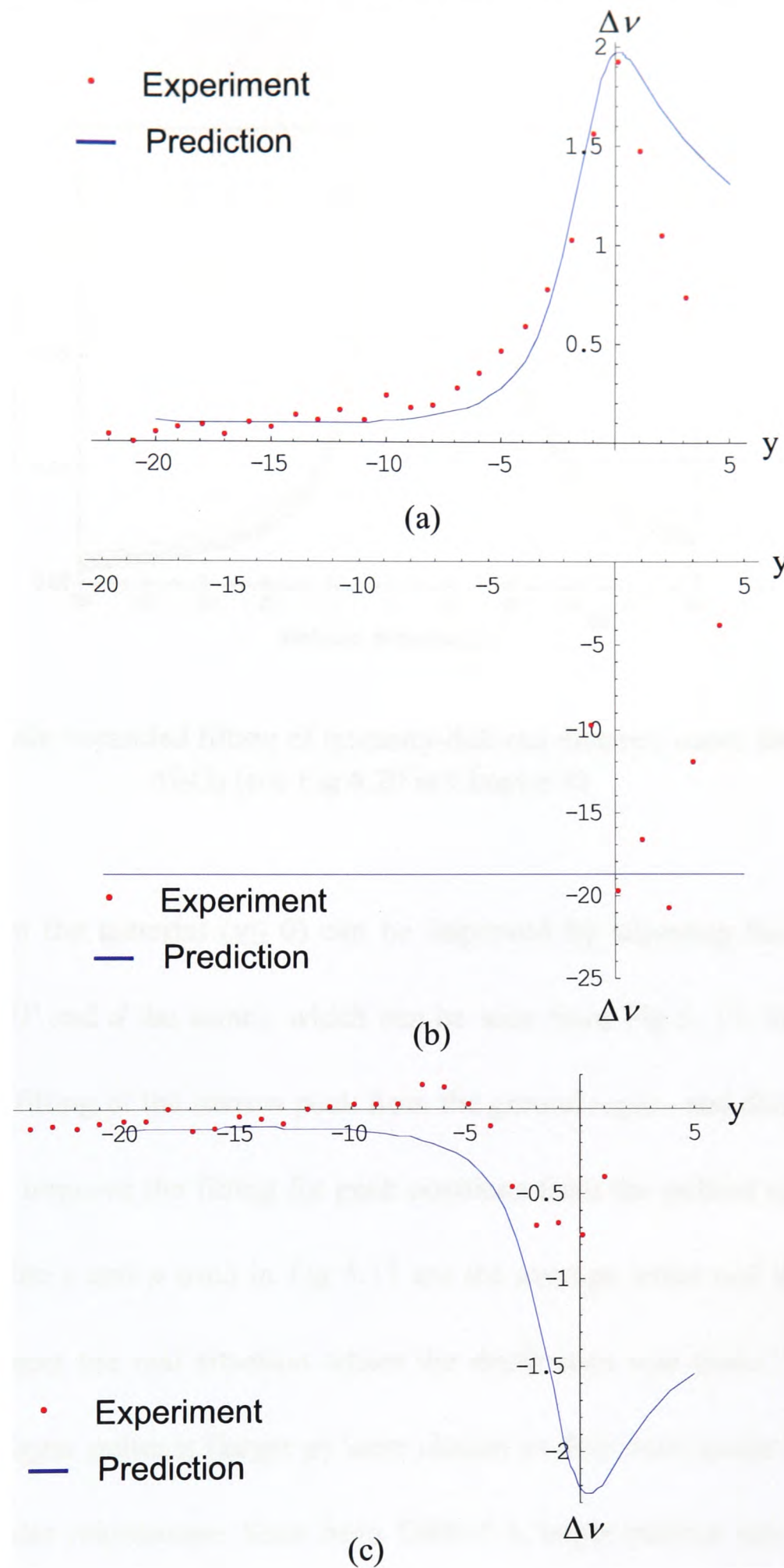


Fig 5.17 Fitting results for 8.0  $\mu\text{m}$  sized ground alumina  
 (a) narrow peaks from ground region; (b) broad peaks from ground region; (c)  
 pullout region. Unit of  $\Delta\nu$  is cm $^{-1}$  and  $y$  is in  $\mu\text{m}$ .

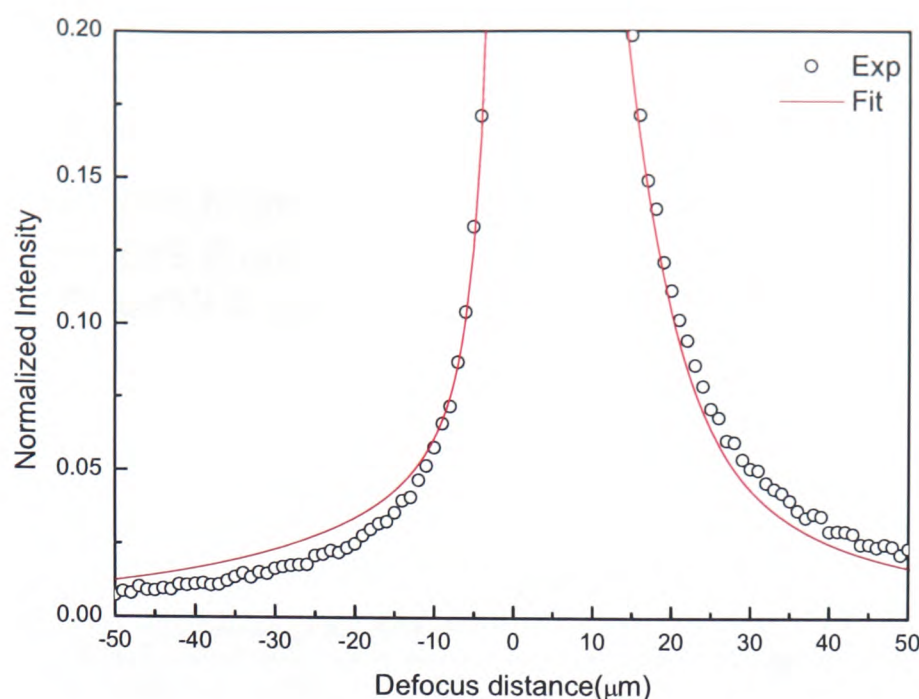


Fig 5.18 Y-scale expanded fitting of intensity-defocus distance curve for 5.6  $\mu\text{m}$   $\text{Al}_2\text{O}_3$  (see Fig 4.20 in Chapter 4)

The fitting within the material ( $y \leq 0$ ) can be improved by adjusting the  $p$  and  $s/p$  values (keeping  $D'$  and  $d$  the same), which can be seen from Fig 5. 19. Increasing  $p$  will improve the fitting of the narrow peak from the ground region and decreasing  $s/p$  will significantly improve the fitting for peak positions from the pullout region. This is reasonable as the  $s$  and  $p$  used in Fig 5.17 are the average value and they do not necessarily represent the real situation where the depth scan was made. In the real measurements, bigger pullouts (larger  $p$ ) were chosen as they were easier to identify with certainty under microscope. Seen from Table 5.3, larger pullout size ( $p$ ) would not significantly affect the fitted  $D'$  and  $d$  values. The predicted variation with  $p$  and  $s$  also explains the variation of depth profiles made at different locations (P1, P2 and P3 from ground regions; P4, P5 and P6 from pullout regions in Fig 5.2). In the following fittings, the typical depth profiles (with middle maximum peak shift) were fitted with

the theoretical prediction, to make the experimental results more representative.

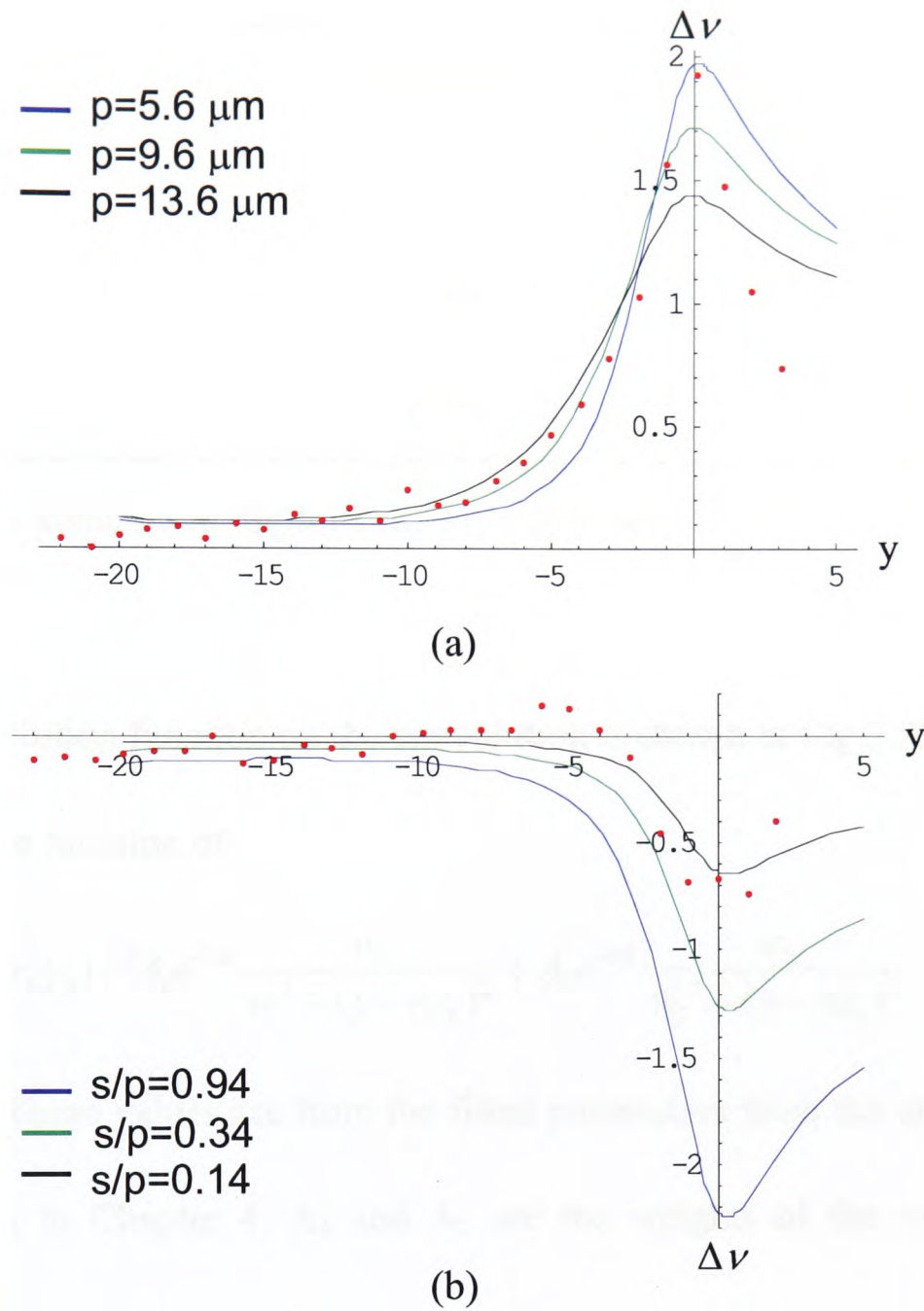


Fig 5.19 Adjustment of  $p$  and  $s/p$  to improve the fitting of 8.0 μm sized ground alumina. (a) narrow peaks from ground region,  $s/p=0.94$ , varying  $p$ ; (b) pullout region,  $p=5.6$ , varying  $s/p$ . Unit of  $\Delta\nu$  is cm<sup>-1</sup> and  $y$  is in μm.

Table 5.3 Sensitivity of the fitted  $D'$  and  $d$  values to pullout size ( $p$ )

| $^1p$ ( $\mu\text{m}$ ) | $s/p=0.94$ |                       |
|-------------------------|------------|-----------------------|
|                         | $D'$ (GPa) | $d$ ( $\mu\text{m}$ ) |
| 5.6                     | 0.59       | 0.54                  |
| 9.6                     | 0.56       | 0.67                  |
| 13.6                    | 0.56       | 0.81                  |

<sup>1</sup>The pullout sizes assumed in Fig 5.19 (a) were used here

The effect of resolution function on the convolution is shown in Fig 5.20. Assuming a double-Lorentzian function of:

$$g(y, y_0) = A_1 e^{-2\alpha} \frac{w_1}{w_1^2 + (y - ny_0)^2} + A_2 e^{-2\alpha} \frac{w_2}{w_2^2 + (y - ny_0)^2} \quad (5.24)$$

$w_1$ : 2.9;  $w_2$ : 72. These values are from the fitted parameters from the depth profile of polished alumina in Chapter 4.  $A_1$  and  $A_2$  are the weights of the two Lorentzian functions.  $A_1$  and  $A_2$  are 0.6 and 0.2 respectively for alumina.

Changing the  $A_2/A_1$  ratio has little effect on the convolution for broad peaks from ground region, although when  $A_2/A_1$  is big the predicted narrow peak from ground regions and peaks from pullout regions will be noticeably different, especially when focusing above the surface. However, within a certain depth below the surface ( $-5 < y \leq 0$ ), where the curve fitting for the experimental results is more reliable (higher signal/noise ratio, more reliant on the central part of the resolution function than the tails), the difference made by this aspect of the resolution function is small. It



confirms that using a simpler resolution function as in Equation. 5.23 ( $A_1=1$ ,  $A_2=0$ ) is sufficient for these purposes.

The same fitting procedure was repeated for ground 2.4  $\mu\text{m}$ , 4.2  $\mu\text{m}$  and 5.6  $\mu\text{m}$  sized alumina. The fitting results are given in Fig 5.21, 5.22 and 5.23, and a summary of the fitted parameters,  $D'$  and  $d$ , together with  $s$  and  $p$ , are listed in Table 5.4.

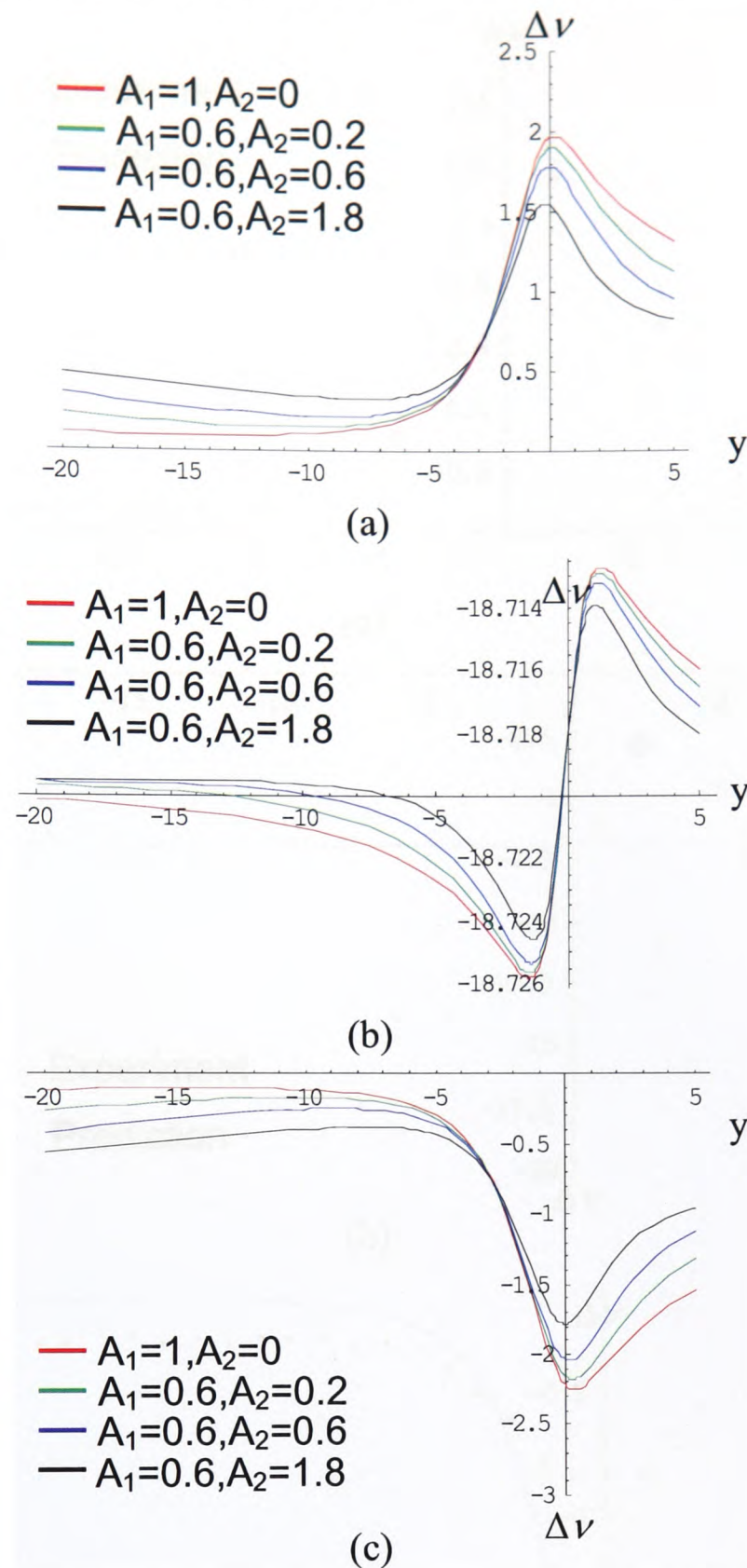


Fig 5.20 Effect of resolution functions on the convolution results for (a) narrow peaks from ground region; (b) broad peaks from ground region; (c) pullout region.  $A_1, A_2$  are the weights of two Lorentzian functions in Equation 5.24. Unit of  $\Delta\nu$  is  $\text{cm}^{-1}$  and  $y$  is in  $\mu\text{m}$ .

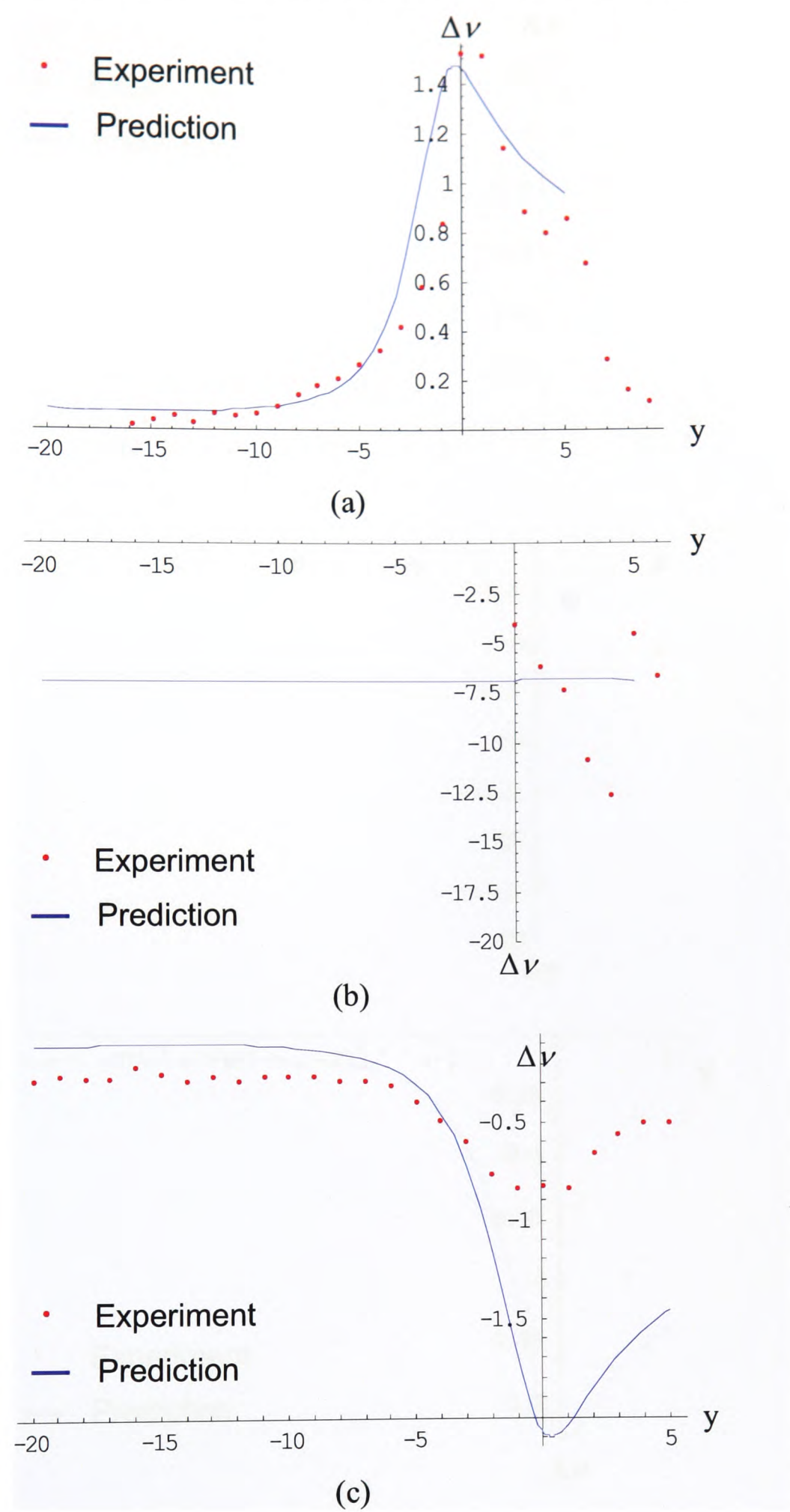


Fig 5.21 Fitting results for 2.4 μm sized ground alumina  
(a) narrow peaks from ground region; (b) broad peaks from ground region; (c)  
pullout region. Unit of  $\Delta\nu$  is cm<sup>-1</sup> and  $y$  is in μm.

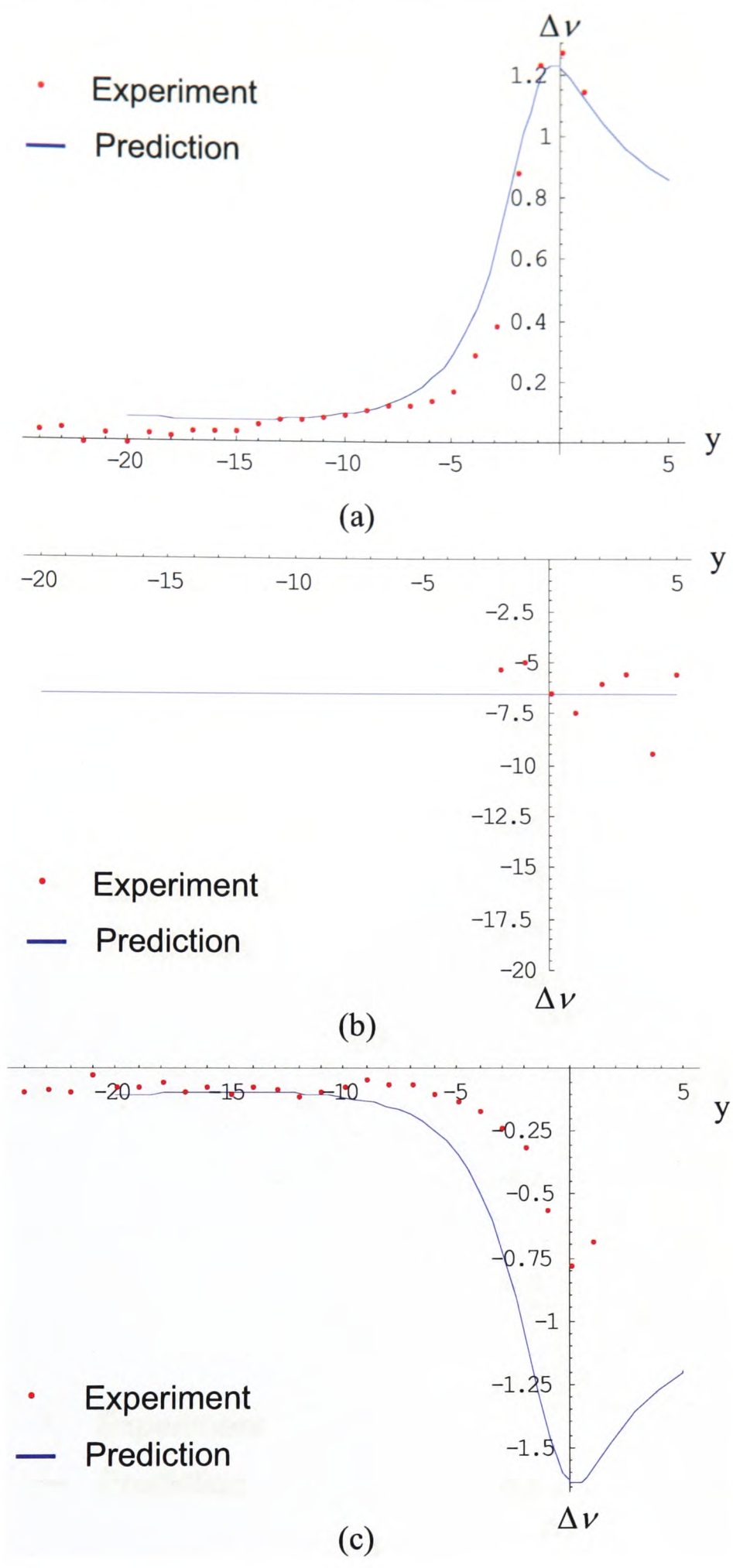


Fig 5.22 Fitting results for 4.2  $\mu\text{m}$  sized ground alumina  
(a) narrow peaks from ground region; (b) broad peaks from ground region; (c) pullout region. Unit of  $\Delta\nu$  is  $\text{cm}^{-1}$  and  $y$  is in  $\mu\text{m}$ .

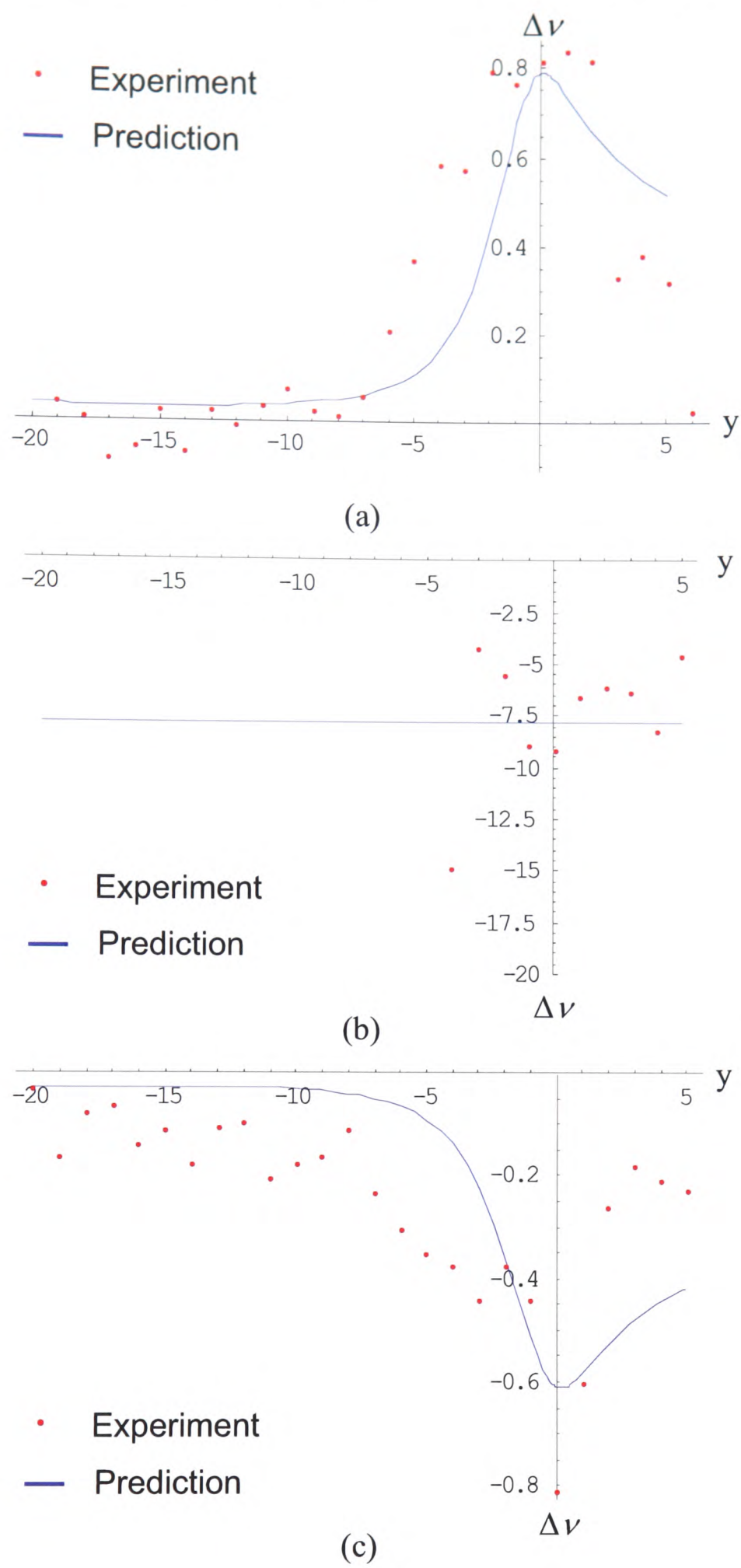


Fig 5.23 Fitting results for 5.6  $\mu\text{m}$  sized ground alumina  
(a) narrow peaks from ground region; (b) broad peaks from ground region; (c)  
pullout region. Unit of  $\Delta\nu$  is  $\text{cm}^{-1}$  and  $y$  is in  $\mu\text{m}$ .



Table 5.4 List of  $D'$ ,  $d$ ,  $s$  and  $p$  for four ground polycrystalline aluminas.  $D'$  and  $d$  are from the best fit of convolution to the experimental results (from ground region);  $s$  and  $p$  are measured from optical images.

| $G(\mu\text{m})$ | $D'$ (GPa) | $d(\mu\text{m})$ | $D' \cdot d$ (GPa $\cdot\mu\text{m}$ ) | $s(\mu\text{m})$ | $p(\mu\text{m})$ | $s/p$ |
|------------------|------------|------------------|----------------------------------------|------------------|------------------|-------|
| 2.4              | 0.28       | 1.34             | 0.38                                   | 5.3              | 6.3              | 0.84  |
| 4.2              | 0.23       | 1.32             | 0.30                                   | 8.1              | 8.4              | 0.96  |
| 5.6              | 0.24       | 0.42             | 0.10                                   | 5.1              | 7.1              | 0.72  |
| 8.0              | 0.59       | 0.54             | 0.32                                   | 5.2              | 5.6              | 0.94  |

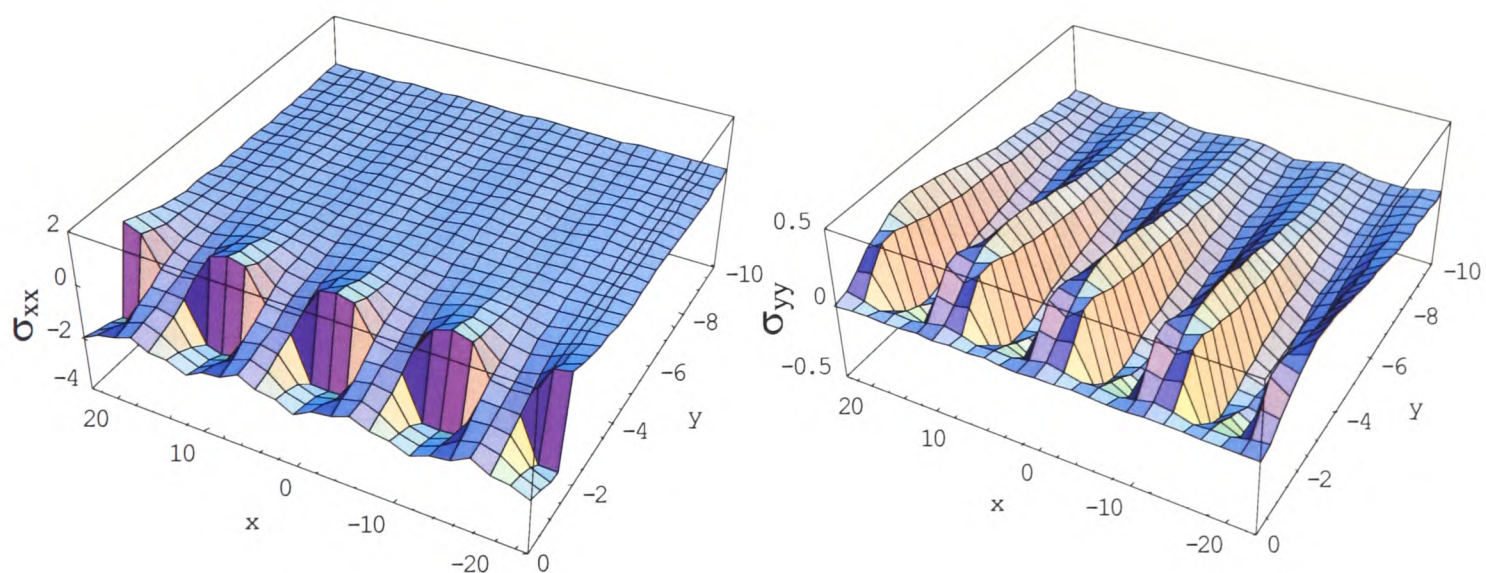
Seen from Table 5.4,  $D'$  and  $d$  are very similar between the 2.4  $\mu\text{m}$  and the 4.2  $\mu\text{m}$  sized alumina. The larger  $D'$  and hence smaller  $d$  values obtained from the 8.0  $\mu\text{m}$  alumina was due to the spectrum chosen for fitting (P1 in Fig 5.2 (d)), which was not representative of the average stress level. The 5.6  $\mu\text{m}$  sized alumina had a smaller  $d$  value compared to other aluminas, which was probably due to its fabrication route as it was hot pressed and the other specimens were pressureless sintered. In addition, considering the complexity and hence the accuracy of the fitting, the difference among the  $d$  values from the four aluminas can be regarded as in the reasonable error range. The magnitude of the depth is in agreement with the results reported by Wu et al. [8]. They estimated the depth of dislocations in the ground alumina by TEM to be  $< 1 \mu\text{m}$ .

By replacing the  $D'$ ,  $d$ ,  $s$  and  $p$  with the values listed in Table 5.4, Fig 5.12 can be replotted (Fig 5.24) to generate the real stress distributions in the four polycrystalline aluminas, without being averaged by the resolution function. In all the four specimens,



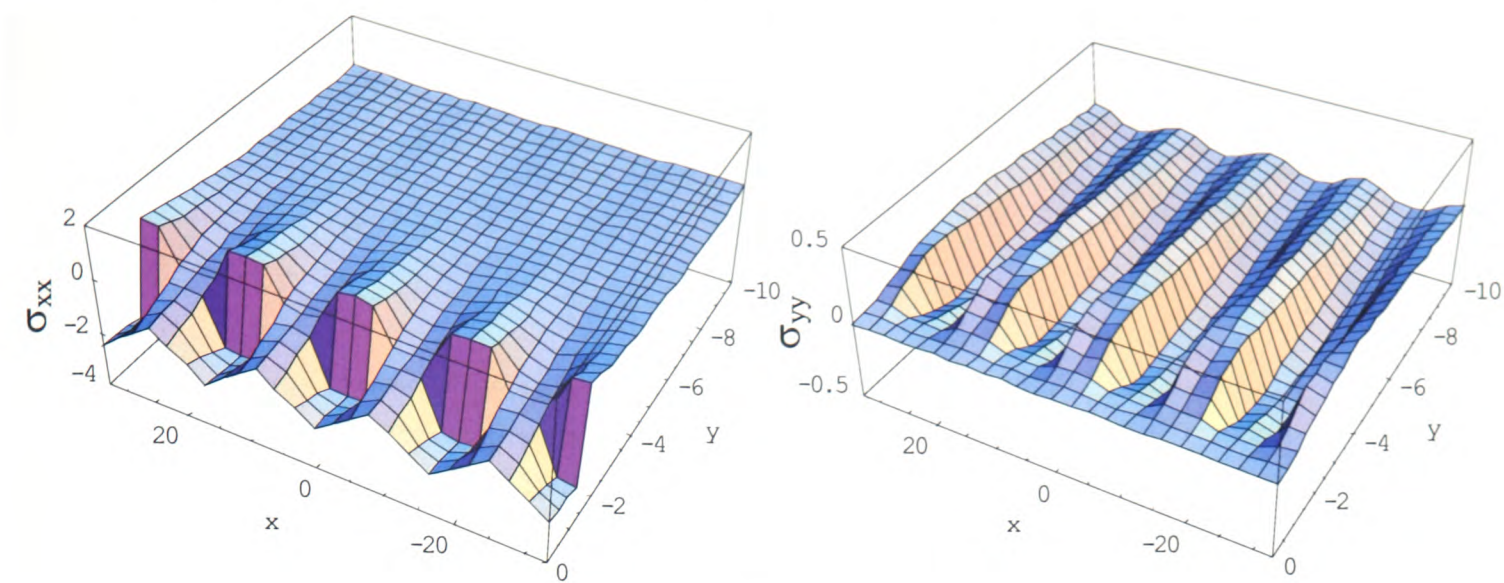
$\sigma_{xx}$  and  $\sigma_{yy}$  are tensile for some distance below the ground regions. As discussed before, these tensile stresses can be responsible for the crack growth and grain pullouts by subsurface lateral cracking respectively. Crack growth is driven by the tensile stress and larger  $\sigma_{xx}$  tends to drag cracks deeper; however, the compressive stress existing in the surface layer will counter-act this.

The total normal stresses along the depth direction, both in the ground region and pullout region, are also given in Fig 5.24. The stresses will shift from compressive to tensile beyond the depth of dislocation array in the ground region, and the maximum of tensile and compressive stresses calculated for these four different grain sized alumina are listed in Table 5.5. The maximum compressive stresses in the pullout regions are also listed.

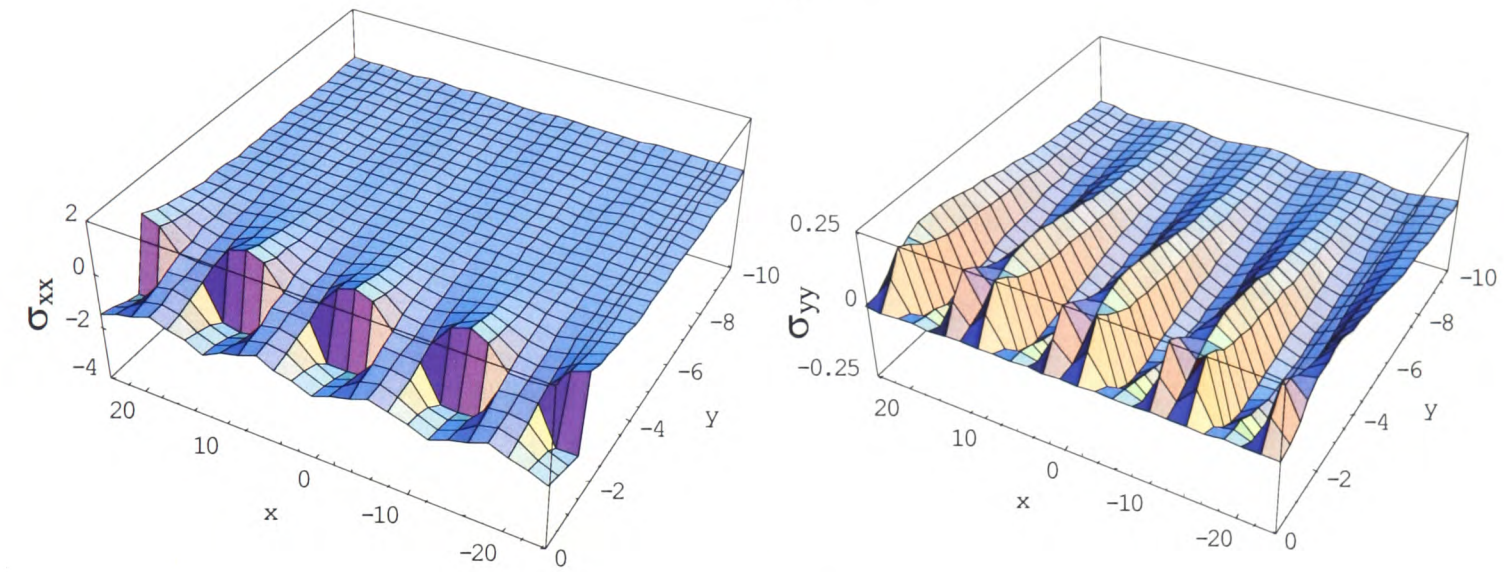


(a)

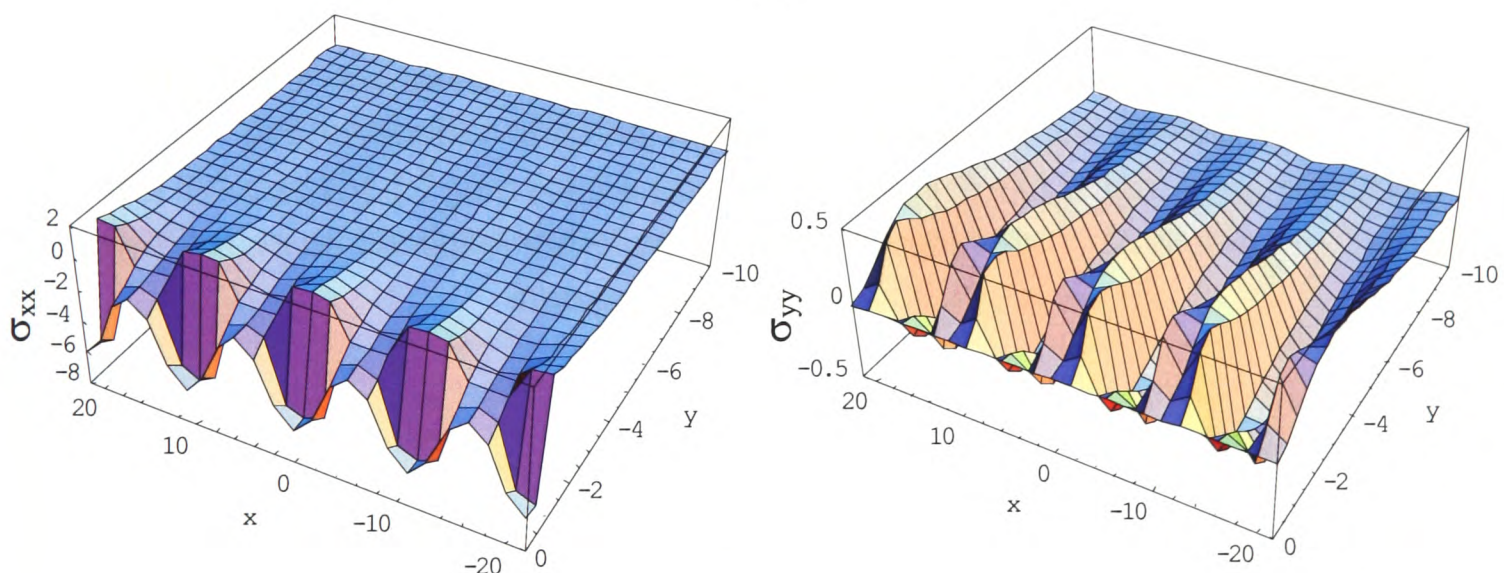




(b)



(c)



(d)

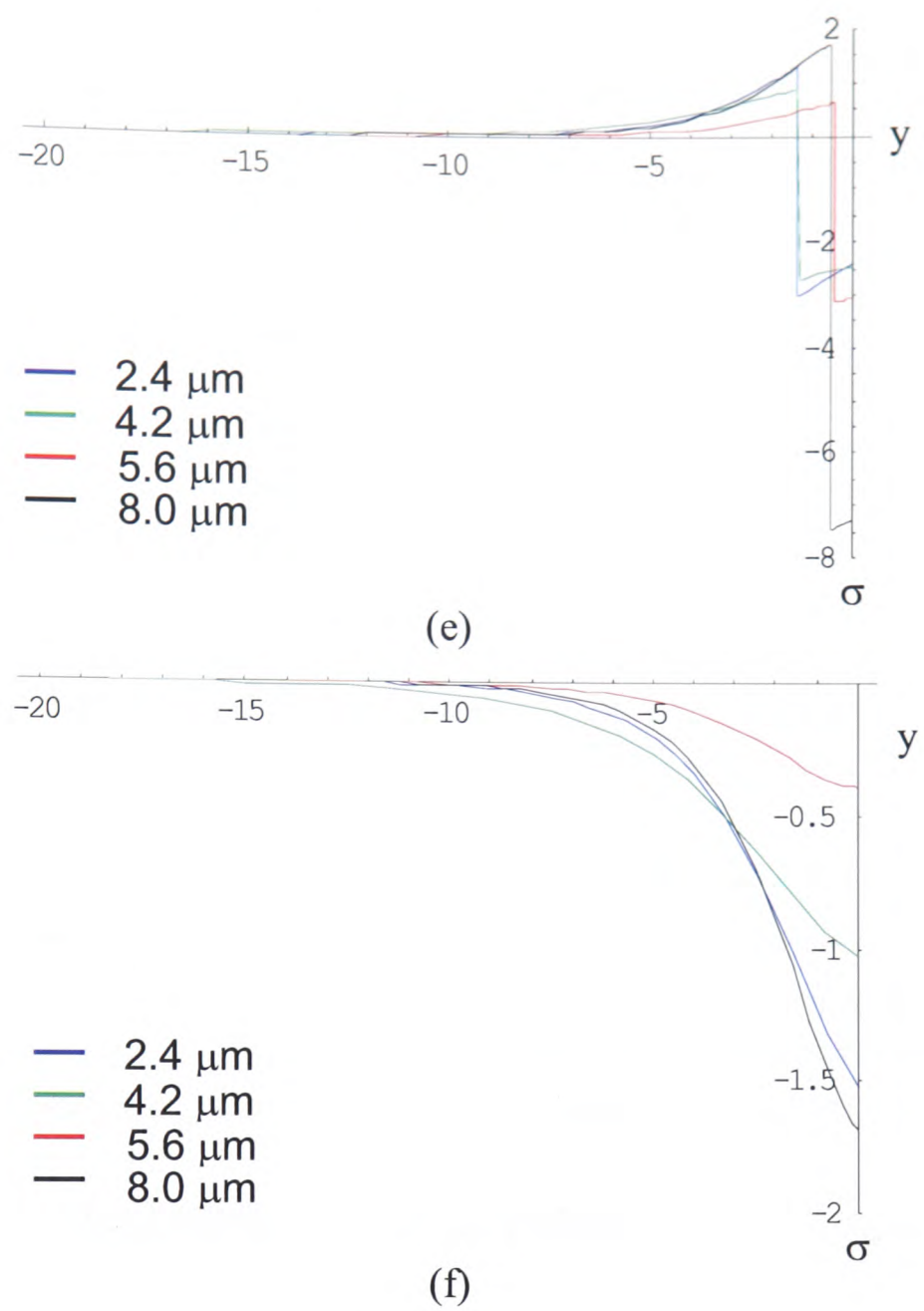


Fig 5.24 Fitted stress components  $\sigma_{xx}$  and  $\sigma_{yy}$  for four grain sizes of ground polycrystalline alumina, using  $D'$ ,  $d$ ,  $s$  and  $p$  listed in Table 5.4. (a) 2.4  $\mu\text{m}$ ; (b) 4.2  $\mu\text{m}$ ; (c) 5.6  $\mu\text{m}$ ; (d) 8.0  $\mu\text{m}$ ; (e) and (f): total normal stress distribution on the depth direction for ground region and pullout region. Units of stresses are GPa and  $y$  is in  $\mu\text{m}$ .

Table 5.5 List of maximum tensile and compressive stresses for ground regions and pullout regions of the four ground aluminas as shown in Fig 5.24

| G(μm) | Ground Region (x=0)       |                           |                         | Pullout Region (x=(s+p)/2) |                         |
|-------|---------------------------|---------------------------|-------------------------|----------------------------|-------------------------|
|       | $\sigma_{xx} (^1t_{max})$ | $\sigma_{xx} (^2c_{max})$ | $\sigma_{yy} (t_{max})$ | $\sigma_{xx} (c_{max})$    | $\sigma_{yy} (c_{max})$ |
|       | (GPa)                     | (GPa)                     | (GPa)                   | (GPa)                      | (GPa)                   |
| 2.4   | 0.94                      | -2.58                     | 0.27                    | -1.23                      | -0.23                   |
| 4.2   | 0.66                      | -2.23                     | 0.18                    | -0.83                      | -0.17                   |
| 5.6   | 0.48                      | -2.53                     | 0.12                    | -0.32                      | -0.08                   |
| 8.0   | 1.32                      | -6.09                     | 0.35                    | -1.35                      | -0.32                   |

| G(μm) | Ground Region (x=0)                       |                                           | Pullout Region (x=(s+p)/2)                |
|-------|-------------------------------------------|-------------------------------------------|-------------------------------------------|
|       | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ |
|       | (t <sub>max</sub> ) (GPa)                 | (c <sub>max</sub> ) (GPa)                 | (c <sub>max</sub> ) (GPa)                 |
| 2.4   | 1.33                                      | -3.03                                     | -1.52                                     |
| 4.2   | 0.89                                      | -2.70                                     | -1.03                                     |
| 5.6   | 0.62                                      | -3.12                                     | -0.32                                     |
| 8.0   | 1.72                                      | -7.48                                     | -1.68                                     |

<sup>1</sup> maximum tensile stresses  
<sup>2</sup> maximum compressive stresses

### 5.6.2 Polishing

In principle, the same technique can be used to measure the residual stresses in polished materials. The fitting results are given in Fig 5.25, 5.26, 5.27 and 5.28, and the  $D'$ ,  $d$  from the best fit to experimental results are listed in Table 5.6. The model shows that, for the polished regions the stresses below the dislocation array will be very close to zero and stresses within it will be compressive and nearly constant (because  $s$  is much larger than  $p$ ). The narrow peaks and broad peaks from polished regions agree with this prediction very well, although scatter for narrow peaks was seen for all four different grain sized aluminas. As a result, the achieved best fit was based on the broad peaks from the polished regions and peaks from the pullout regions.

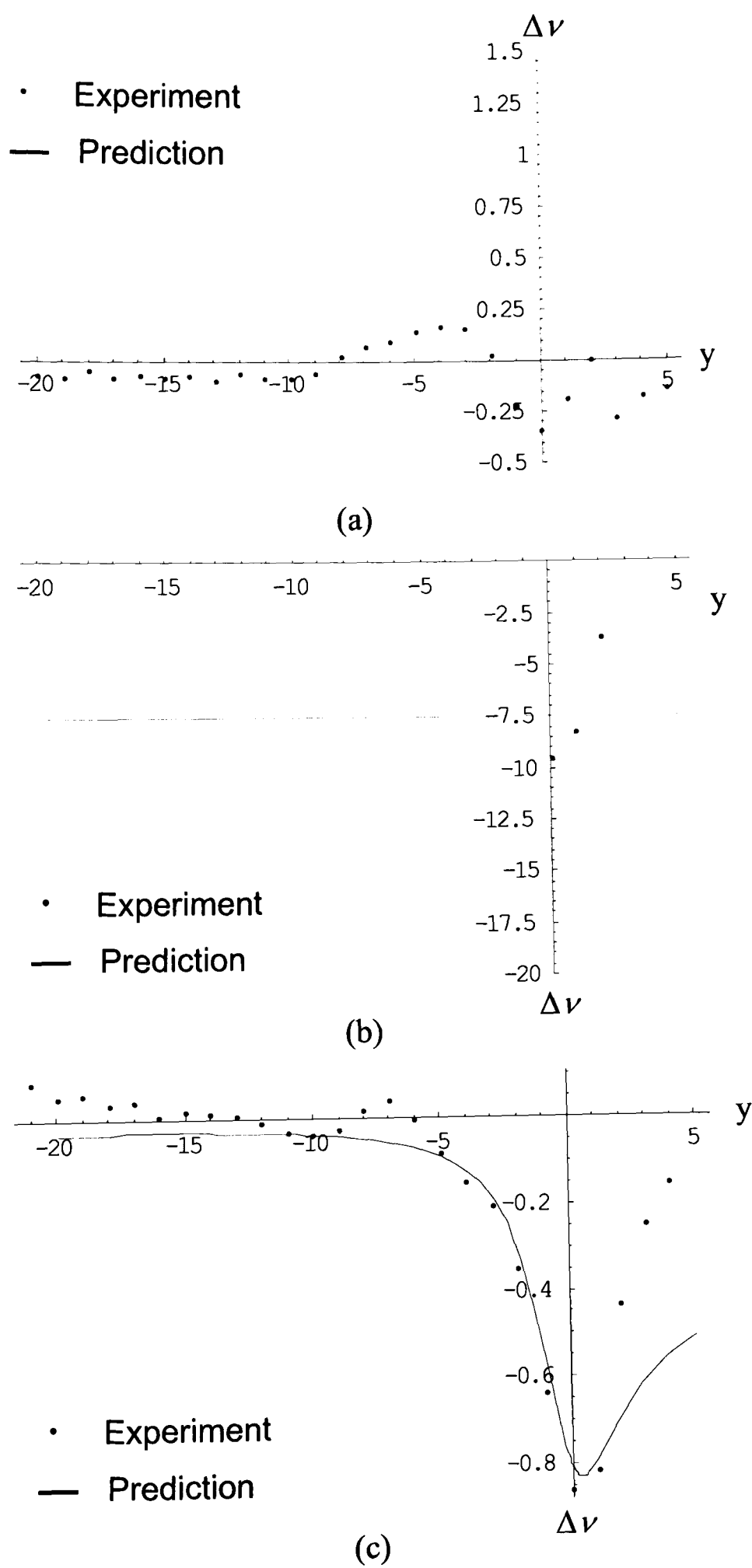


Fig 5.25 Fitting results for 2.4 μm sized polished alumina  
(a) narrow peaks from polished region; (b) broad peaks from polished region; (c)  
pullout region. Unit of  $\Delta\nu$  is cm<sup>-1</sup> and  $y$  is in μm.



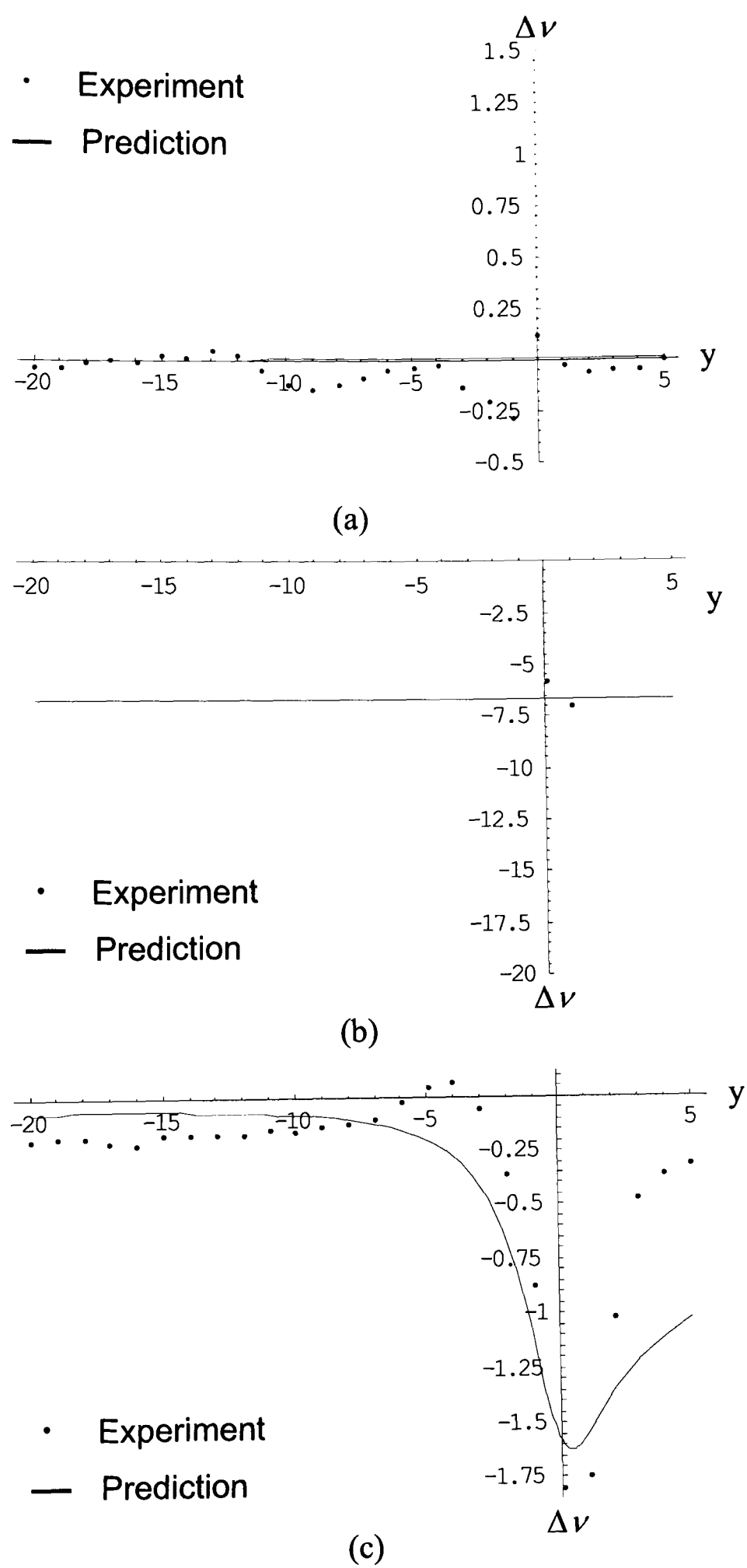


Fig 5.26 Fitting results for 4.2 μm sized polished alumina  
(a) narrow peaks from polished region; (b) broad peaks from polished region; (c) pullout region. Unit of  $\Delta\nu$  is cm<sup>-1</sup> and  $y$  is in μm.

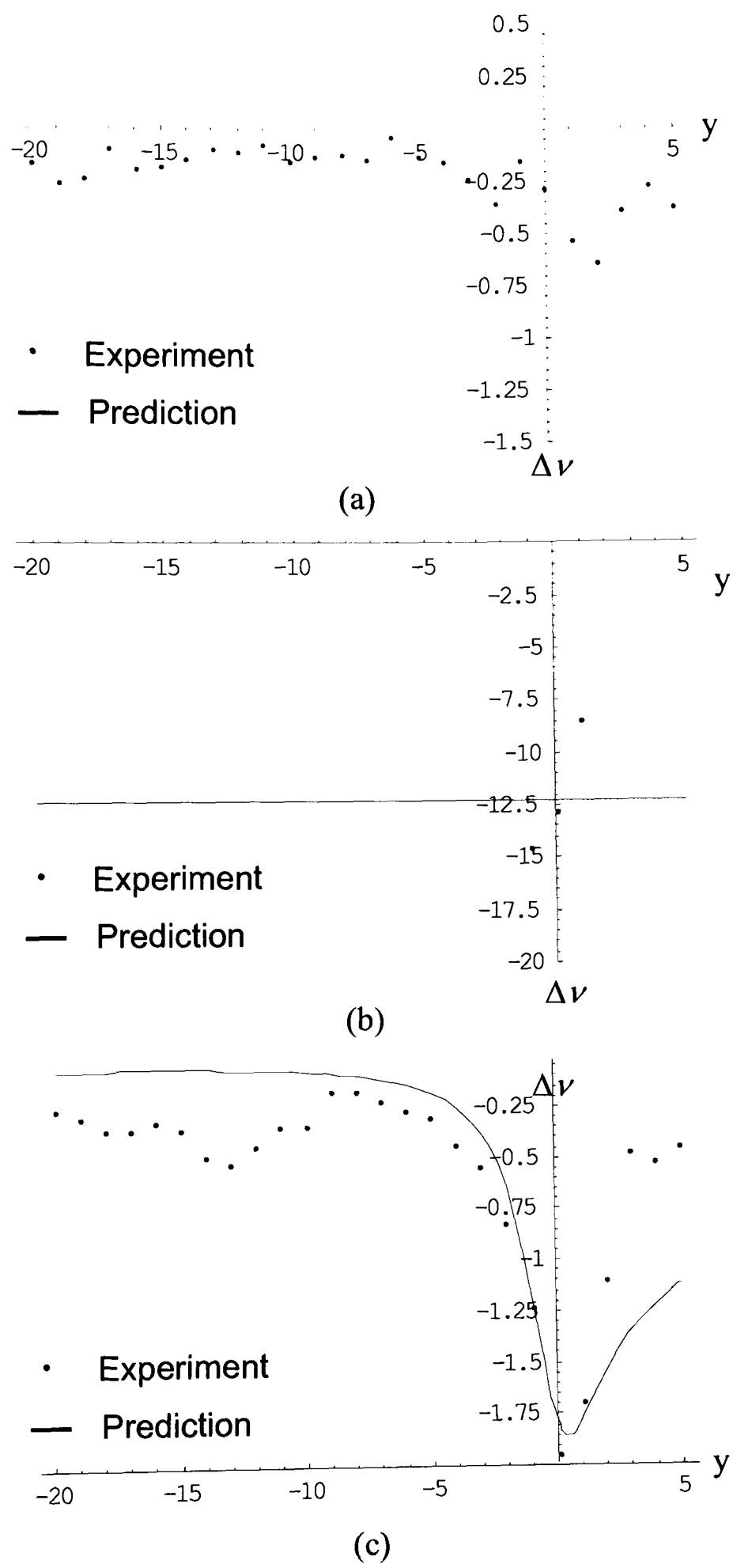


Fig 5.27 Fitting results for 5.6  $\mu\text{m}$  sized polished alumina  
(a) narrow peaks from polished region; (b) broad peaks from polished region; (c)  
pullout region. Unit of  $\Delta\nu$  is  $\text{cm}^{-1}$  and  $y$  is in  $\mu\text{m}$ .

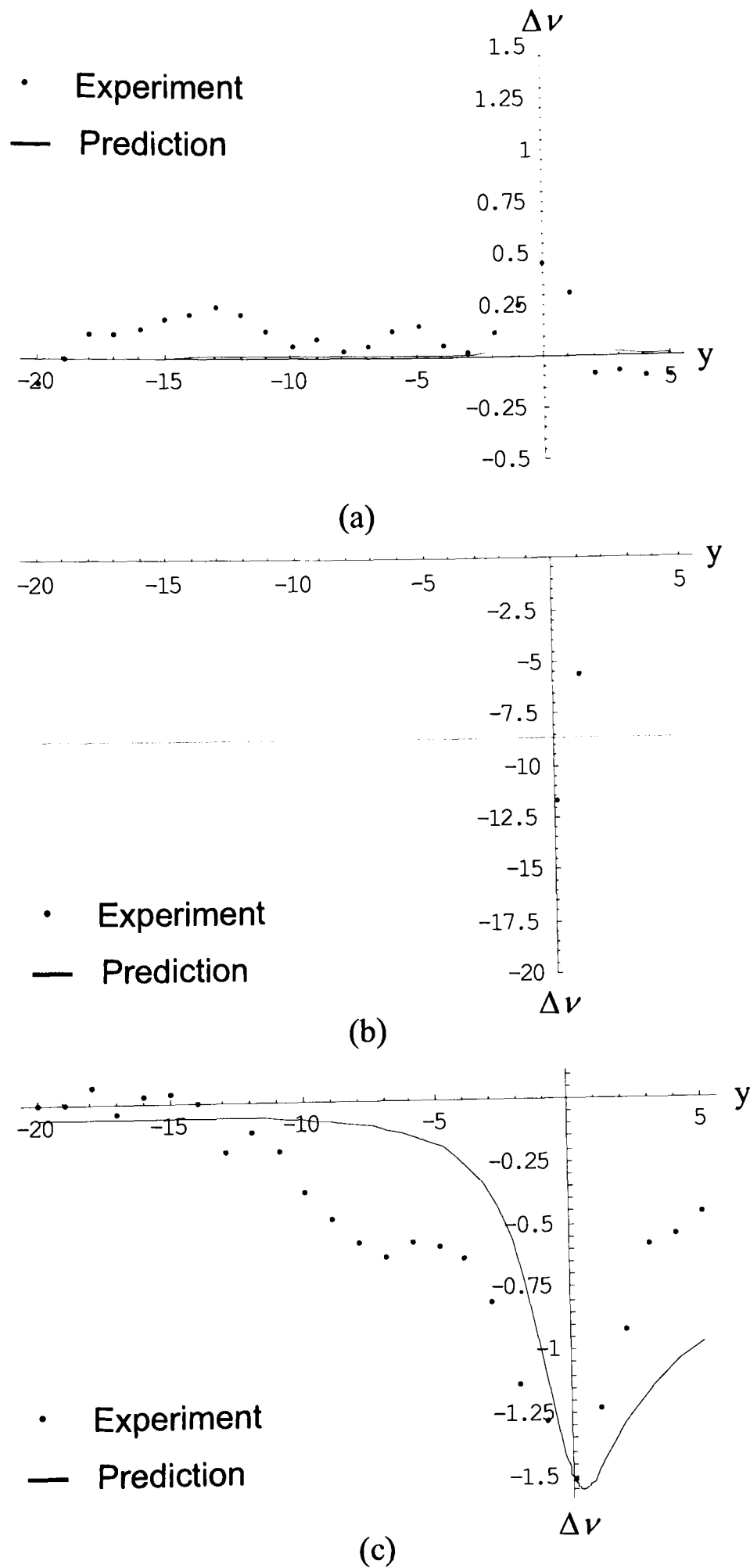
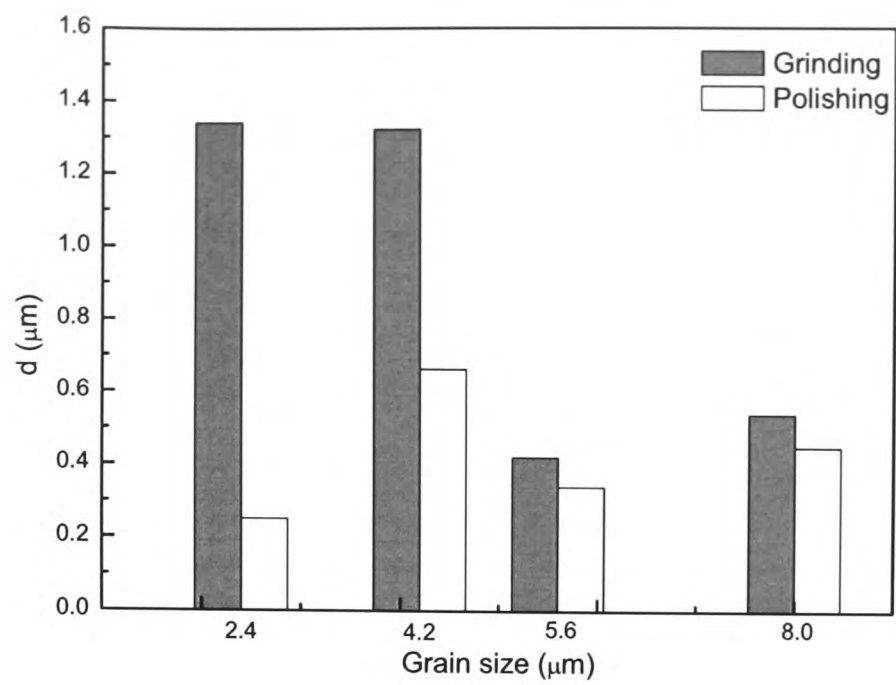


Fig 5.28 Fitting results for 8.0 μm sized polished alumina  
 (a) narrow peaks from polished region; (b) broad peaks from polished region; (c)  
 pullout region. Unit of  $\Delta\nu$  is cm<sup>-1</sup> and  $y$  is in μm.

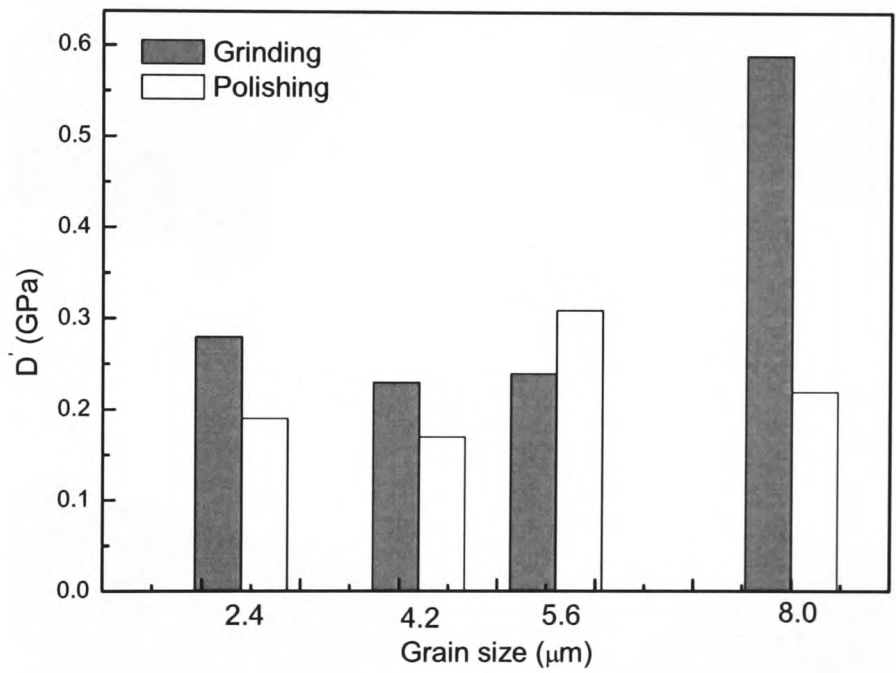
Table 5.6 List of  $D'$ ,  $d$ ,  $s$  and  $p$  for four polished polycrystalline alumina.  
 $D'$  and  $d$  are from the best fit of convolution to the experimental results;  $s$  and  $p$  are measured from optical images.

| $G(\mu\text{m})$ | $D'(\text{GPa})$ | $d(\mu\text{m})$ | $D'\cdot d \text{ (GPa}\cdot\mu\text{m)}$ | $s(\mu\text{m})$ | $p(\mu\text{m})$ | $s/p$ |
|------------------|------------------|------------------|-------------------------------------------|------------------|------------------|-------|
| 2.4              | 0.19             | 0.25             | 0.05                                      | 114.5            | 2.0              | 56.7  |
| 4.2              | 0.17             | 0.66             | 0.11                                      | 94.5             | 2.4              | 38.6  |
| 5.6              | 0.31             | 0.34             | 0.10                                      | 184.6            | 1.6              | 112.6 |
| 8.0              | 0.22             | 0.45             | 0.10                                      | 53.0             | 2.3              | 22.8  |

Compared with the grinding condition, the depth of plastic deformation in the polishing condition is smaller than in the grinding condition, as shown in Fig 5.29. This is in agreement with the TEM investigation by Wu et al. [8].  $D'$ , which is a measure of the extent of plastic deformation within the surface layer, is only slightly smaller in polishing than in grinding, except for the 8.0  $\mu\text{m}$  grain sized alumina owing to its high value obtained in grinding.



(a)

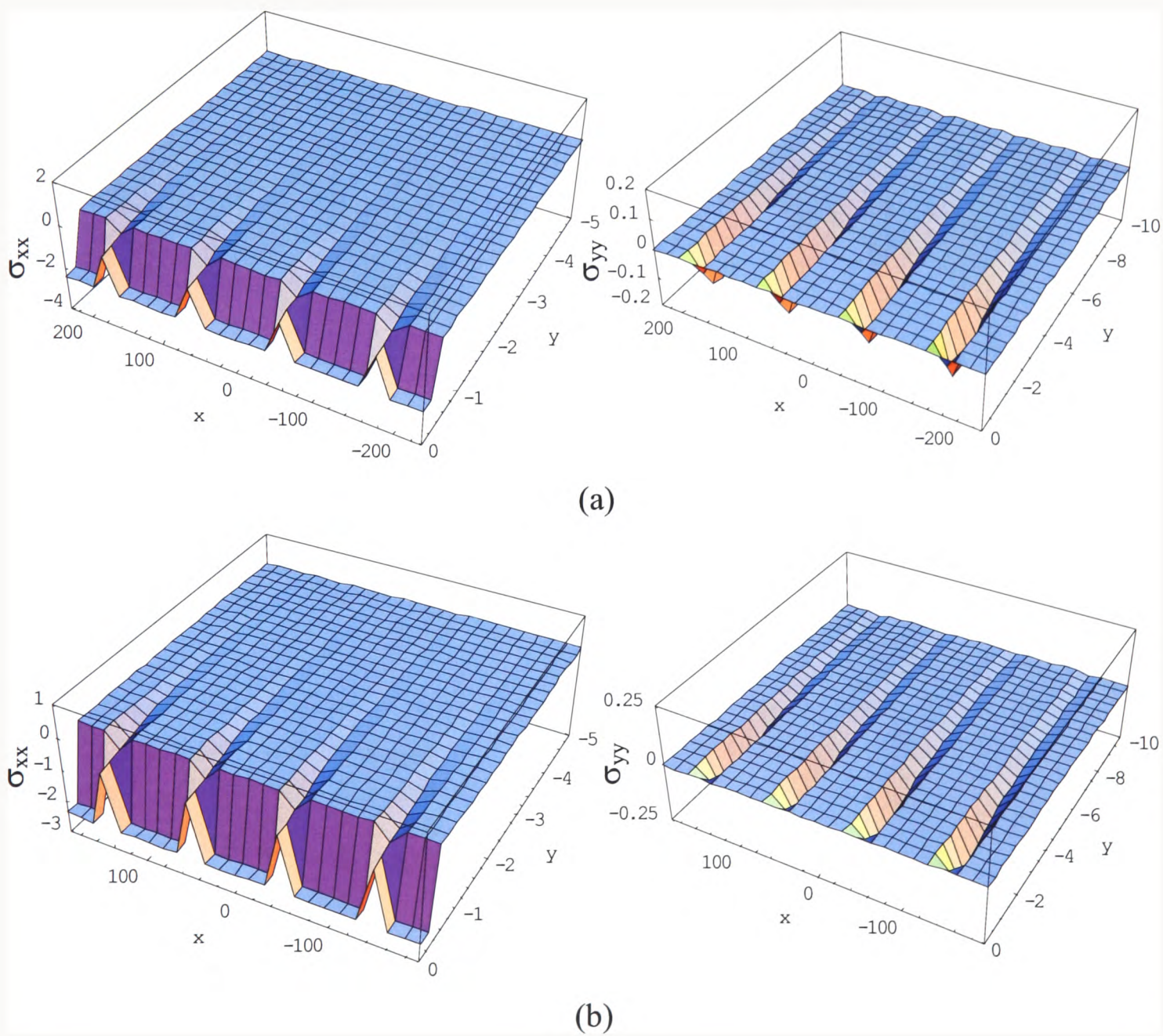


(b)

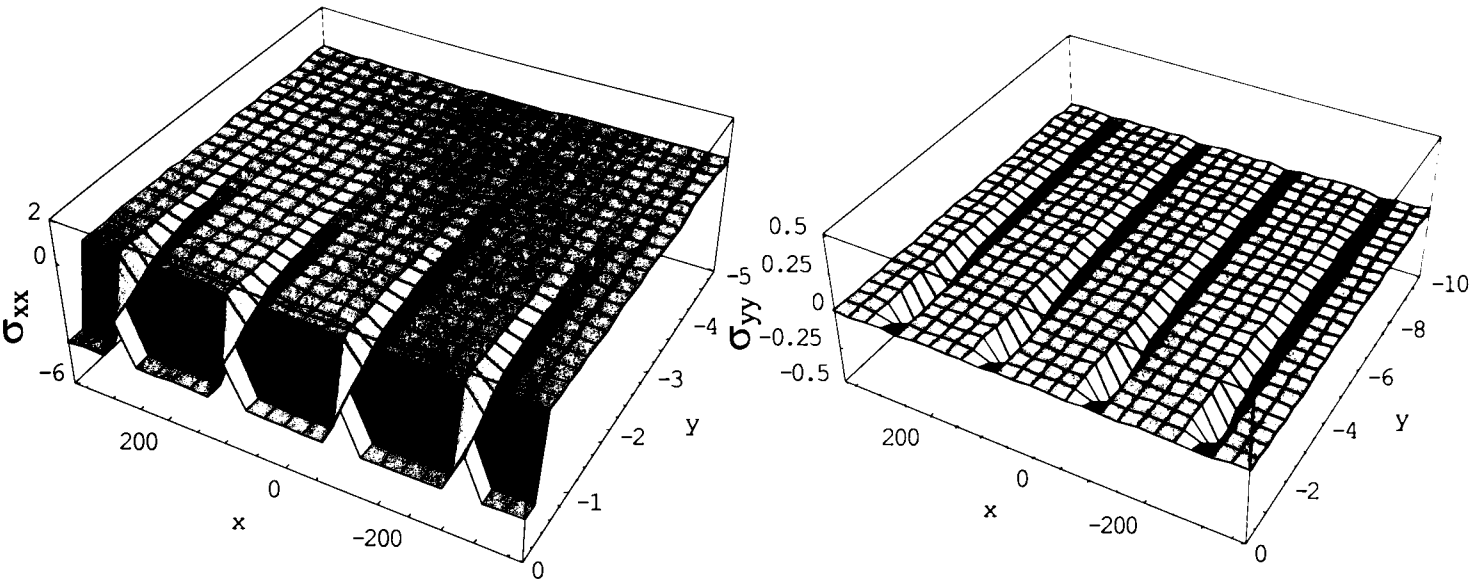
Fig 5.29 Comparison of  $d$  (a) and  $D'$  (b) for the four grain sized alumina fitted from grinding and polishing conditions

The real stress components  $\sigma_{xx}$  and  $\sigma_{yy}$  are shown in Fig 5.30, by using the values of  $D'$ ,  $d$ ,  $s$  and  $p$  in Table 5.6. The one-dimensional total normal stress distribution on the depth direction only is also shown. Significantly different to grinding, tensile stresses beyond the depth of the dislocation array are not observed in the polishing conditions.

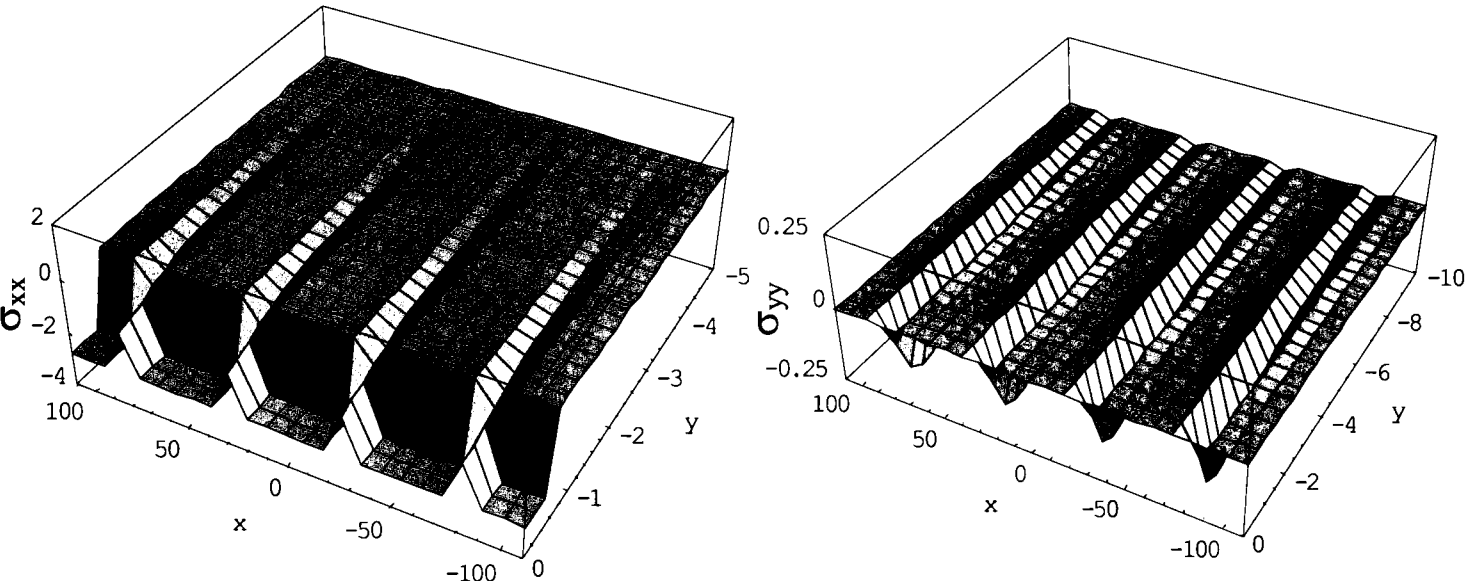
The pullouts in grinding condition cause tensile stresses that lead cracks to grow and degrade strength; in polishing, lack of pullouts cannot produce tensile stresses while remaining the compressive stresses. The fewer pullouts in polishing are mainly due to two reasons: smaller scratches result in fewer cracks being nucleated; smaller  $d$  and  $D'$  generate smaller tensile stresses when the pullouts are formed. The maximum compressive stresses on the polished region and pullout region are listed in Table 5.7.



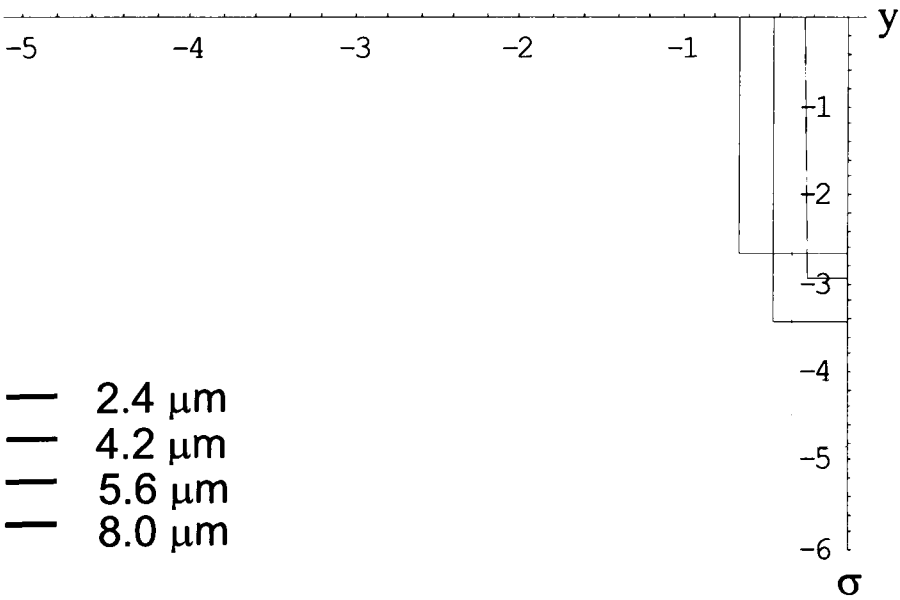




(c)



(d)



- 2.4  $\mu\text{m}$
- 4.2  $\mu\text{m}$
- 5.6  $\mu\text{m}$
- 8.0  $\mu\text{m}$

(e)

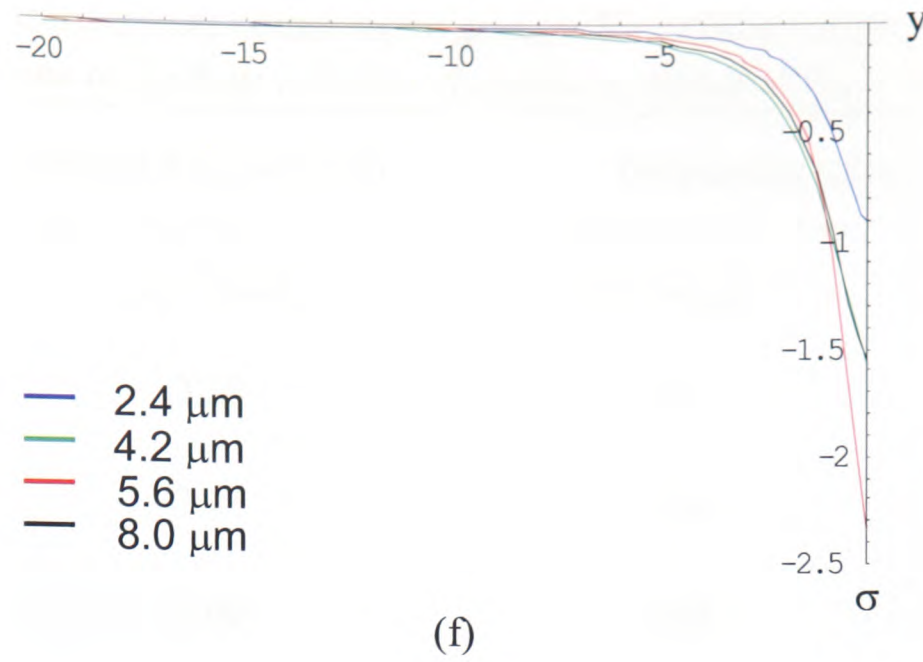


Fig 5.30 Fitted stress components  $\sigma_{xx}$  and  $\sigma_{yy}$  for four grain sized polished polycrystalline alumina, using  $D'$ ,  $d$ ,  $s$  and  $p$  listed in Table 5.6.

(a) 2.4  $\mu\text{m}$ ; (b): 4.2  $\mu\text{m}$ ; (c): 5.6  $\mu\text{m}$ ; (d): 8.0  $\mu\text{m}$ ; (e) and (f): total normal stress distribution on the depth direction for ground region and pullout region. Units of stresses are GPa and  $y$  is in  $\mu\text{m}$ .

Table 5.7 List of maximum compressive stresses for polished regions and pullout regions of the four polished aluminas as shown in Fig 5.30

| G(μm) | Ground Region (x=0)       | Pullout Region (x=(s+p)/2) |                         |
|-------|---------------------------|----------------------------|-------------------------|
|       | $\sigma_{xx} (^1c_{max})$ | $\sigma_{xx} (c_{max})$    | $\sigma_{yy} (c_{max})$ |
| 2.4   | -2.39                     | -0.73                      | -0.14                   |
| 4.2   | -2.13                     | -1.26                      | -0.20                   |
| 5.6   | -3.90                     | -1.89                      | -0.32                   |
| 8.0   | -2.76                     | -1.25                      | -0.22                   |

| G(μm) | Ground Region (x=0)                       | Pullout Region (x=(s+p)/2)                |
|-------|-------------------------------------------|-------------------------------------------|
|       | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ |
|       | (c <sub>max</sub> ) (GPa)                 | (c <sub>max</sub> ) (GPa)                 |
| 2.4   | -2.96                                     | -0.90                                     |
| 4.2   | -2.64                                     | -1.56                                     |
| 5.6   | -4.83                                     | -2.34                                     |
| 8.0   | -3.42                                     | -1.55                                     |

<sup>1</sup> maximum compressive stress

5.7 Summary

1) The residual stresses on the ground and polished polycrystalline aluminas depend greatly on the local environment: ground/polished region or pullout region. Both tensile and compressive stresses exceeding 1 GPa were present in the ground surface of aluminas.

2) Peak fitting for spectra near the ground/polished surfaces provides an important method of separating stresses in the ground region into two physically distinct regions: broad peaks representing the plastically deformed surface layer, and narrow peaks representing the elastically deformed material beneath.

3) A model taking into account the pullout and modelling the residual stress field using an array of dislocations was proposed. The experimental results supported the main features of the model when the resolution function of the microscope is taken into account.

4) The depth of plastic deformation was found to be deeper in the ground condition than in polished condition, for the same grain sized alumina. Tensile stresses produced by the pullouts in the ground condition would lead to crack growth and strength degradation; however, in the polished condition, the lack of pullouts prevented tensile stresses arising while the compressive stresses remained. The residual stress level in the ground condition is slightly higher than that in the polished condition.

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## Chapter 6

### Grinding of Al<sub>2</sub>O<sub>3</sub>/SiC Nanocomposites

#### 6.1 Introduction

Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites are polycrystalline alumina containing small fractions of sub-micron sized SiC particles. The typical microstructure of Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites is composed of a polycrystalline alumina matrix with an average grain size of 1-5  $\mu\text{m}$ , and SiC particles with size ranging from 100 to 200 nm which exist either inside the matrix or on the grain boundaries. They have attracted interest from researchers since Niihara [1-3] first reported a much improved strength and toughness compared to alumina with equivalent grain size. Further researches have confirmed the strength increase (but much less than Niihara claimed) of Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites over polycrystalline alumina, but a significant increase in toughness is not observed [4-9]. The lack of strengthening mechanism in Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites has motivated lots of research work and different explanations with their experimental support. Proposed strengthening mechanisms include microstructure refinement [4], grain-boundary strengthening [10, 11], decrease of grain pullout [12], crack deflection and bridging [13, 14], and also an increase in the compressive residual stresses after machining [9, 15, 16]. The effect of residual stress is the main topic in this chapter. Previous results on measuring the residual stresses in Al<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites have been reviewed in Chapter 2. They either measured the thermal stress (i.e., not machining induced residual stress) [17-20] or the



residual stress induced by indentation or scratching [21-23], or used other techniques rather than the Cr<sup>3+</sup> fluorescence microscopy to measure the grinding induced stress or strain [15, 16, 24-26]. In this chapter, the grinding induced residual stress in alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites was investigated using the Cr<sup>3+</sup> fluorescence microscopy method, specifically by using the depth scanning method described in Chapter 5. The same model used in Chapter 5 convoluting the predictions of an elastic model for the surface and the PRF in the depth direction was applied.

## 6.2 Experimental Results

### 6.2.1 Microstructure

An alumina specimen of average grain size 5.6 μm and three Al<sub>2</sub>O<sub>3</sub>/x vol. % SiC (x=2, 5, 10) nanocomposites with similar grain size to alumina (see Table 3.1 in Chapter 3) were ground using 45 μm grit diamond on an epoxy resin bonded alumina wheel for 4 minutes. The scanning electron microscope (SEM) observed ground surfaces are shown in Fig 6.1, with the measured mean pullout size and area fraction of pullout given in Table 6.1.

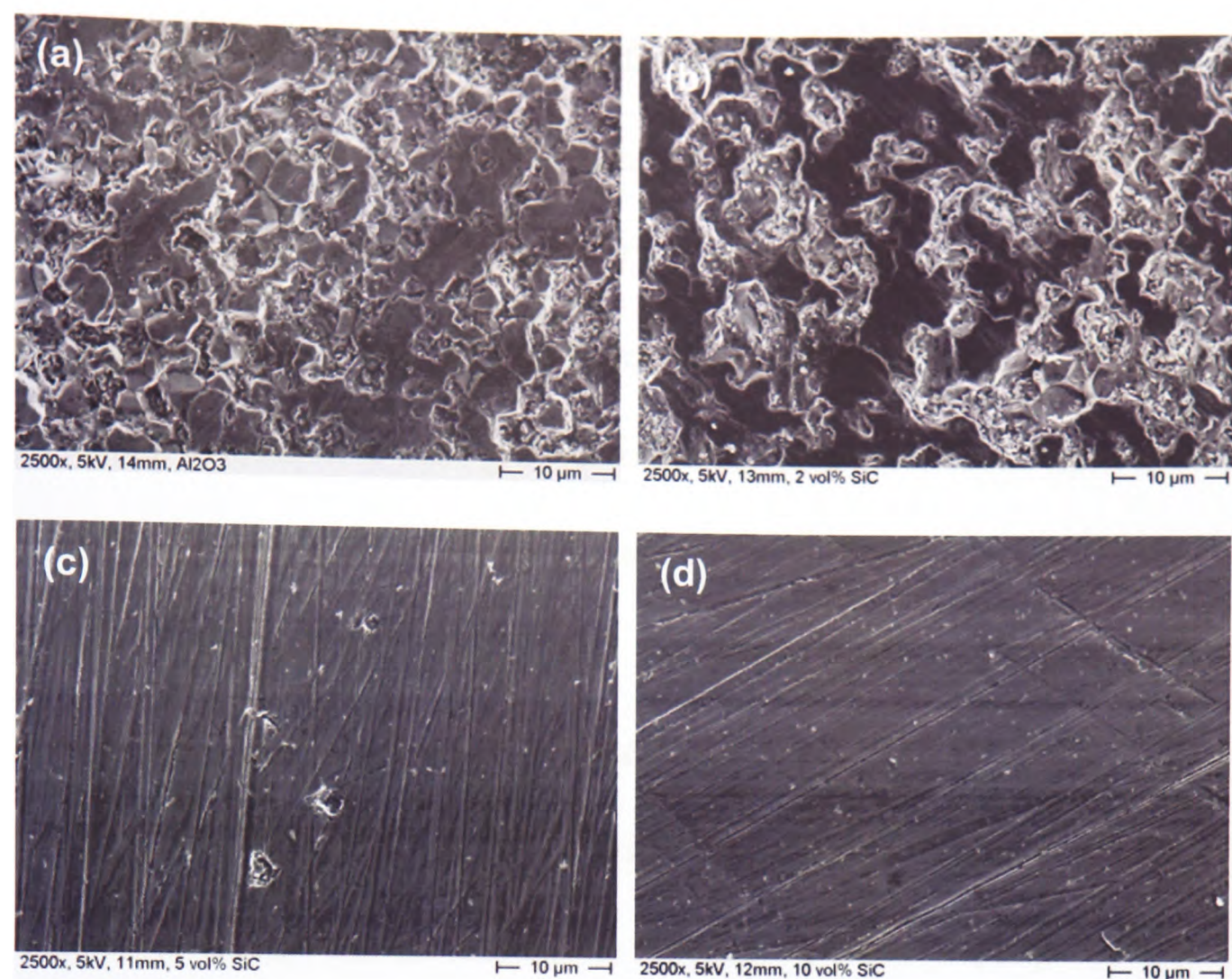


Fig 6.1 SEM images of ground surfaces of alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites (a) Al<sub>2</sub>O<sub>3</sub> (b) 2 vol. % SiC (c) 5 vol. % SiC and d) 10 vol. % SiC. Courtesy of Apichart Limpichaipanit.

Table 6.1 List of average grain size of alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites, and their pullout size and fraction of pullout after 45 μm grit grinding (courtesy of Apichart Limpichaipanit)

| Materials                      | Average Grain Size<br>(μm) | Mean Pullout Size<br>(μm) | Area Fraction<br>of Pullout (%) |
|--------------------------------|----------------------------|---------------------------|---------------------------------|
| Al <sub>2</sub> O <sub>3</sub> | 5.6                        | 5.5                       | 60±7                            |
| 2 vol.% SiC                    | 6.2                        | 4.1                       | 50±1                            |
| 5 vol.% SiC                    | 5.3                        | 1.9                       | 3±1                             |
| 10 vol.% SiC                   | 4.9                        | 1.7                       | 1.2±0.4                         |

There are two phenomena can be clearly observed from the SEM images:

1) The nanocomposites have better surface quality after grinding, which can be attributed to the decrease of pullout size and also the suppression of pullouts [9, 12]. The pullout size decreases as the amount of SiC addition increases. Todd et al. [12, 27] used a simple geometrical model to explain this as the result of fracture mode from inter-granular in alumina to trans-granular in nanocomposites. Nanocomposites have a much smaller area fraction of pullouts compared with alumina, especially when the volume percentage of SiC reaches 5%; for 5 vol. % SiC and 10 vol. % SiC nanocomposites, only tiny amount of pullout can be observed. The large amount of nano-sized SiC addition (10 vol. %) can possibly block the formation of long twins and dislocation pileups that are responsible for crack initiation [27, 28].

2) For alumina, the material removal is mainly by grain dislodgement, or intergranular fracture; when 2 vol. % of SiC is added, the main material removal mechanism changes from inter-granular fracture to trans-granular fracture. When more than 5vol% of SiC is added, the main material removal mechanisms become plastic deformation, in contrast to the brittle fracture in alumina, which is reflected in the grooves seen in Fig 6.1 (c) and (d).

These phenomena are also in agreement with observations from previous reports [12, 15, 25]. More details about the difference of grinding behavior between alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites can be found from the above mentioned literature. The main topic in this chapter is the measuring and modeling of the residual stresses

introduced by grinding, with the aim of determining how different the residual stresses in alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites are, and whether the difference in residual stresses, if any, is the mechanism behind the different strength between alumina and the nanocomposites. In the following sections, the residual stresses in ground alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites will be investigated by the depth scanning method used in Chapter 5.

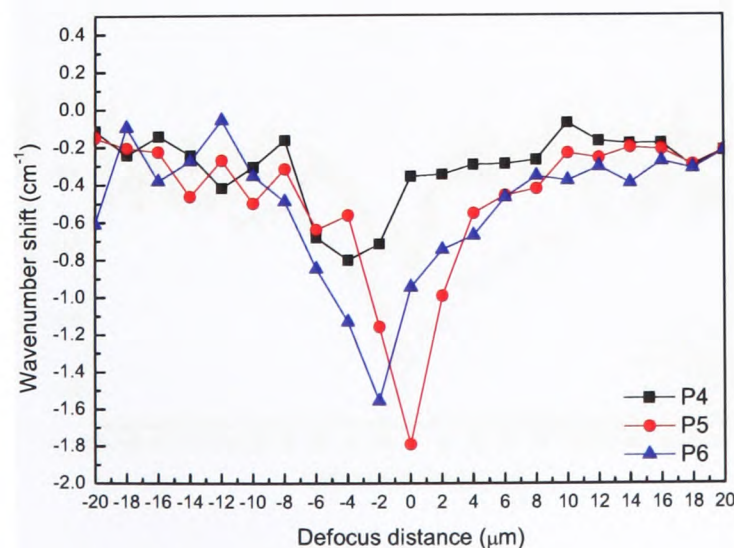
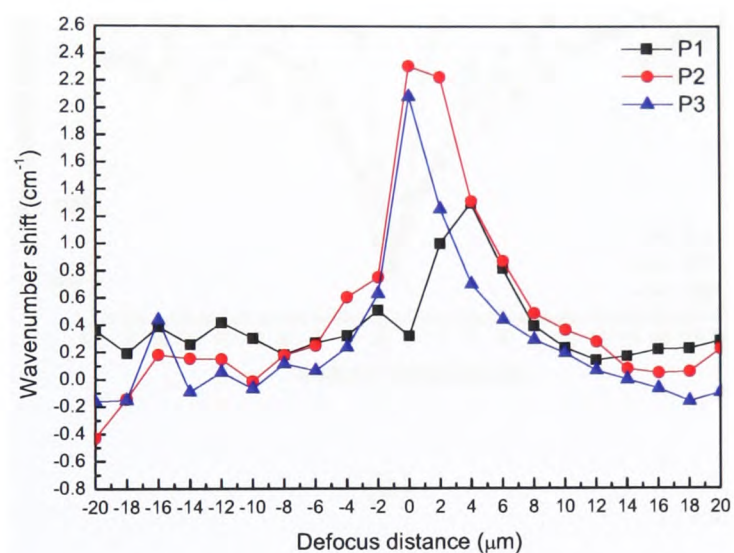
### 6.2.2 Depth Profiling

Depth scans through the ground regions and the pullout regions were carried out on alumina and  $\text{Al}_2\text{O}_3/x$  vol. % SiC ( $x=2, 5, 10$ ) nanocomposites. The depth profiles of these four materials are given in Fig 6.2, with the abscissa being the distance away from surface and the ordinate being R1 wavenumber shift compared to the reference position. The reference positions were determined from the averaged R1 positions (spectra collected on 20 random points) on the individual 1  $\mu\text{m}$  grit polished specimens.

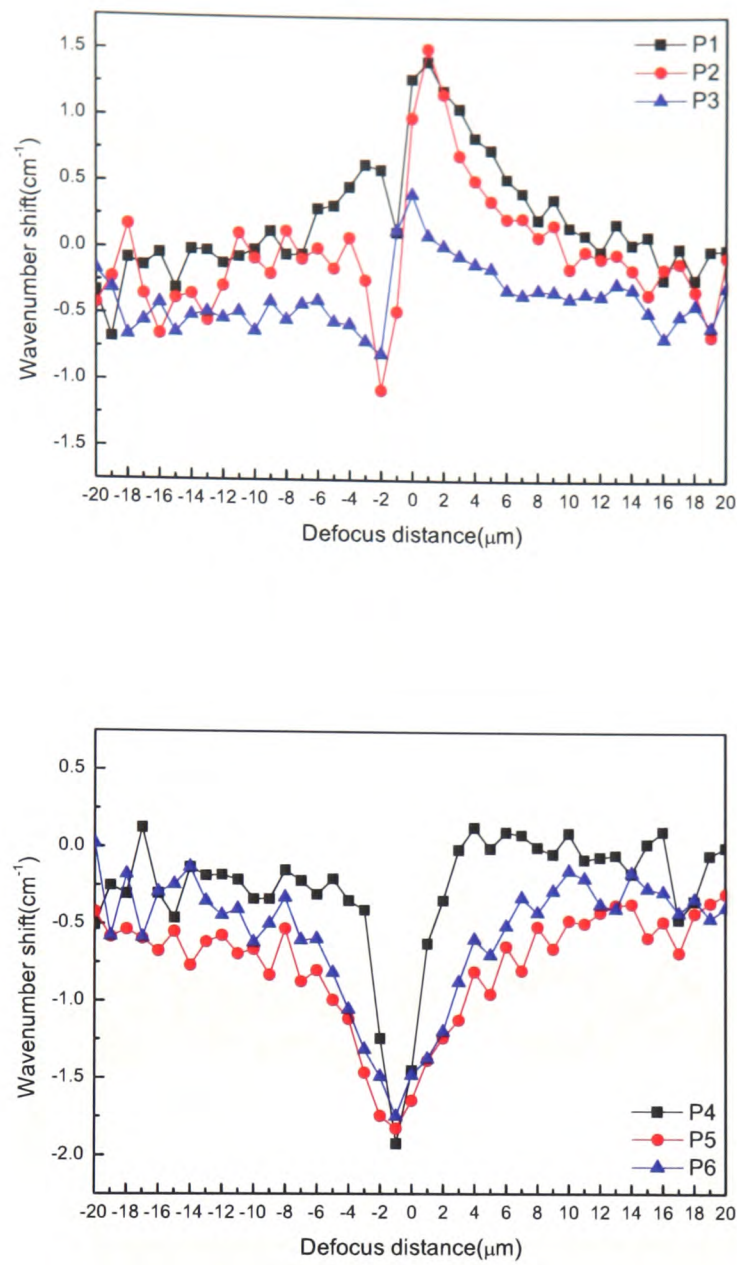
Seen from Fig 6.2, the measured wavenumber shifts from pullout regions were all negative for both alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites, which indicates the compressive stresses in these regions. This is the same as was observed for polycrystalline alumina with different grain sizes in Chapter 5. For the 10 vol. % SiC nanocomposite, it was difficult to locate the pullouts under the optical microscope, so only depth profiles from ground regions were given. Positive wavenumber shifts were



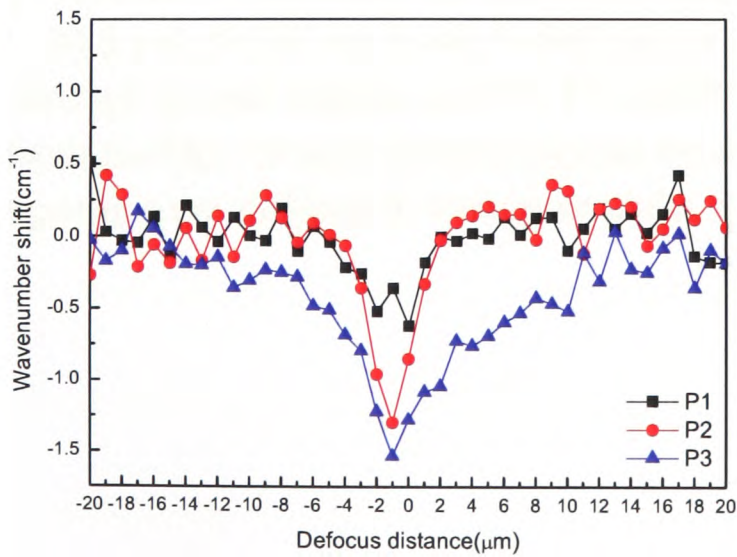
found in the ground regions of alumina and 2 vol. % SiC nanocomposite, indicating the existence of tensile stresses. However, for 5 and 10 vol. % SiC nanocomposites depth profiles from the ground regions showed less consistent results. The smaller pullout size and lower pullout fraction in the 3  $\mu\text{m}$  grit polished alumina were responsible for the disappearance of tensile stresses as has been explained in section 5.6.2 of Chapter 5. The possibility that this is also the explanation for the similar results in the 5 and 10 vol.% nanocomposites will be discussed in the following section.



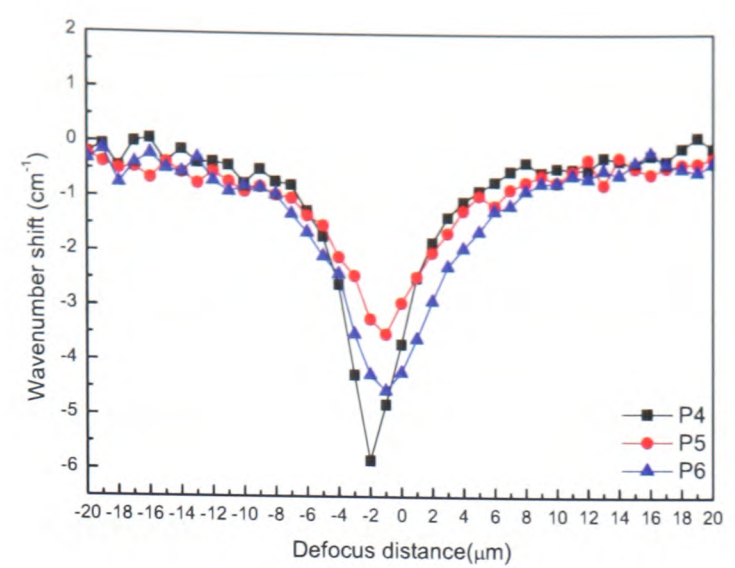
(a)



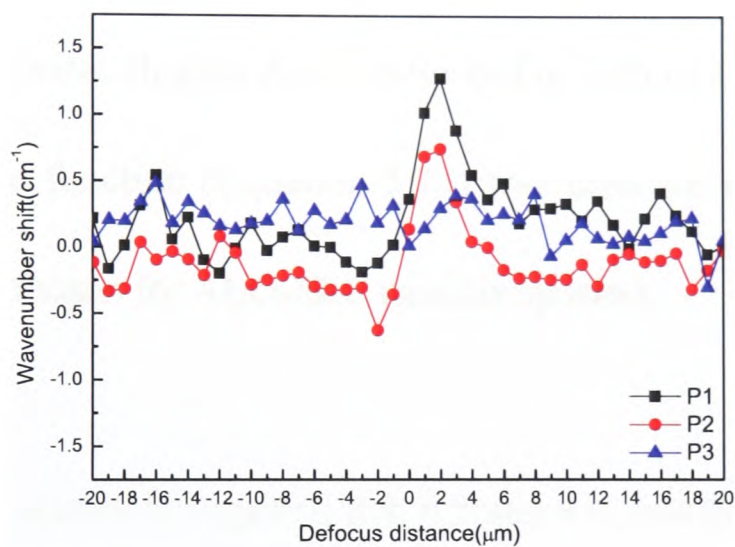
(b)







(c)



(d)

**Fig 6.2** Depth profiles of alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites  
(a) Al<sub>2</sub>O<sub>3</sub> (b) 2 vol. % SiC (c) 5 vol. % SiC (d) 10 vol. % SiC  
P1, P2 and P3 are through ground regions and P4, P5 and P6 are through pullout regions. In (d) only depth profiles through ground regions are shown as pullouts in 10 vol. % SiC nanocomposite were difficult to recognize under the optical microscope

### 6.3 Fitting Results and Discussion

The same fitting procedure as was used for polycrystalline alumina in Chapter 5 was applied to the nanocomposites, i.e. using the convolution of stress function and resolution function to fit the narrow peaks and broad peaks from ground regions, and also peaks from pullout regions. Note that in Chapter 5, a simplified resolution function was used (Equation 5.23) instead of the double-Lorentzian function (Equation 5.24) proposed in Chapter 4. Ignoring the second Lorentzian function has a small effect on the convolution for alumina but its effect is not negligible for  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites (bigger  $A_2/A_1$  ratio in Fig 5.20 of Chapter 5). As a result, the double-Lorentzian function (Equation 5.24) was used for the resolution function when doing the convolution for  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites.

The fitting results are shown in Figs 6.3, 6.4, 6.5 and 6.6, and the  $D'$ ,  $d$  from the fitting are listed in Table 6.2. For nanocomposites, the achieved best fit was judged from the broad peaks from the ground regions and peaks from pullout region (similar to the case for the 3  $\mu\text{m}$  grit polished polycrystalline alumina), as the narrow peaks from ground regions were difficult to fit, particularly for 5 and 10 vol. % SiC nanocomposites (see Fig 6.5 (a) and Fig 6.6 (a)). The same fitting criterion was used for alumina here to avoid the comparison being biased. The predicted wavenumber shift for narrow peaks from ground regions in 2 vol. % SiC nanocomposite, and for pullout regions in 5 vol.% SiC nanocomposites, were not closing to zero far below surface ( $y \ll 0$ ), as seen from Fig 6.4(a) and Fig 6.4 (c) and Fig 6.5 (c), which deviated

from the experimental results. It is because all the signal comes from near the surface due to absorption, even if the focus position is well below the surface. In this case, it is the detailed shape of the tails of the resolution function that determines the mean stress sampled and this is the reason that neither the real nor the predicted peak shifts approach zero as the focus depth increases. The disagreement between theory and experiment is also possibly due to the tail of the probe response function, which has been discussed in Chapter 5 (see Fig 5.18). This can also explain the unsatisfactory fittings above surface ( $y>0$ ) as discussed in section 5.6.1 in Chapter 5. As a result, the height of peak is the main thing to fit. Note that in Figs 6.5 (a) and 6.6 (a), the prediction suggests a nearly zero shift all along the depth, significantly different to those in Figs 6.3 (a) and 6.4 (a), which is certainly due to the convolution.

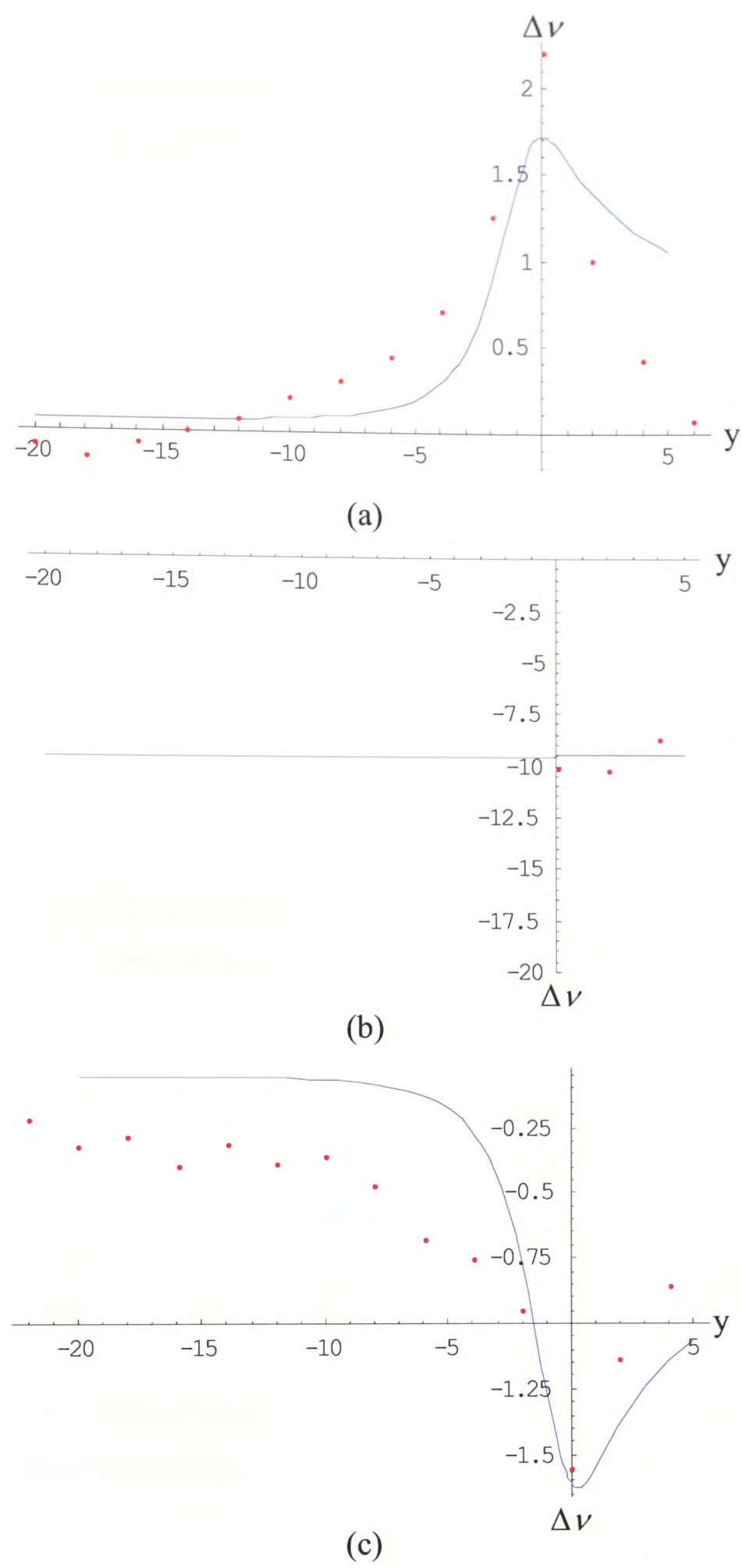
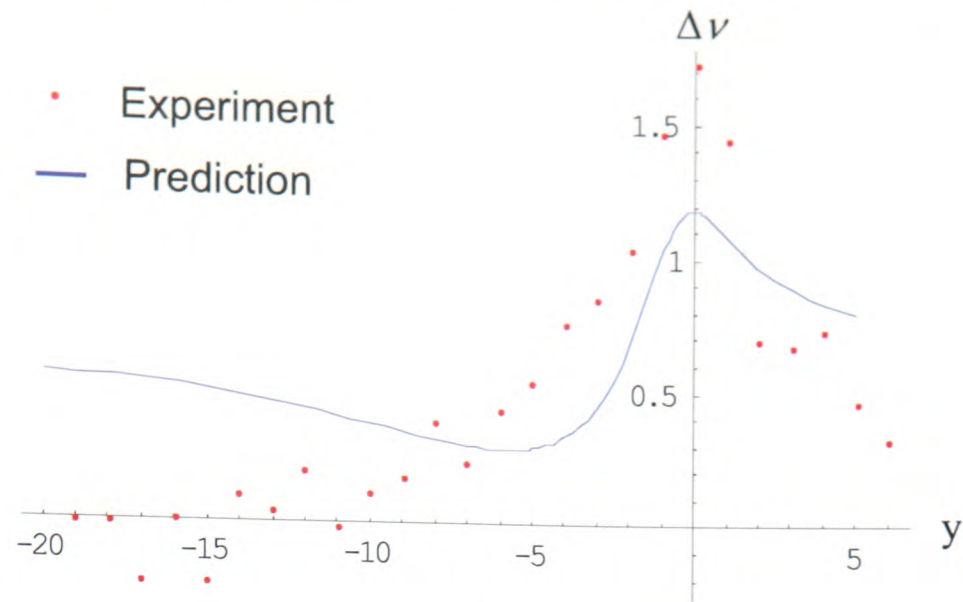
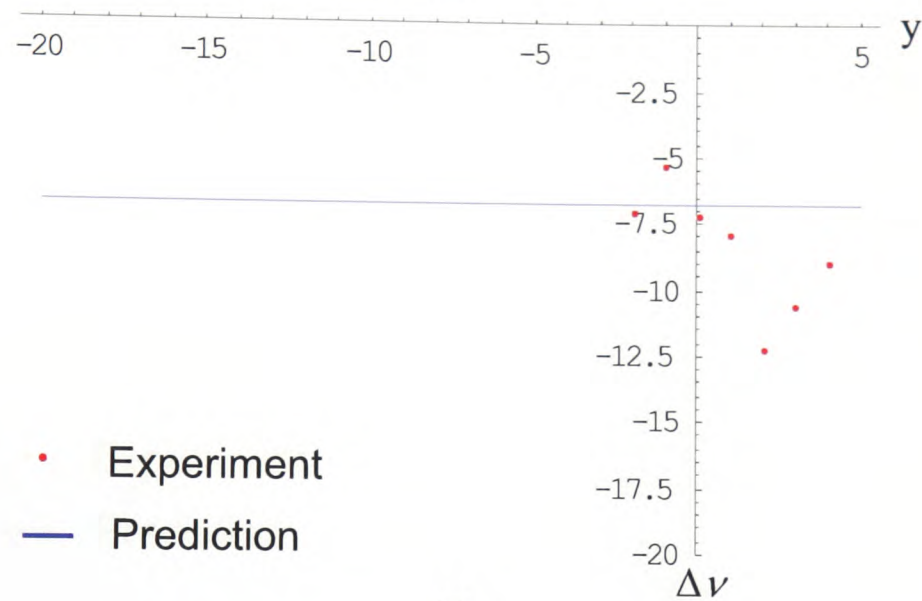


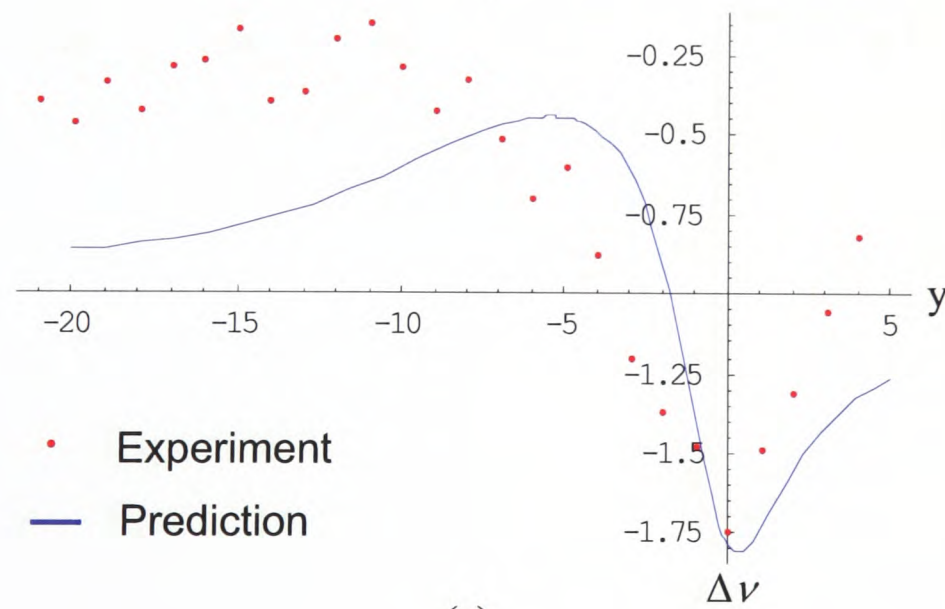
Fig 6.3 Fitting results for 5.6  $\mu\text{m}$  grain-sized Al<sub>2</sub>O<sub>3</sub>  
(a) narrow peaks from ground region; (b) broad peaks from ground region; (c) pullout region. Unit of  $y$  is in  $\mu\text{m}$  and  $\Delta\nu$  is in  $\text{cm}^{-1}$ .



(a)



(b)



(c)

Fig 6.4 Fitting results for 2.vol. % SiC  
 (a) narrow peaks from ground region; (b) broad peaks from ground region; (c) pullout region. Unit of  $y$  is in  $\mu\text{m}$  and  $\Delta\nu$  is in  $\text{cm}^{-1}$ .

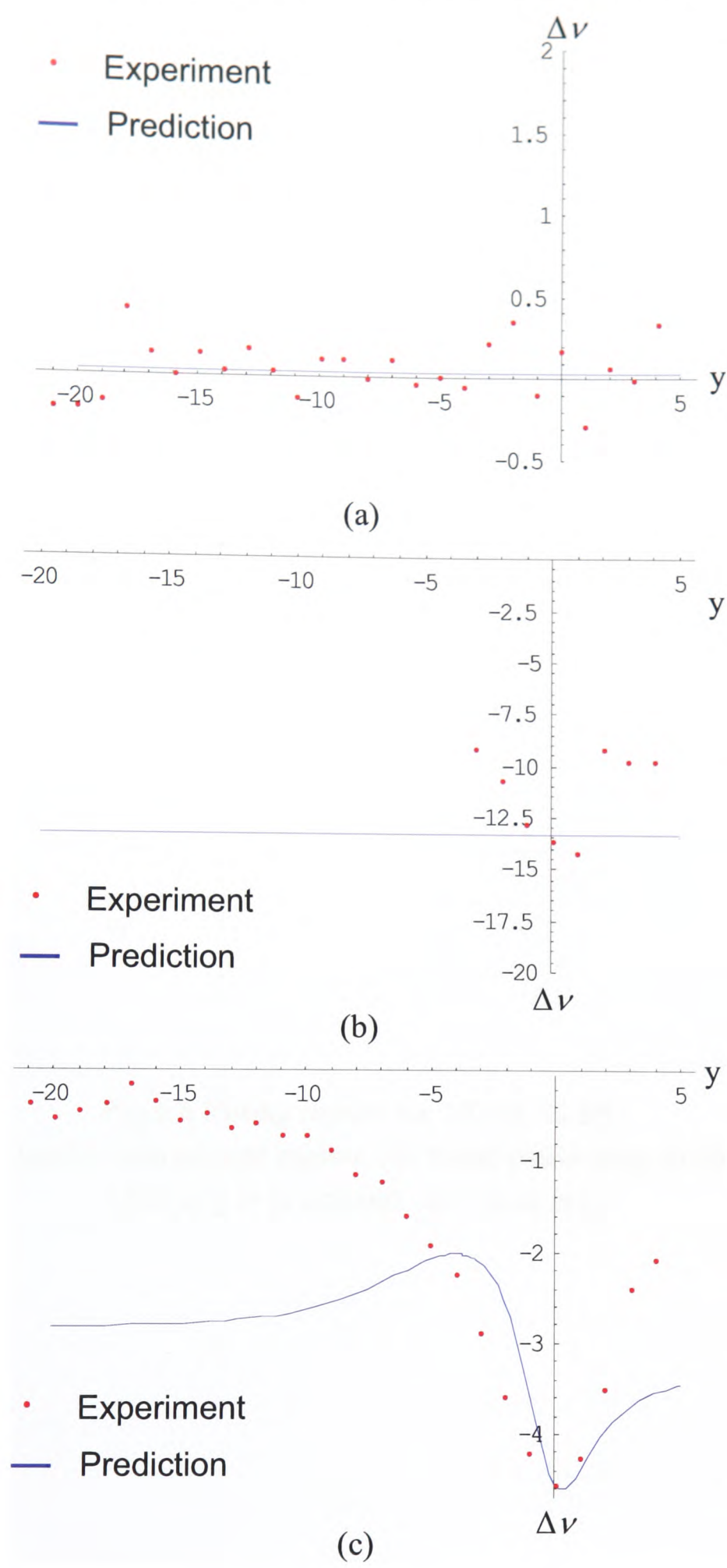


Fig 6.5 Fitting results for 5 vol. % SiC  
(a) narrow peaks from ground region; (b) broad peaks from ground region; (c) pullout region. Unit of  $y$  is in  $\mu\text{m}$  and  $\Delta\nu$  is in  $\text{cm}^{-1}$ .



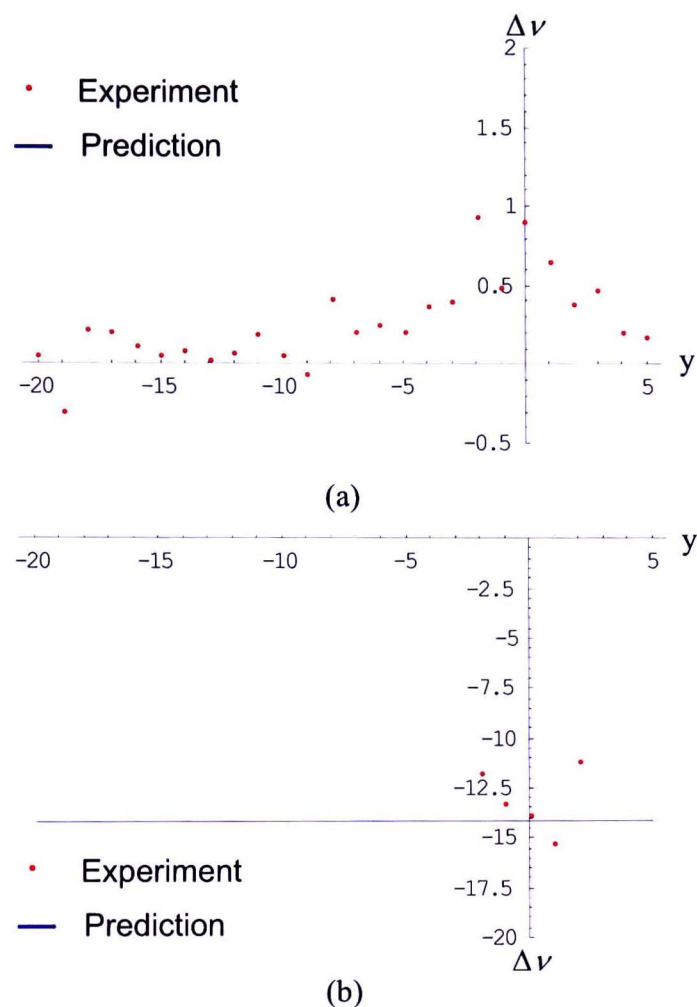


Fig 6.6 Fitting results for 10 vol. % SiC  
(a) narrow peaks from ground region; (b) broad peaks from ground region;  
Unit of y is in  $\mu\text{m}$  and  $\Delta \nu$  is in  $\text{cm}^{-1}$ .

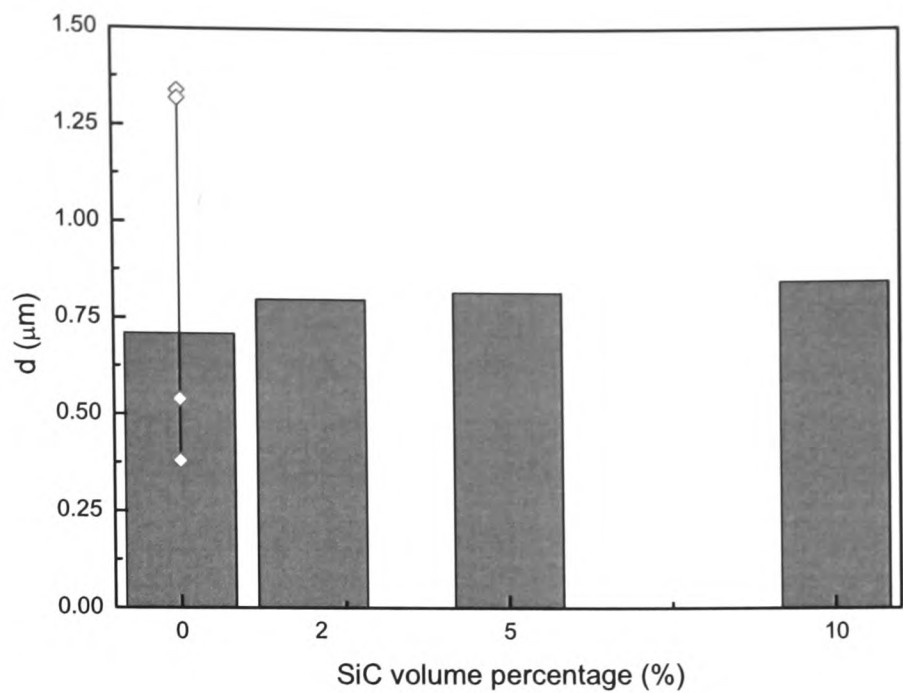
Table 6.2 List of  $D'$ ,  $d$ ,  $s$  and  $p$  for alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites  
 $D'$  and  $d$  are from the best fit of convolution to the experimental results;  $s$  and  $p$  are measured from SEM images

| Materials                      | $D'$ (GPa) | $d$ ( $\mu\text{m}$ ) | $D'\cdot d$ (GPa $\cdot\mu\text{m}$ ) | $s$ ( $\mu\text{m}$ ) | $p$ ( $\mu\text{m}$ ) | $s/p$ |
|--------------------------------|------------|-----------------------|---------------------------------------|-----------------------|-----------------------|-------|
| Al <sub>2</sub> O <sub>3</sub> | 0.37       | 0.71                  | 0.26                                  | 3.7                   | 5.5                   | 0.67  |
| 2 vol. % SiC                   | 0.25       | 0.80                  | 0.2                                   | 4.1                   | 4.1                   | 1     |
| 5 vol. % SiC                   | 0.34       | 0.82                  | 0.28                                  | 61.4                  | 1.9                   | 32.3  |
| 10 vol. % SiC                  | 0.36       | 0.85                  | 0.31                                  | 142.4                 | 1.7                   | 83.8  |

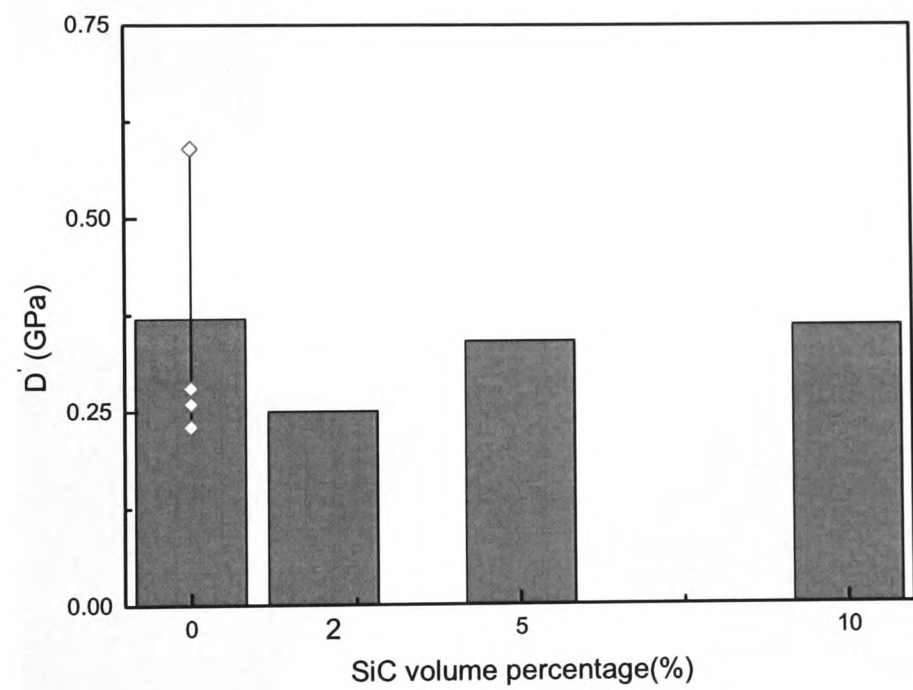
Seen from Table 6.2 and Fig 6.7,  $D'$  and  $d$  of alumina were similar to those of the Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites, considering the accuracy of the fitting parameters and the obtained  $D'$  and  $d$  values for aluminas investigated in Chapter 5 (Table 5.1). The depth of deformation in ground alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites agrees well with the results measured by high-resolution grazing-incidence parallel-beam X-ray powder diffraction ( $\sim 0.5\ \mu\text{m}$  for alumina and  $\sim 0.4\ \mu\text{m}$  for 5 vol.% SiC nanocomposite) [24], but is different to conclusions reported by Wu et al. [25]. Wu et al. [25] used TEM to observe the dislocation depth in 150 grit ground alumina and 5 vol.% SiC nanocomposite with similar grain sizes of about  $3\ \mu\text{m}$ . The depths of dislocations they reported for alumina and nanocomposite were quite different, being  $<1.0\ \mu\text{m}$  and  $3.0\text{-}10\ \mu\text{m}$ , respectively. The TEM-observed dislocation depth for Al<sub>2</sub>O<sub>3</sub> is similar to the findings here but Wu et al.'s dislocation depth of the nanocomposites is much larger than that is predicted from the results of this work. Although the difference could be due to the larger grit diamond wheel and the more aggressive grinding condition used in Wu et al.'s work, it has to be noted that the TEM method is not ideal

for identifying the depth of the residual stresses: the dislocation density was different at different depths underneath the surface with higher densities near the surface. Wu et al. also drew attention to the larger uncertainty of the depth of the dislocations for Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites. It is therefore reasonable to infer that the residual stress is also concentrated nearer the surface than the maximum depth of dislocations as the TEM observation might suggest. Certainly, the single layer dislocation model used in this work is just approximate to the real deformation under the grinding condition.

The two dimensional residual stress distributions on the xy plane and also one dimensional distribution along depth are given in Fig 6.8. The details of the maximum tensile stresses and compressive stresses on the ground regions and pullout regions for alumina and Al<sub>2</sub>O<sub>3</sub>/ x vol. % SiC (x= 2, 5, 10) nanocomposites are listed in Table 6.3. Seen from Fig 6.8, tensile stress components  $\sigma_{xx}$  and  $\sigma_{yy}$  were found only in alumina and 2 vol.% SiC nanocomposite. These would aid crack growth normal to the surface and the formation of further grain pullouts by lateral cracking respectively. In the 5 and 10 vol. % SiC nanocomposites, tensile stresses were not found due to the lack of pullouts and greater compressive stress of  $\sim 4.5$  GPa existed in the stressed layer. In this case the greater stress level was due to the lack of pullouts; although  $D'$  and  $d$  were similar in all materials, fewer pullouts means more plastically deformed ground surface, which was the source of the compressive stresses. The results also suggest that the main difference between ground alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites is the brittle fracture behavior rather than the plastic deformation.



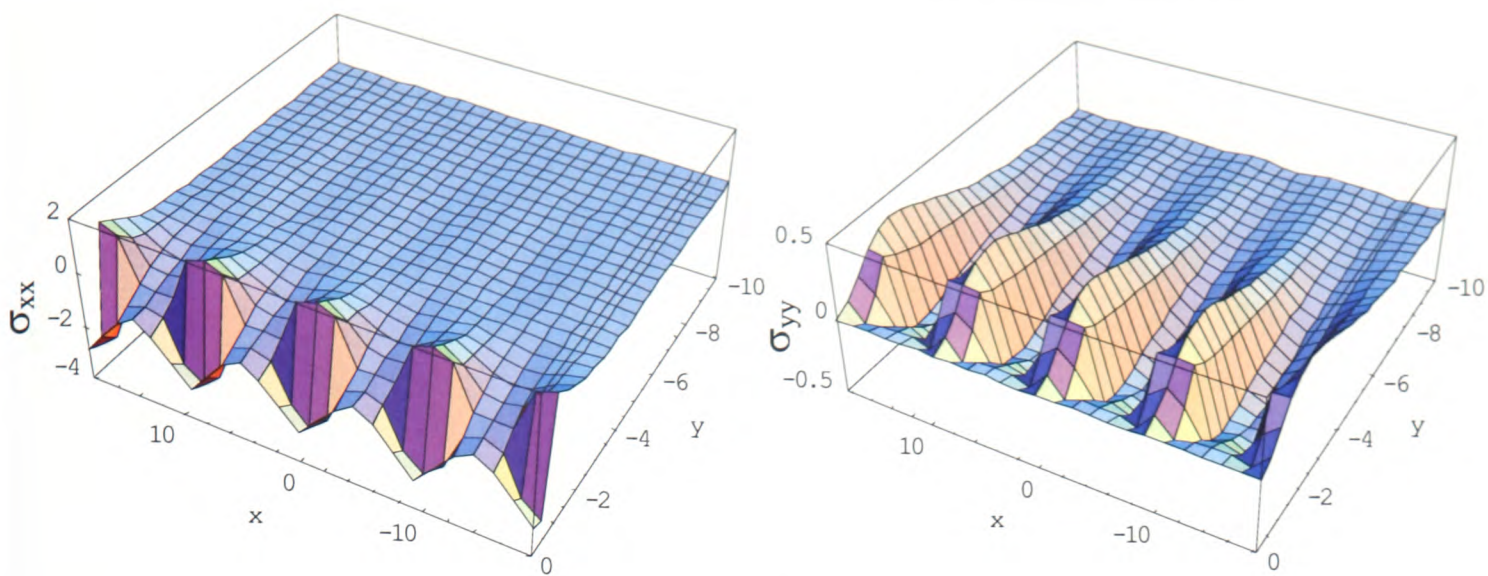
(a)



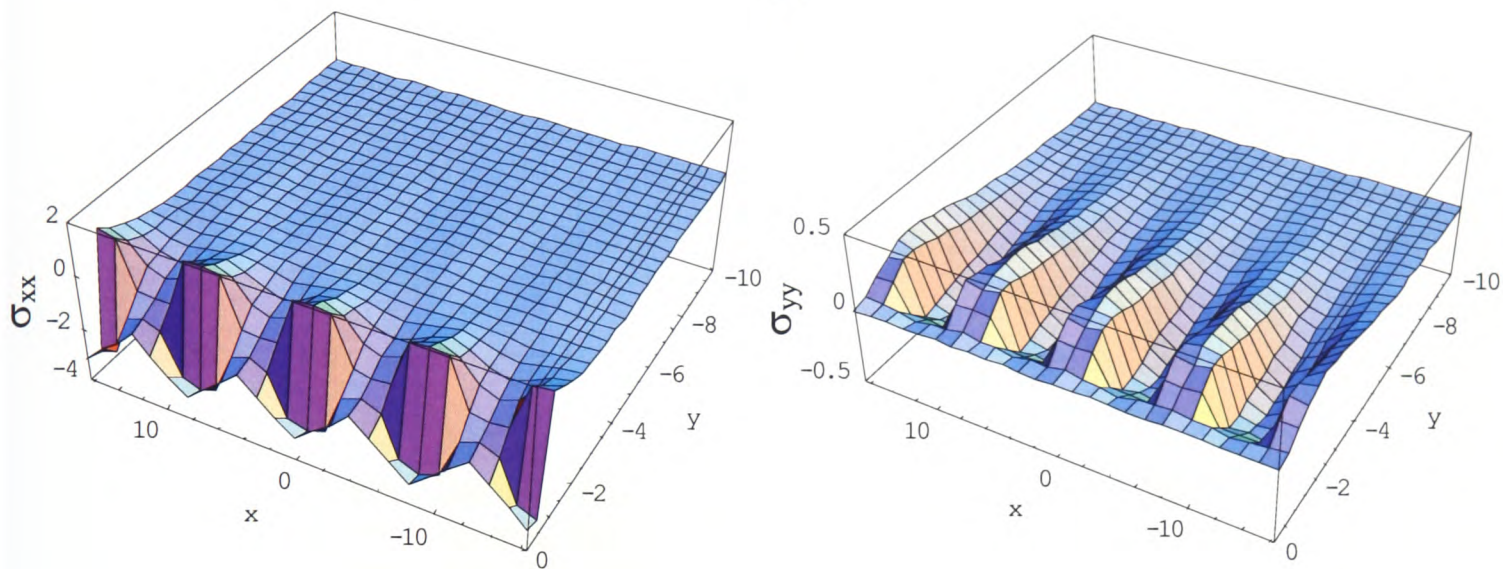
(b)

Fig 6.7 Comparison of (a)  $d$  and (b)  $D'$  of alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites. The extra four  $D'$  and  $d$  values for alumina came from the 45  $\mu\text{m}$  grit ground aluminas investigated in Chapter 5 (see Table 5.1).

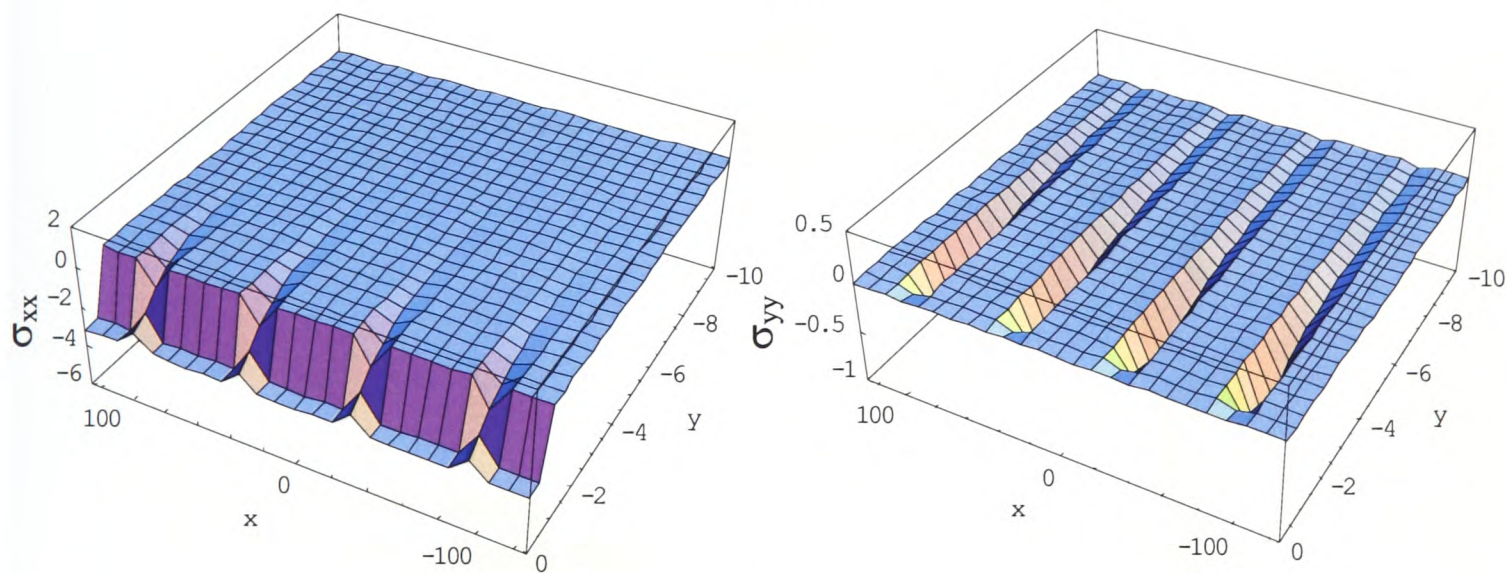




(a)



(b)



(c)



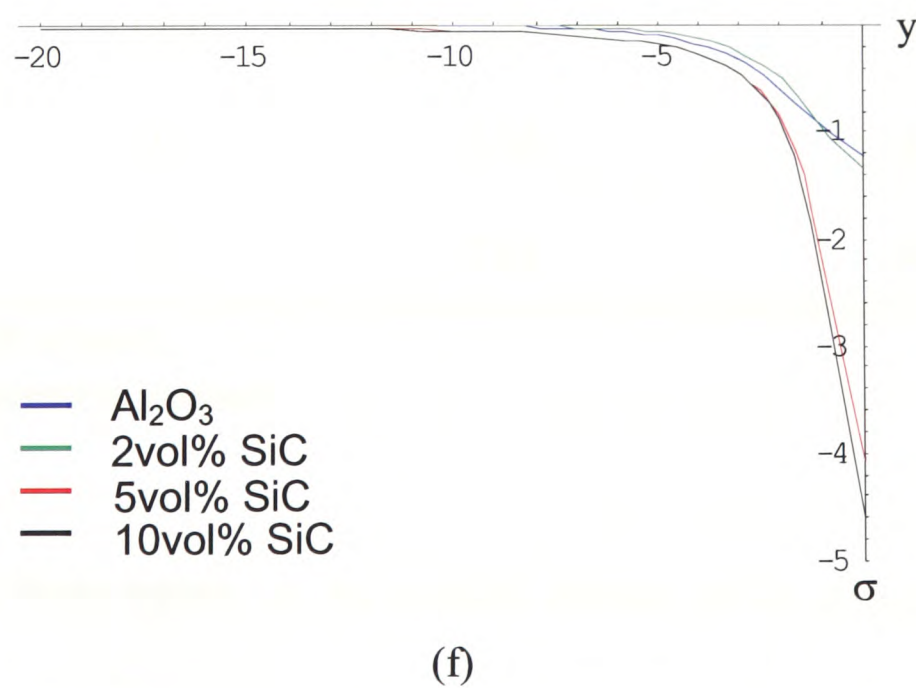
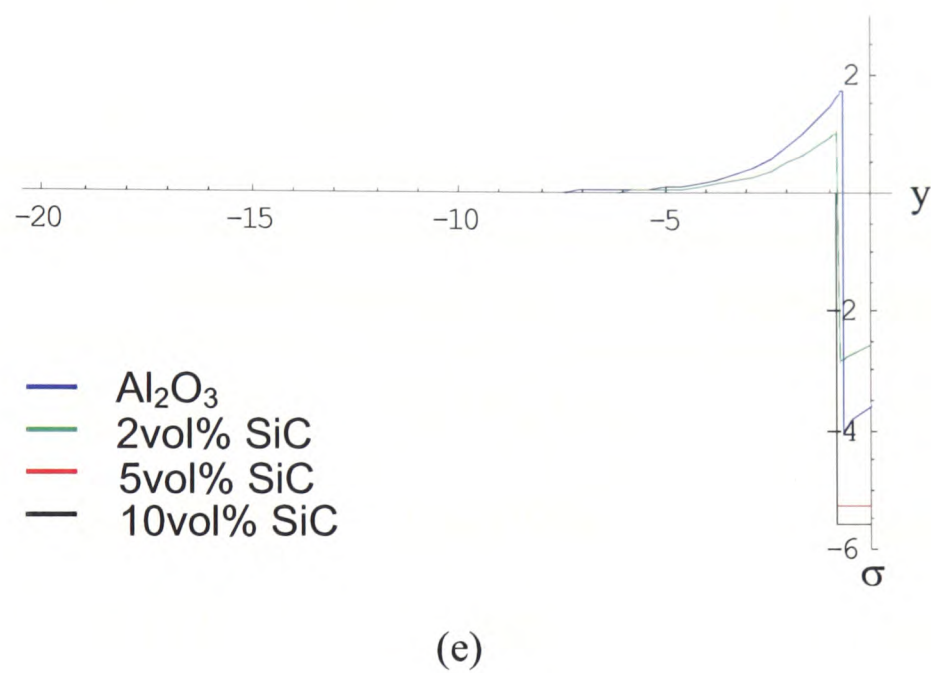
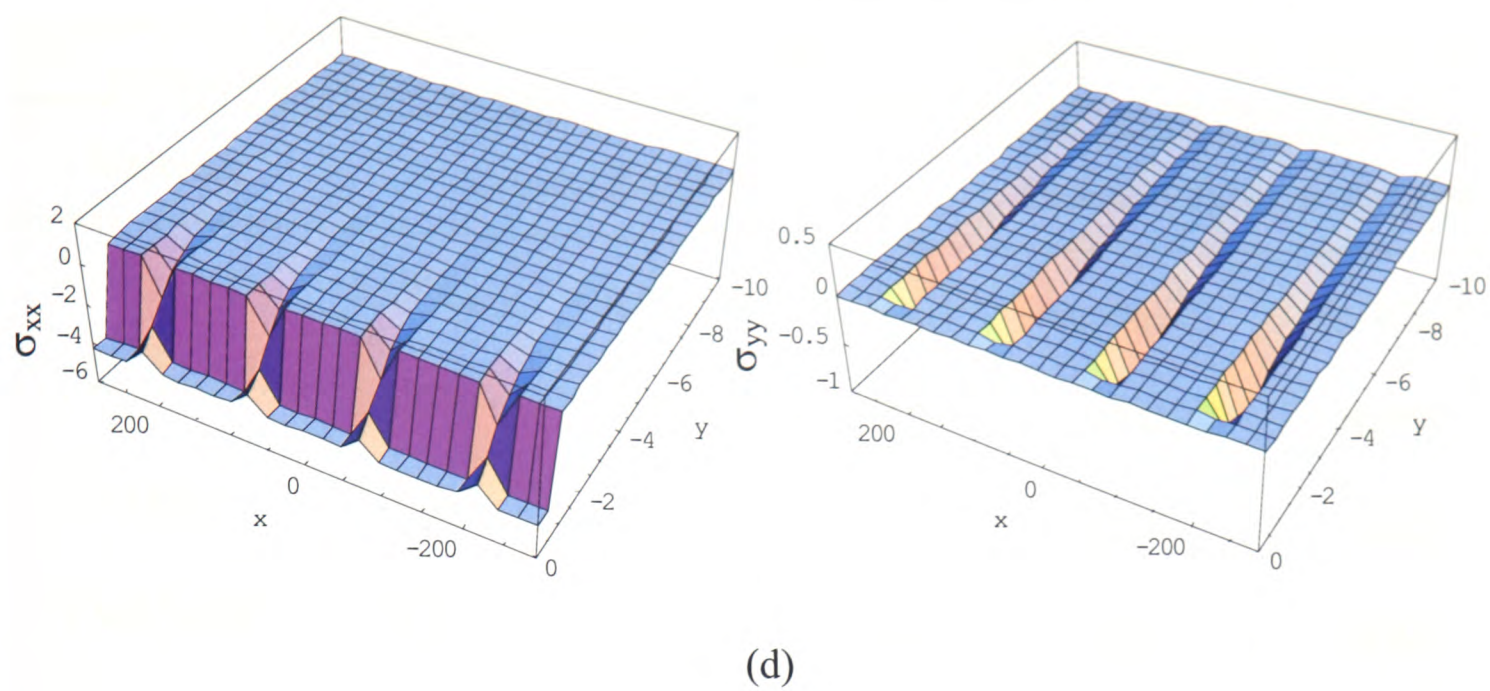


Fig 6.8 Stress distributions over  $xy$  plane for alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites, using  $D'$ ,  $d$ ,  $s$  and  $p$  listed in Table 6.1  
(a) Al<sub>2</sub>O<sub>3</sub>; (b): 2 vol. % SiC; (c): 5 vol. % SiC; (d): 10 vol. % SiC; (e) and (f): total normal stress distribution on the depth direction for ground region and pullout region. Units of stresses are in GPa and  $y$  is in  $\mu\text{m}$ .



Table 6.3 List of maximum tensile and compressive stresses for ground regions and pullout regions for alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites as shown in Fig 6.8

| Materials                      | Ground Region (x=0)       |                           |                         | Pullout Region (x=(s+p)/2) |                         |
|--------------------------------|---------------------------|---------------------------|-------------------------|----------------------------|-------------------------|
|                                | $\sigma_{xx} (^1t_{max})$ | $\sigma_{xx} (^2c_{max})$ | $\sigma_{yy} (t_{max})$ | $\sigma_{xx} (c_{max})$    | $\sigma_{yy} (c_{max})$ |
|                                | (GPa)                     | (GPa)                     | (GPa)                   | (GPa)                      | (GPa)                   |
| Al <sub>2</sub> O <sub>3</sub> | 1.26                      | -3.39                     | 0.34                    | -0.99                      | -0.22                   |
| 2 vol.% SiC                    | 0.75                      | -2.39                     | 0.21                    | -1.09                      | -0.21                   |
| 5 vol.% SiC                    | 0                         | -4.26                     | 0                       | -3.28                      | -0.44                   |
| 10 vol.% SiC                   | 0                         | -4.52                     | 0                       | -3.70                      | -0.47                   |

| Materials                      | Ground Region (x=0)                       |                                           | Pullout Region (x=(s+p)/2)                |
|--------------------------------|-------------------------------------------|-------------------------------------------|-------------------------------------------|
|                                | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ | $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ |
|                                | (t <sub>max</sub> ) (GPa)                 | (c <sub>max</sub> ) (GPa)                 | (c <sub>max</sub> ) (GPa)                 |
| Al <sub>2</sub> O <sub>3</sub> | 1.72                                      | -4.05                                     | -1.23                                     |
| 2 vol.% SiC                    | 1.03                                      | -2.86                                     | -1.35                                     |
| 5 vol.% SiC                    | 0                                         | -5.28                                     | -4.06                                     |
| 10 vol.% SiC                   | 0                                         | -5.61                                     | -4.59                                     |

<sup>1</sup> maximum tensile stresses  
<sup>2</sup> maximum compressive stresses

Compared to previous reports on the residual stresses in the ground alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites [15, 16, 25, 26], the maximum compressive stresses within the stressed layer obtained in this work were significantly higher. The most important difference between the results listed in Table 6.3 and all those reported by

other researchers is the former being maximum local stresses without being volume averaged, and they reflect the real stress distribution existing within the stressed layer.

The difference might also arise from the following aspects: 1) the grinding condition in this work was different to those in literature. The 45  $\mu\text{m}$  diamond grit grinding was much less severe compared to the grinding conditions in the literature. 2) In Reference [16] and [25], the calculations of residual stresses were dependent on the measured thickness of the stressed layer (by curvature method and TEM method, respectively).

The disadvantage of using TEM to determine the depth of the residual stresses has already been discussed; in the curvature method the upper limit of thickness was used and hence the calculated stresses actually defined a lower limit. There was also a significant scatter between sample batches: 63-496 MPa for alumina and 35-438 MPa for the 5 vol.% SiC nanocomposite.

This work shows that  $D'$  and  $d$ , i.e. the damage due to the plastic deformation are similar in alumina and the nanocomposites. The maximum compressive stress is greater in the nanocomposites solely because there are fewer pullouts in them, as the crack initiation is inhibited due to the SiC particles blocking the formation of long twins and dislocation pileups (see section 6.2.1). The mean compressive stress will also be greater in the nanocomposites. Furthermore, the pullouts in the alumina increase the incidence of local tensile stresses, which can lead to further pullouts and weaken the material.

## 6.4 Summary

- 1) Tensile stresses were found on the ground surfaces of alumina and Al<sub>2</sub>O<sub>3</sub>/ 2 vol. % SiC nanocomposite; when SiC addition reached 5 vol.%, the pullout size decreased and so did the pullout fraction, which prevented the arising of the tensile stresses.
- 2) The depths of deformation of alumina and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites were similar (~ 1 μm), which was different to the previous TEM results. The compressive stresses in the 5 and 10 vol.% SiC nanocomposites (maximum stress of about -5.5 GPa) were higher than those in alumina and 2 vol.% SiC nanocomposite.

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## Chapter 7

### Indentation

#### 7.1 Introduction

Vickers indentation is commonly used to characterize the toughness of brittle materials, by creating controlled flaws on the surface and measuring the crack length [1]. The existence of the residual stresses around the indentation and particularly around the cracks induced by indentation, certainly affects the toughness measurement and hence it is very important to understand their effect. The crack system around the indentation has been investigated intensively to understand the indentation process [2-7].  $\text{Cr}^{3+}$  fluorescence microscopy is an ideal technique to measure the residual stresses induced by indentation in alumina ceramics due to its higher spatial resolution and simpler experimental procedure compared to diffraction methods [8-11]. Often a line scan through the indentation center is carried out to study the residual stress distribution [8-11], or an area mapping including the whole indentation impression is utilized to give two-dimensional stress information [8, 9]. However, the spatial resolution used in the previous works was not high enough, and quantitative comparisons between experimental results and Yoffe's model are rarely seen.

In this chapter, the residual stresses around the 1 kg loaded indentations in alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were investigated using the high resolution optical



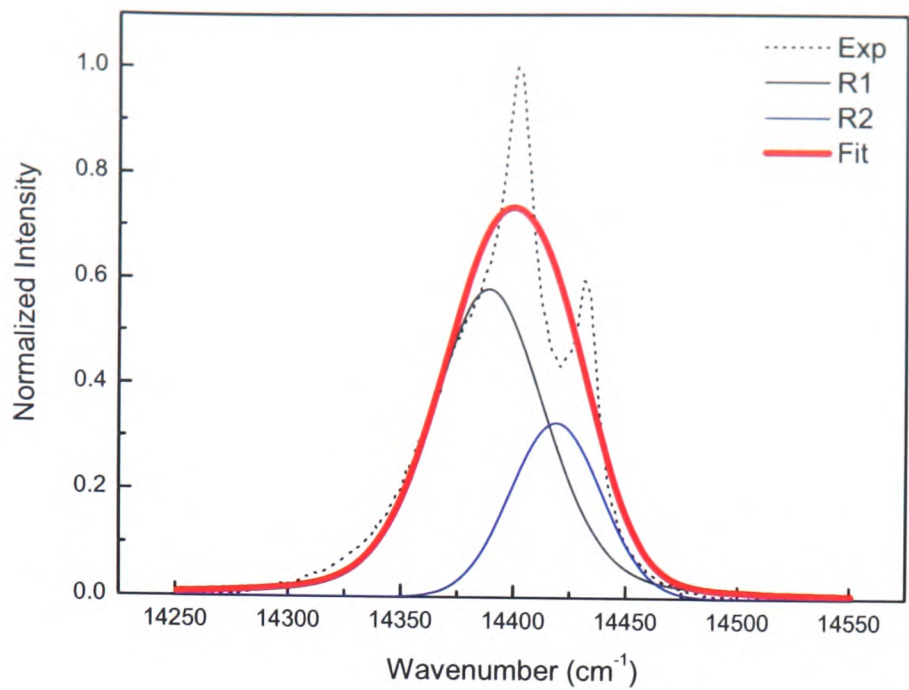
fluorescence technique and the experimental results were compared with Yoffe's model quantitatively. The curve fitting of the spectral lines close to the indentation center were given great care in order to extract important information related to the plastic zone. Line scans and area mapping were carried out in different materials, including sapphire, polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites, to study the cracking behavior and residual stress distributions. For sapphire, its anisotropy made it unsuitable for quantitative comparison to Yoffe's model which is based on the assumption of mechanical isotropy. As a result, mainly qualitative analyses were made on sapphire.

## 7.2 Curve Fitting

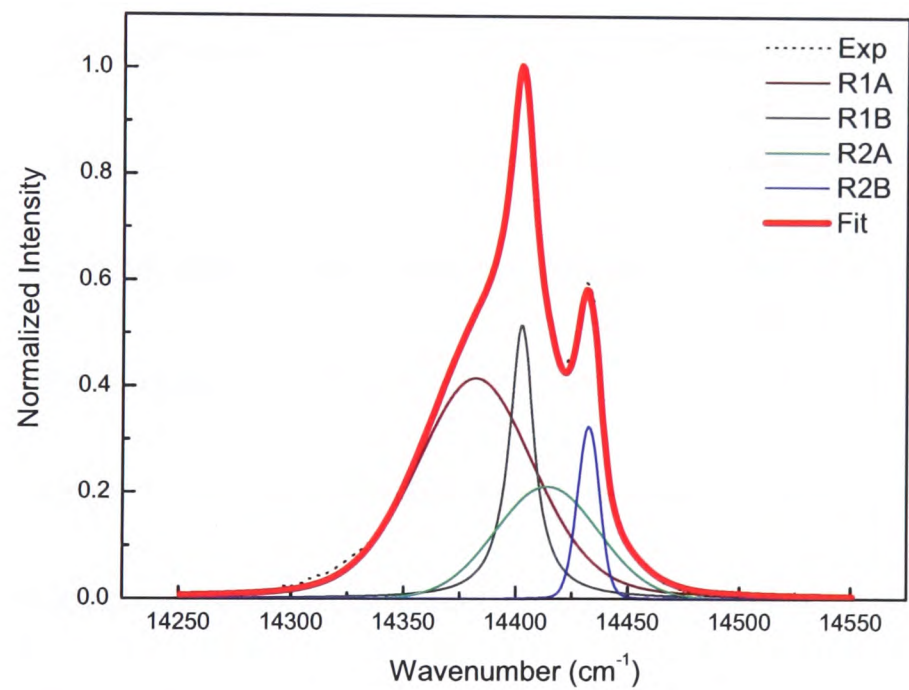
Curve fitting is crucial to the determination of the peak shift of R-lines and hence the level of the residual stresses. The misfit issue for complicated spectra collected from stressed regions was analyzed in Chapter 5 and Chapter 6 where a four-peak fitting procedure could achieve a good fitting for those spectra that could not be fitted using a two-peak fitting. A physical interpretation was also given to the broad and narrow peaks, the two sets of peaks separated by the four-peak fitting, the broad peaks corresponding to the region within a stressed and plastically deformed layer and the narrow peaks corresponding to the region underneath that layer. The superimposition of very different stress states from different regions was responsible for the misfit with two-peak fitting. In the measurement of residual stresses around indentations using the  $\text{Cr}^{3+}$  fluorescence microscopy technique, spectra that could not be fitted well

using two-peak fitting were also found and the fitting could be improved by using the four-peak fitting (see Fig 7.1). The broad peaks and narrow peaks can be physically related to the two distinct regions well known for indentations: the plastically deformed zone and regions outside this zone where the deformation, if any, is elastic. The fact that the spectra requiring four-peak fitting were collected from regions close to the indentations also supports the above idea from the consideration of the sampling volume, as shown in Fig 7.2: the sampled regions include volumes both inside and outside the plastic zone where the stress states are quite different.

In Chapter 5, the R1/R2 area ratio was used as a criterion to determine when to use two-peak fitting or four-peak fitting. This was applied to the curve fitting for the indentation experiments as well. Fig 7.3 shows the peak position distribution along a line scan through the indentation center on a sapphire specimen using two-peak fitting. Within a distance of 12  $\mu\text{m}$  from the indentation center, the apparent R1 and R2 peak shifts were quite different. The R1/R2 area ratios for the spectra collected when  $r \leq 12$   $\mu\text{m}$  ( $r$  is the distance from the indentation center) were found to be much bigger than the fairly constant value when  $r > 12$   $\mu\text{m}$ . The spectra with abnormal R1/R2 area ratios were then treated using four-peak fitting (for line scan only). The reasoning behind this and the detailed fitting procedure have been described in Chapter 3.



(a)



(b)

Fig 7.1 Examples of the misfit for a spectrum collected close to the indentation using a two-peak fitting (a) and the improvement of the curve fit by using a four peak fitting (b). Symbols A and B in (b) indicate the broad peaks and narrow peaks, which correspond to the two regions shown in Fig 7.2 using the same symbols.

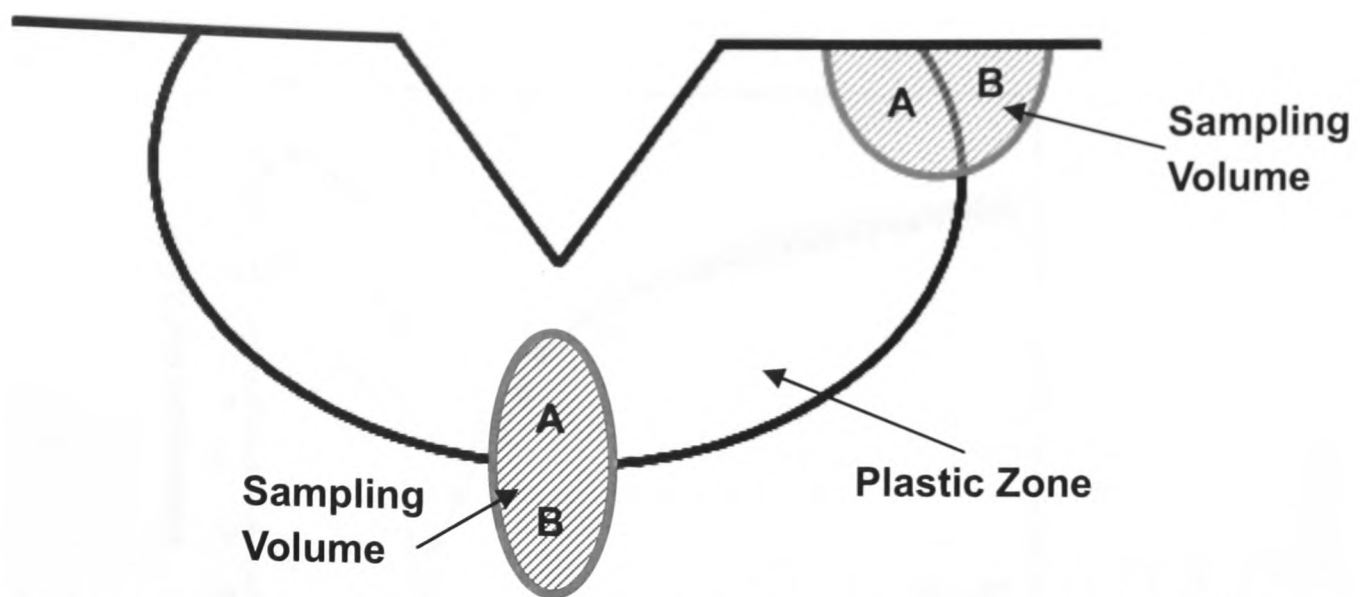
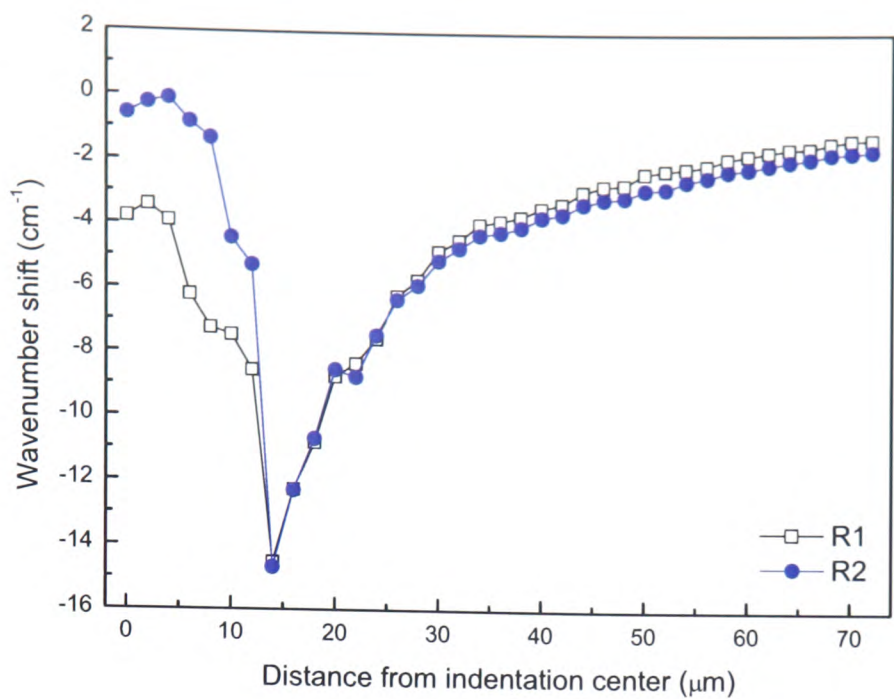
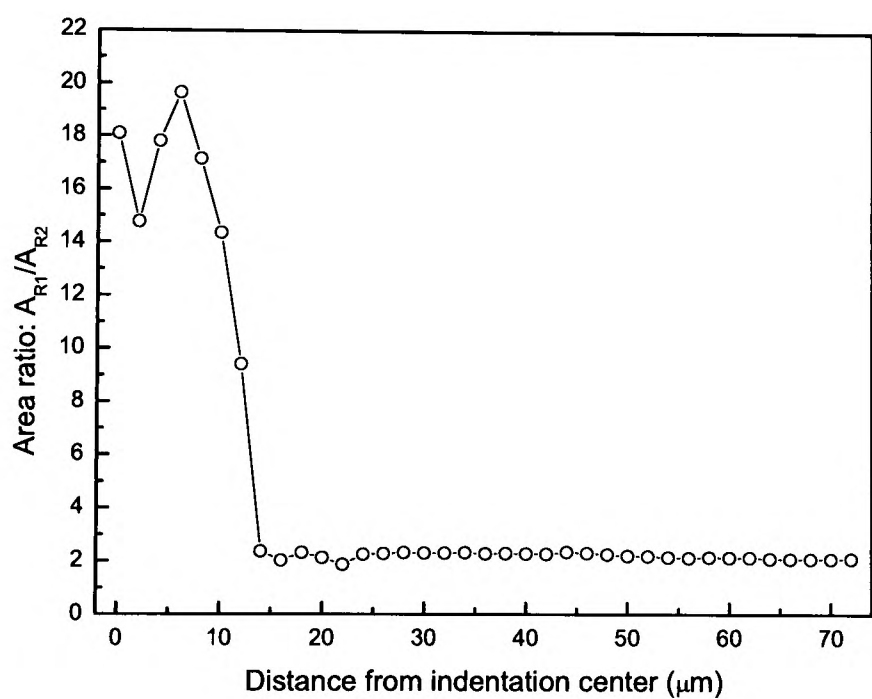


Fig 7.2 Schematic of the sampling volume close to the plastic zone, where both regions inside the plastic zone (represented by symbol A) and outside it (represented by symbol B) are sampled

The assumed existence of distinct broad peaks and narrow peaks means that the sampled volume includes part of the plastic zone. Stress information on the plastic zone is reflected by the broad peaks. In the following experimental results section, for spectra where four-peak fitting was used to determine the peak positions, only the peak position of the broad peak (R1) will be used in the peak shift distribution plot. The distance at some point to the indentation center beyond which the two-peak fitting can fit the spectrum well is an effective indication of the size the plastic zone. In principle, the depth of the plastic zone can be estimated similarly from the depth scan.



(a)



(b)

Fig 7.3 Wavenumber shifts of R1 and R1 (a) and R1/R2 area ratio (b) for spectra collected from a line scan through the indentation center on a sapphire specimen using the two-peak fitting

### 7.3 Experimental Results

Line scans, depth scans and area mapping were carried out on 1 kg Vickers indentations on 1  $\mu\text{m}$  diamond grit polished single crystal sapphire, polycrystalline  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites (the same materials as used for the grinding stress investigation in Chapter 6). For line scans the laser was first focused on the polished surface away from the indentation and the scan then started from the indentation center (without refocusing) and in the direction normal to the edge of the indentation. A step size of 2  $\mu\text{m}$  was used and the laser detection time was 20 seconds per point for all the materials. Note that due to the sloping surface inside the indentation impression, the laser was actually focused slightly above the real surface when the laser was scanning within the indentation impression. However, for the 1 kg loaded specimen this has a small effect considering the shallow depth of the indentation (maximum depth of  $\sim 4 \mu\text{m}$ ), and also the difficulty of focusing precisely on the sloping surface. An area of  $240 \times 200 \mu\text{m}^2$  was used to map the stresses around the indentations for all the materials, and the step size was  $4 \times 4 \mu\text{m}$  with a collection time of 20 seconds per point. The origin point ( $x=0, y=0$ ) was set to be the indentation center, but on the depth of that of the polished surface. Schematic representation of line scan and area mapping are shown in Fig 7.4. A volume scan was also carried out on the sapphire specimen to examine the stress distribution on a three-dimensional scale ( $160 \times 140 \times 70 \mu\text{m}^3$ , with step size  $5 \times 5 \times 4 \mu\text{m}$  and 20 seconds of collection time). The origin point ( $x=0, y=0, z=0$ ) was set as the same position as for the area mapping. The same indents were used for line scans, depth scans, area mapping and volume



mapping.

To investigate the probe effect on the measured R-line peak shift, non-confocal microscope settings (slit size = 50  $\mu\text{m}$ , 100 $\times$  objective lens) were used (hence worse resolution, see section 3.5.2 in Chapter 3) on  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites. For comparison, the line scans were made on the specimens with the same direction on the same indentation. Depth scans were made through three different positions with distance of  $r'$   $\mu\text{m}$  to the indentation center (they were along the direction of the line scans, as shown in Fig 7.4 (a) and (b)) on  $\text{Al}_2\text{O}_3$  and 10 vol.% SiC nanocomposite specimens using both confocal and non-confocal modes. The zero  $z$  position ( $z$  is the distance away from the focal plane) was set to be that of the polished surface surrounding the indentation, as for the line scan. A step size of 2  $\mu\text{m}$  was used and the collection time was 60 seconds per point. The depth scanning aimed to quantitatively examine the PRF effect and the validity of Yoffe's model by using the convolution method similar to that used in Chapters 5 and 6.

The R1 peak position was used to characterize the peak shift unless stated otherwise. The reference positions for were determined from the average stress-free R1 positions (spectra collected on 20 random points far away from the indentation on the individual polished surface of specimens).

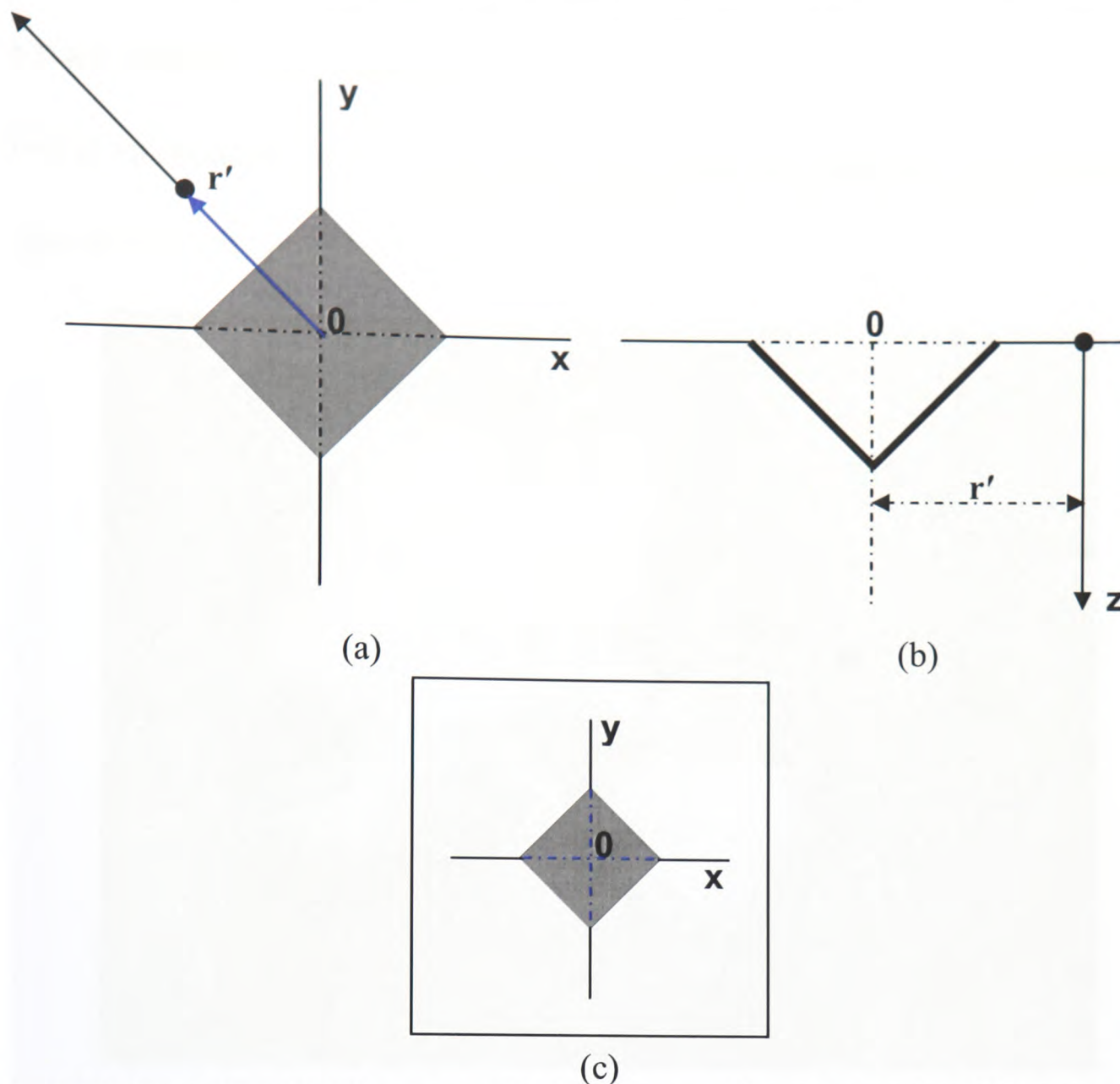


Fig 7.4 Schematic of (a) line scan; (b) depth scan; (c) area mapping. The arrows in (a) and (b) indicate the scanning direction.

### 7.3.1 Sapphire

An optical micrograph of the indentation on the sapphire is given in Fig 7.5. The diagonal length of the indentation print was  $28\text{ }\mu\text{m}$ , corresponding to a Vickers hardness of 24 GPa. Radial cracks could be clearly seen propagating from the corners of the indentation. Hazy features around the indentation but below the surface were also very obvious. These were believed to be subsurface lateral cracks, visible due to the transparency of the sapphire. The lateral cracks showed clear anisotropic characteristics, which was further proved from the area mapping in Fig 7.7. Twin like features were also observed, as indicated by the parallel arrows in Fig 7.5. Twinning is

the main deformation mechanism in sapphire and evidence of twinning is generally found at indentations on non-basal planes (the surface orientation of the sapphire used in this work is (012)) [13-15].

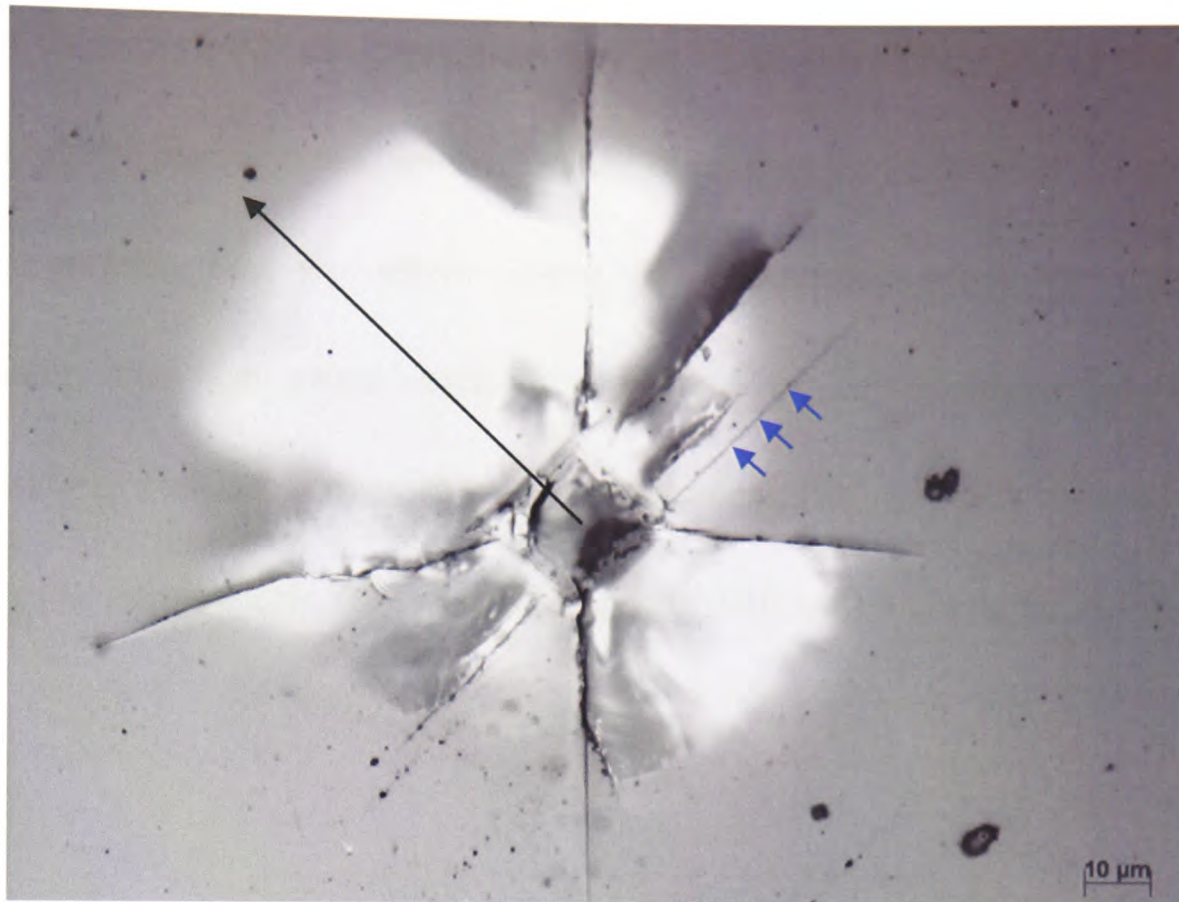


Fig 7.5 Optical micrograph of a 1 kg loaded indentation on sapphire. The arrow across the indentation center indicates the direction of the line scan.

#### 7.3.1.1 Line Scan

The peak position distribution along the line indicated by the arrow shown in Fig 7.5 is given in Fig 7.6. Note that the line scan crosses over a subsurface lateral crack. It will be shown later that the lateral cracks would not affect the line scans on the surface significantly. The peak shifts were all negative along the line suggesting net compressive residual stresses. The maximum compressive stress occurred at 6  $\mu\text{m}$  away from the indentation center and became less compressive at longer distances. The edge of the indentation print was roughly at 10  $\mu\text{m}$  from the indentation center ( $\sqrt{2}d/4 \approx 10 \mu\text{m}$ ,  $d$  is the diagonal length of the indentation print), so the most

compressive position was within the indentation print. Note that the exact position where the most compressive stress existed and the precise level of this maximum stress were influenced significantly by the curve fitting details of the spectra. As described in section 7.2, the peak positions for  $r \leq 12 \mu\text{m}$  shown in Fig 7.6 were the positions of the broad peaks, which were mathematically ill-defined. However, it is safe to conclude that the stress inside the indentation print was much more compressive than the stress outside it, instead of being similar or even less compressive as indicated incorrectly by the two-peak fitting results shown in Fig 7.3 (a). The peak shifts within the indentation print ( $r < 10 \mu\text{m}$ ) were in the range of -23 to  $-18 \text{ cm}^{-1}$ .

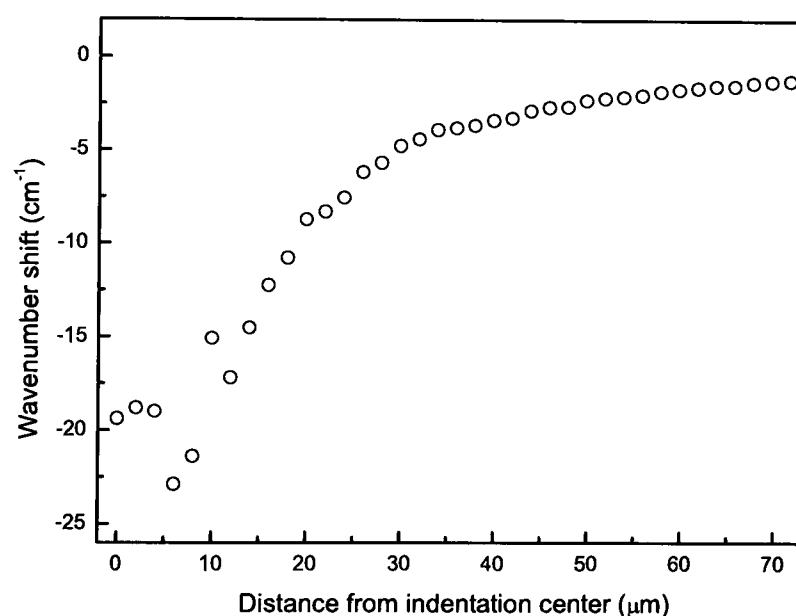


Fig 7.6 Peak position distribution along a line through the indentation center on sapphire

### 7.3.1.2 Area and Volume Mapping

Line scanning is a quick way to detect the residual stress distribution but the



information obtained is limited to the set line. An area mapping can provide more information with a single test, as shown in Fig 7.7. Although the peak positions were all fitted using two-peak fitting only, meaning that the peak positions close to the indentation print were not very accurate, some important information could still be extracted: compressive stresses were found inside the indentation print, which was in agreement with the line scan results; tensile stresses were found on the tips of radial cracks and circling around the indentation, and at the tips of lateral cracks (see Fig 7.8 also). Area mapping provided direct evidence of tensile stresses and connected them with the appearance of the indented regions in the optical microscope.

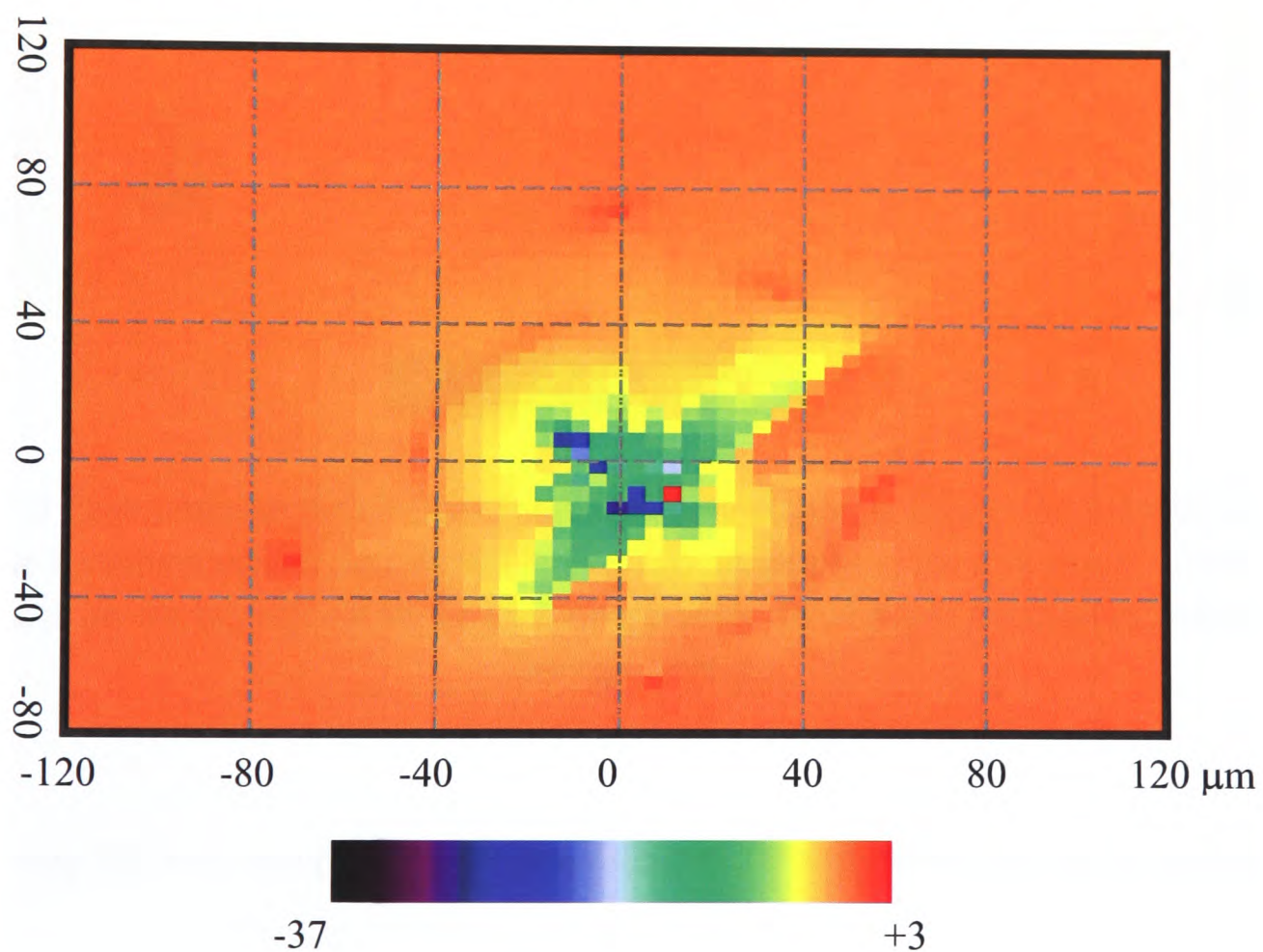


Fig 7.7 Peak position distribution from an area mapping around the indentation (the same specimen orientation shown in Fig 7.5) on sapphire, the unit is  $\text{cm}^{-1}$  shift.

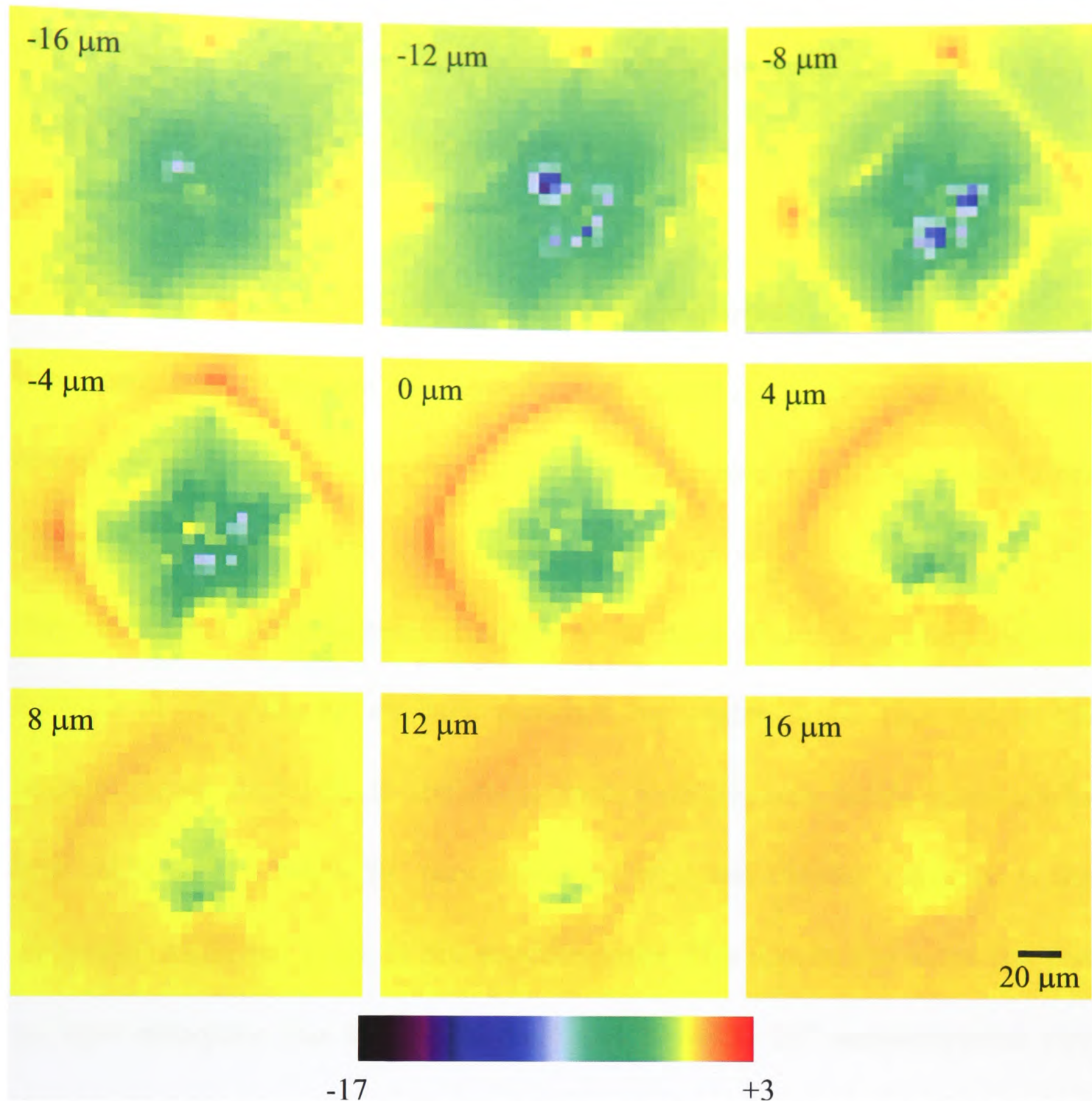


Fig 7.8 Peak position distribution from a volume mapping around the indentation (the same specimen orientation shown in Fig 7.5) on sapphire, the unit is  $\text{cm}^{-1}$  shift. The number on the top left corner of each image is the focal depth position ( $h$ ) relative to the top surface.

By doing the area mapping on different depths, three dimensional stress information could be obtained, as shown in Fig 7.8. The stress development information at different depths was reflected but limited by the resolution (the resolution gets worse with depth). Seen from Fig 7.8, the lateral cracks went all the way round the indentation linking the radial crack tips, which was not observed in the optical micrograph (Fig 7.5). It showed that the optical micrographs might be deceptive as



only cracks with more than a certain opening could be observed. The depth of lateral cracks was  $<16\text{ }\mu\text{m}$  as far below the surface they could not be detected.

### 7.3.2 Polycrystalline $\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiC}$ Nanocomposites

Line scans, depth scans and area mapping were carried out on polycrystalline  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/x\text{ vol.}\%\text{ SiC}$  nanocomposite ( $x=2, 5, 10$ ) specimens with similar grain sizes. Optical micrographs of the 1 kg loaded indentations are shown in Fig 7.9. The diagonal lengths of the indentation print were all about  $30\text{ }\mu\text{m}$ , suggesting a similar hardness of  $\sim 21\text{ GPa}$  for all these materials, but slightly lower than that of the sapphire crystal. Radial cracks and lateral cracks were also seen, but the lateral cracks in 5 vol.% and 10 vol.% SiC nanocomposites were less obvious compared to the monolithic  $\text{Al}_2\text{O}_3$  and 2 vol. % SiC nanocomposite. This may be due to the fact that the light absorption was stronger in the 5 and 10 vol.% SiC nanocomposites (see Chapter 4), although cracking is known to be suppressed in these materials as well [10]. Note that the indenter contained a small flaw as seen in the left corner of each indentation. The area mapping shown later in Fig 7.14 demonstrates that this did not affect the stresses measured significantly.

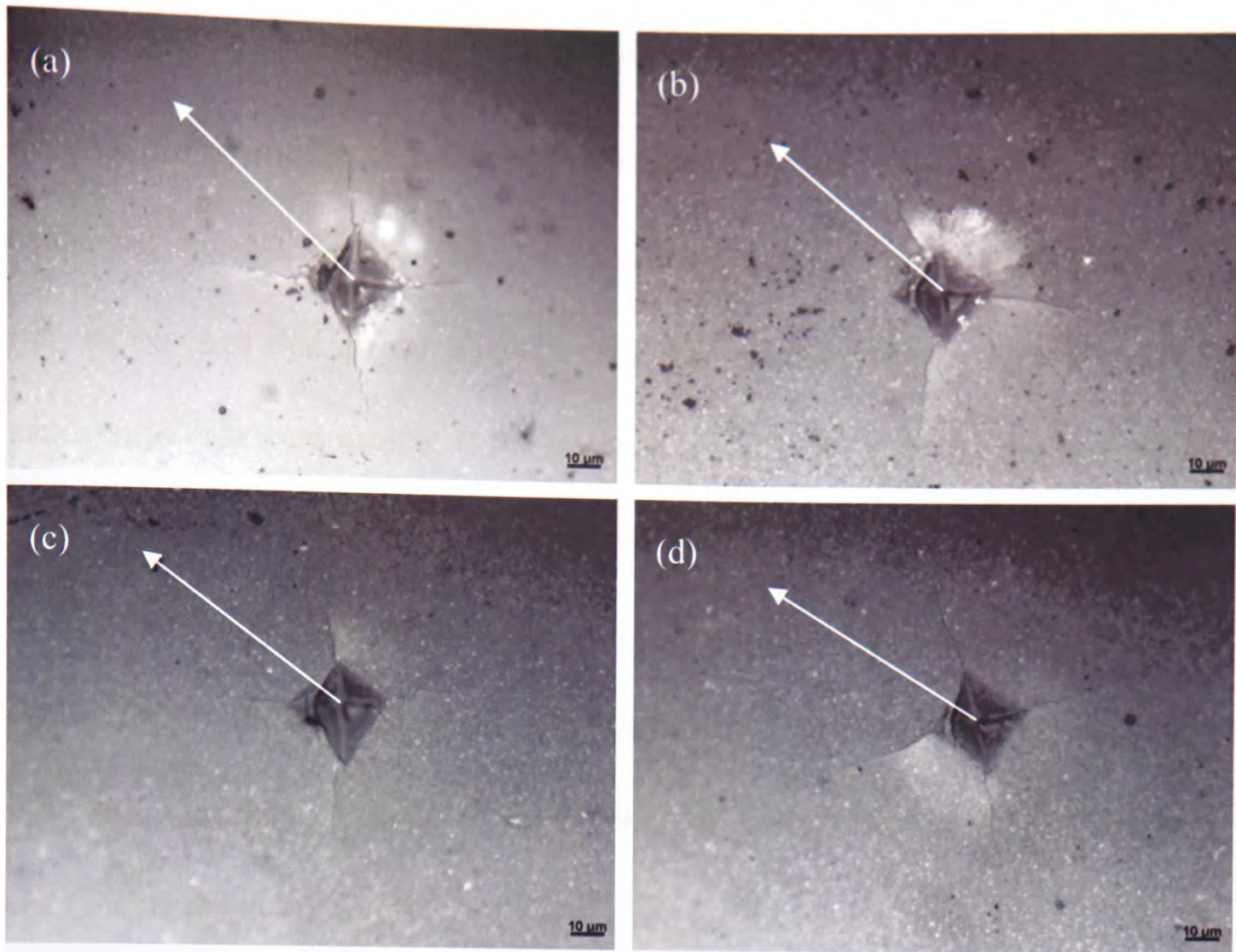


Fig 7.9 Optical micrographs of a 1 kg loaded indentation on (a)  $\text{Al}_2\text{O}_3$ ; (b) 2 vol.% SiC; (c) 5 vol.% (d) 10 vol.% SiC. The arrows indicate the direction of the line scan.

### 7.3.2.1 Line Scan

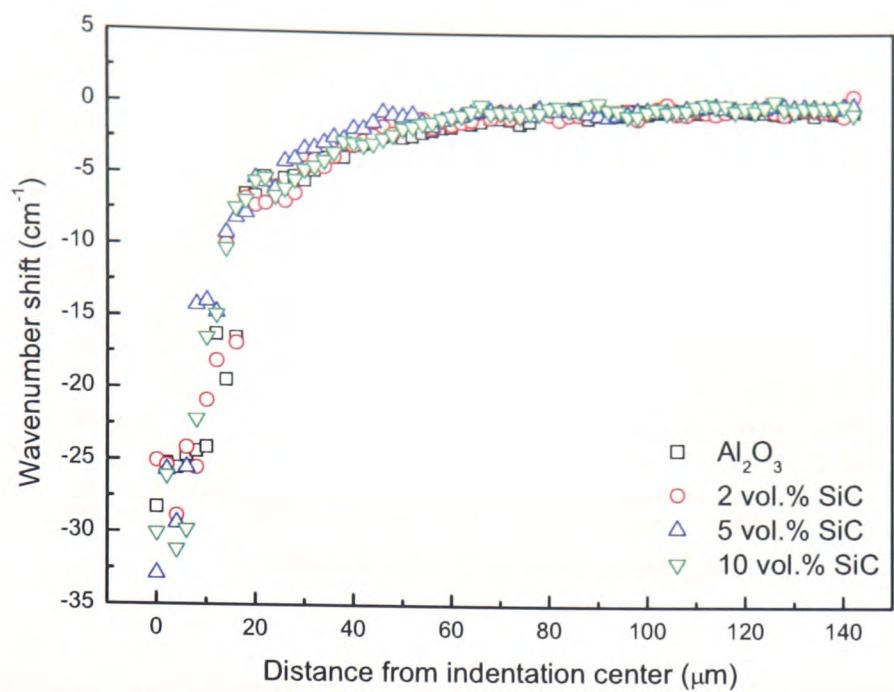
The peak positions along the lines through the indentation center in  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites indicated by the arrows in Fig 7.9 are given in Fig 7.10 (a). Very similar wavenumber shifts were found on these four materials: the most compressive stress was close to the indentation center ( $r=0 \mu\text{m}$  for  $\text{Al}_2\text{O}_3$  and 5 vol.% SiC nanocomposite;  $r=4 \mu\text{m}$  for 2 and 10 vol.% SiC nanocomposites) and became less compressive with the increasing distance from it, finally closing to zero stress at a distance of  $\sim 70 \mu\text{m}$ . The maximum peak shift in alumina was about  $-28 \text{ cm}^{-1}$  shift, larger than that in the sapphire ( $-23 \sim -18 \text{ cm}^{-1}$  shift). This is in agreement with the prediction by Yoffe as the stresses are proportional to  $(P/H)^{3/2}$ ,  $P$  and  $H$  being load

and hardness respectively (see section 7.4.1). Larger stresses are expected in the softer alumina. From the R1/R2 area ratio in Fig 7.10(b), the distance to the indentation center beyond which a two-peak fitting can fit the spectra well for alumina was  $\sim 20$   $\mu\text{m}$ , longer than that of sapphire (12  $\mu\text{m}$ ). The maximum peak shifts in  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were similar, in the range of  $-33 \sim -29$   $\text{cm}^{-1}$  shift. The critical distances to the indentation center beyond which the two-peak fitting can be used were in the range of 14-20  $\mu\text{m}$ . In a previous work [11], Limpichaipanit showed that the line scan results from different indents on 1 kg loaded alumina were quite reproducible: the peak shifts (at same distance to the indentation center) showed a small scatter ( $< 1$   $\text{cm}^{-1}$  shift) outside the plastic zone.

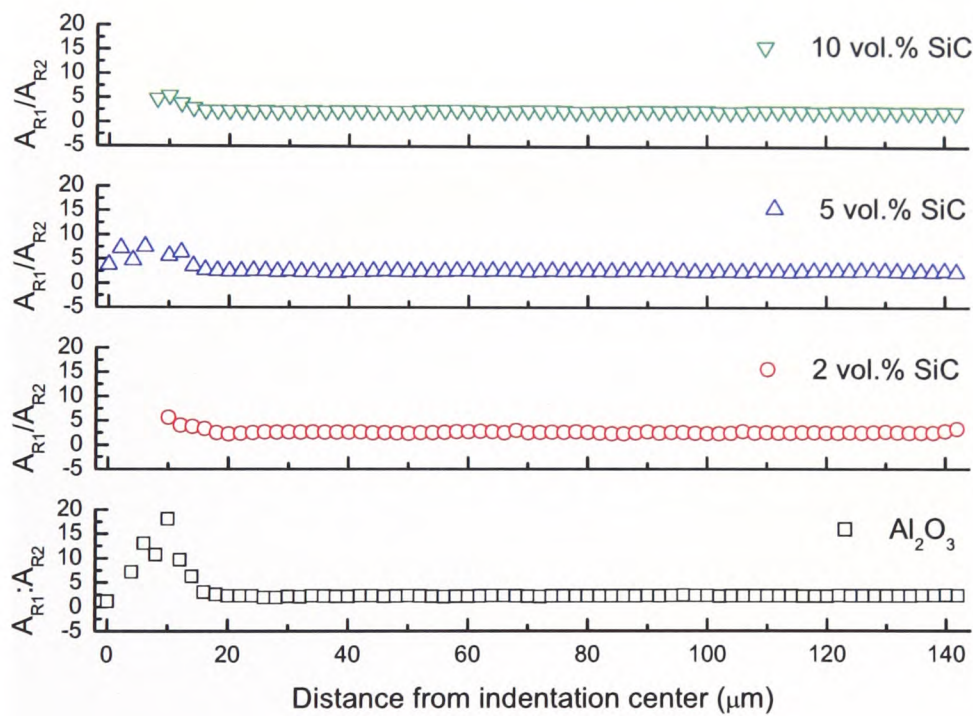
The same line scans were carried out under non-focal microscope settings and the results are given in Fig 7.11. Compared to Fig 7.10 (a), the maximum peak shift of  $\text{Al}_2\text{O}_3$  decreased to about  $-21$   $\text{cm}^{-1}$  shift; the maximum peak shifts for  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were about  $-32 \sim -27$   $\text{cm}^{-1}$  shift, not significantly different to the results obtained under the confocal mode. The difference made on the peak shift results between  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites due to confocal and non-confocal microscope settings was further proved in Fig 7.12: for  $\text{Al}_2\text{O}_3$ , along the distance to the indentation center the measured peak shifts using non-confocal mode were smaller than those measured using confocal mode; this effect was much less significant in  $\text{Al}_2\text{O}_3/10$  vol. % SiC nanocomposite. In Fig 7.12 (b), the comparison was made outside the plastic zone ( $r > 20$   $\mu\text{m}$ ) so as to avoid the confusion that the



difference might come from the curve fitting.



(a)



(b)

Fig 7.10 Peak position distribution (a) and R1/R2 area ratio (b) along a line through the indentation center on Al<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>/ x vol.% SiC nanocomposite (x=2, 5,10). For 2 and 10 vol.% SiC nanocomposite, the area ratios for spectra with r<10 μm were larger than 20 and hence could no been seen on the plots.

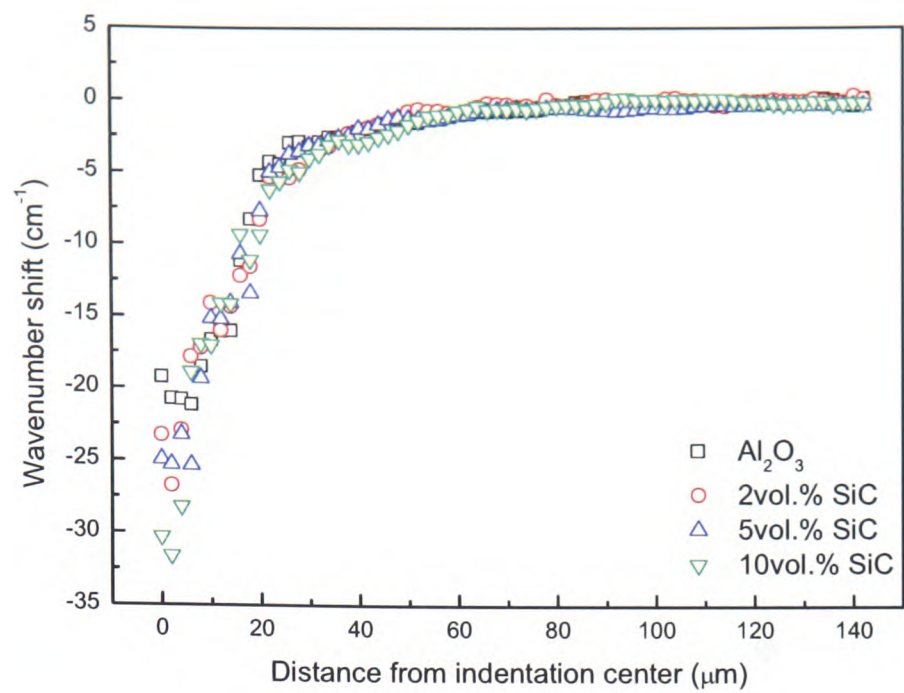
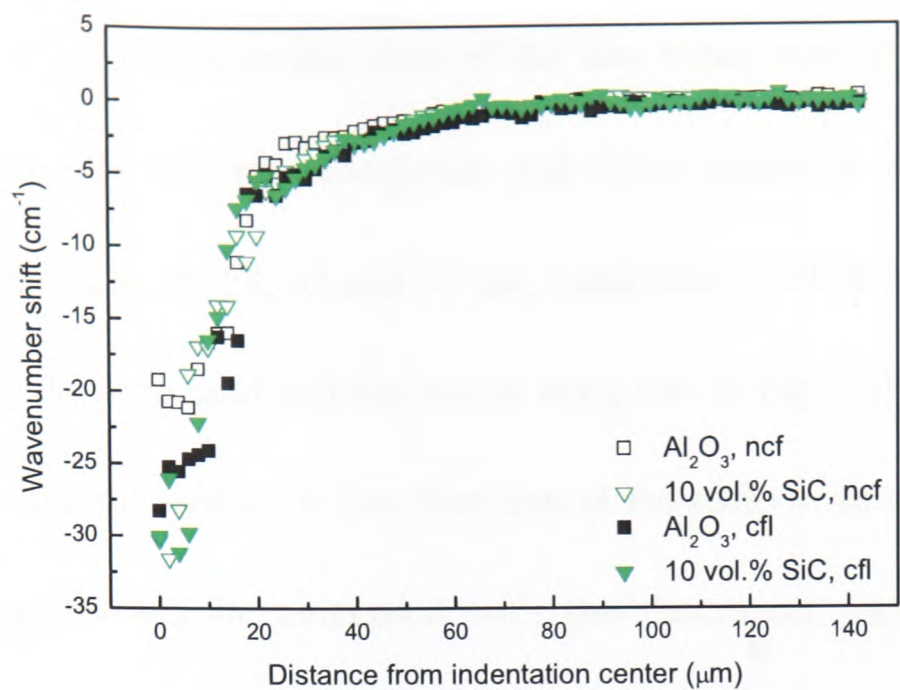
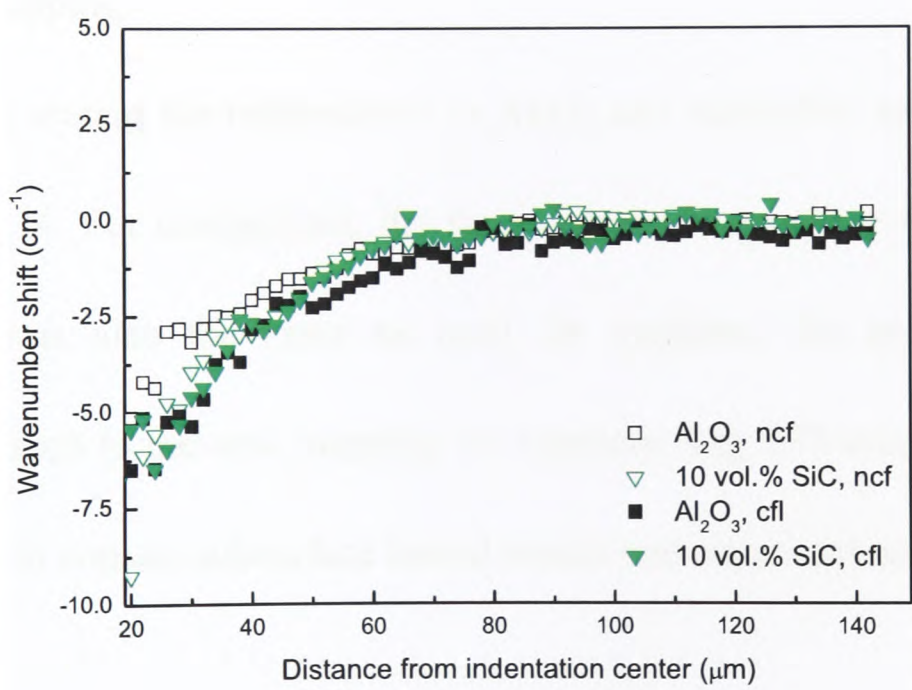


Fig 7.11 Peak position distribution along a line through the indentation center on Al<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>/ x vol.% SiC nanocomposite (x=2, 5,10), using non-confocal microscope settings



(a)



(b)

Fig 7.12 (a) Comparison of peak shift distribution along a line scan through the indentation center on Al<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>/ 10 vol.% SiC nanocomposite specimens, using non-confocal (denoted by *ncf*) and confocal mode (denoted by *cfl*), respectively. In (b), the X scale was expanded to start from outside the plastic zone.

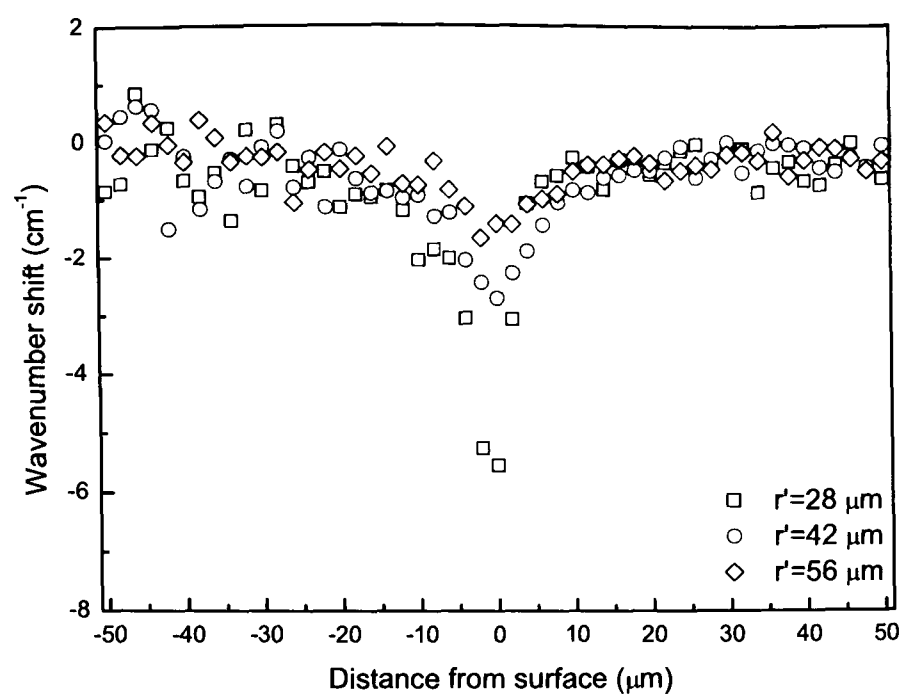


### 7.3.2.2 Depth Scan

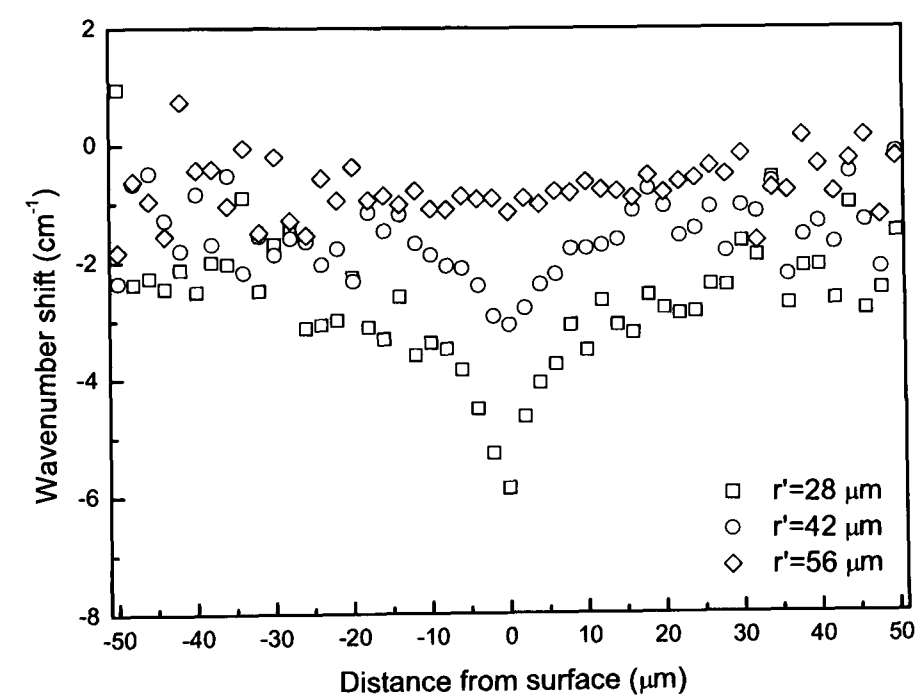
Depth scans through points on the route of the line scans were also made on  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/10$  vol. % SiC nanocomposite and these positions were chosen to be outside the plastic zone ( $r'=28, 42$  and  $56 \mu\text{m}$ , respectively). Both confocal mode and non-confocal mode were used and the results are given in Fig 7.13. The results show that the shift below the surface is less than that at the surface. In section 7.4.2, these experimental results will be compared with the theoretical predictions based on Yoffe's model, taking the PRF into account.

### 7.3.2.3 Area Mapping

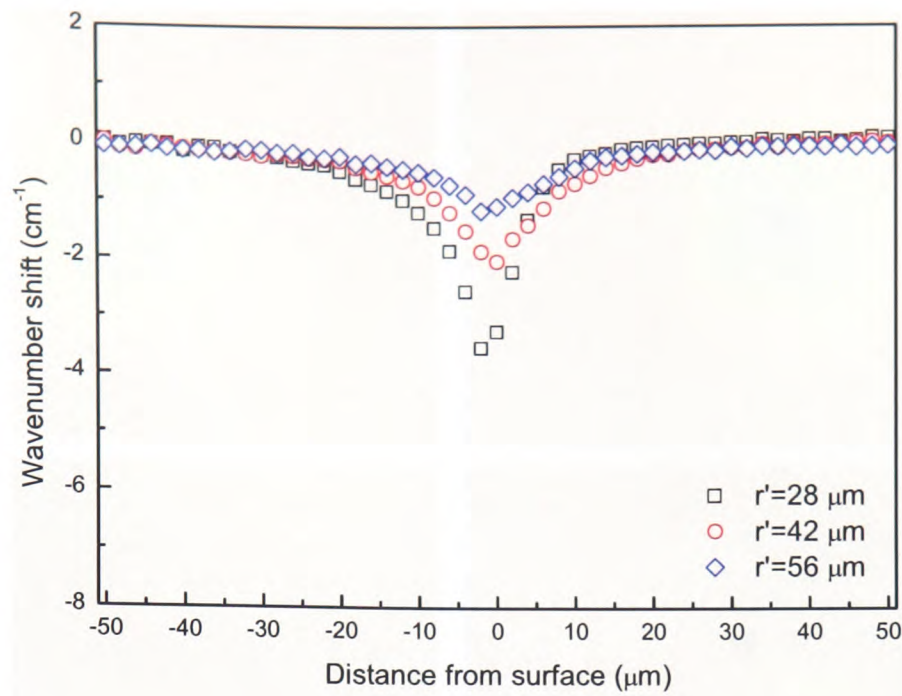
Area mappings around the indentations in  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites are given in Fig 7.14. For comparison, the same false color gradient for peak shift was used, and it was also the same as used for sapphire. No asymmetrical stress distribution as seen in the area mapping for sapphire (Fig 7.7) was found even when some appeared to contain subsurface lateral cracks and others did not.



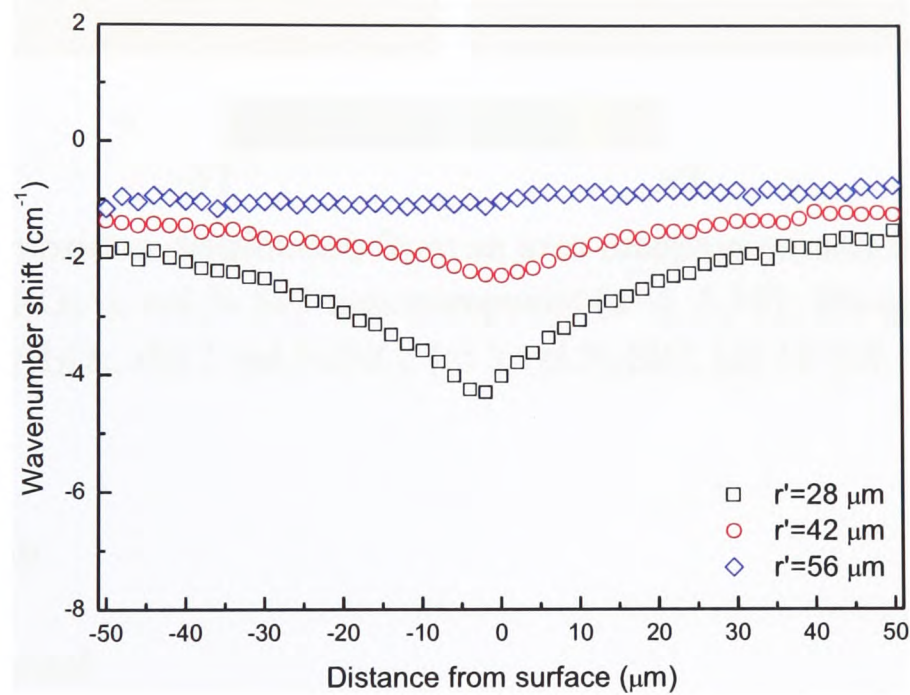
(a)



(b)



(c)



(d)

Fig 7.13 Peak shifts distribution on the depth direction, through three positions (with distance to the indentation center of  $r'$ , as defined in Fig 7.4 (a) and (b)) on the surface of  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/10\ \text{vol.}\% \text{SiC}$  nanocomposite specimen.  
confocal mode: (a)  $\text{Al}_2\text{O}_3$ ; (b) 10 vol.% SiC;  
non-confocal mode: (c)  $\text{Al}_2\text{O}_3$ ; (d) 10 vol.% SiC.

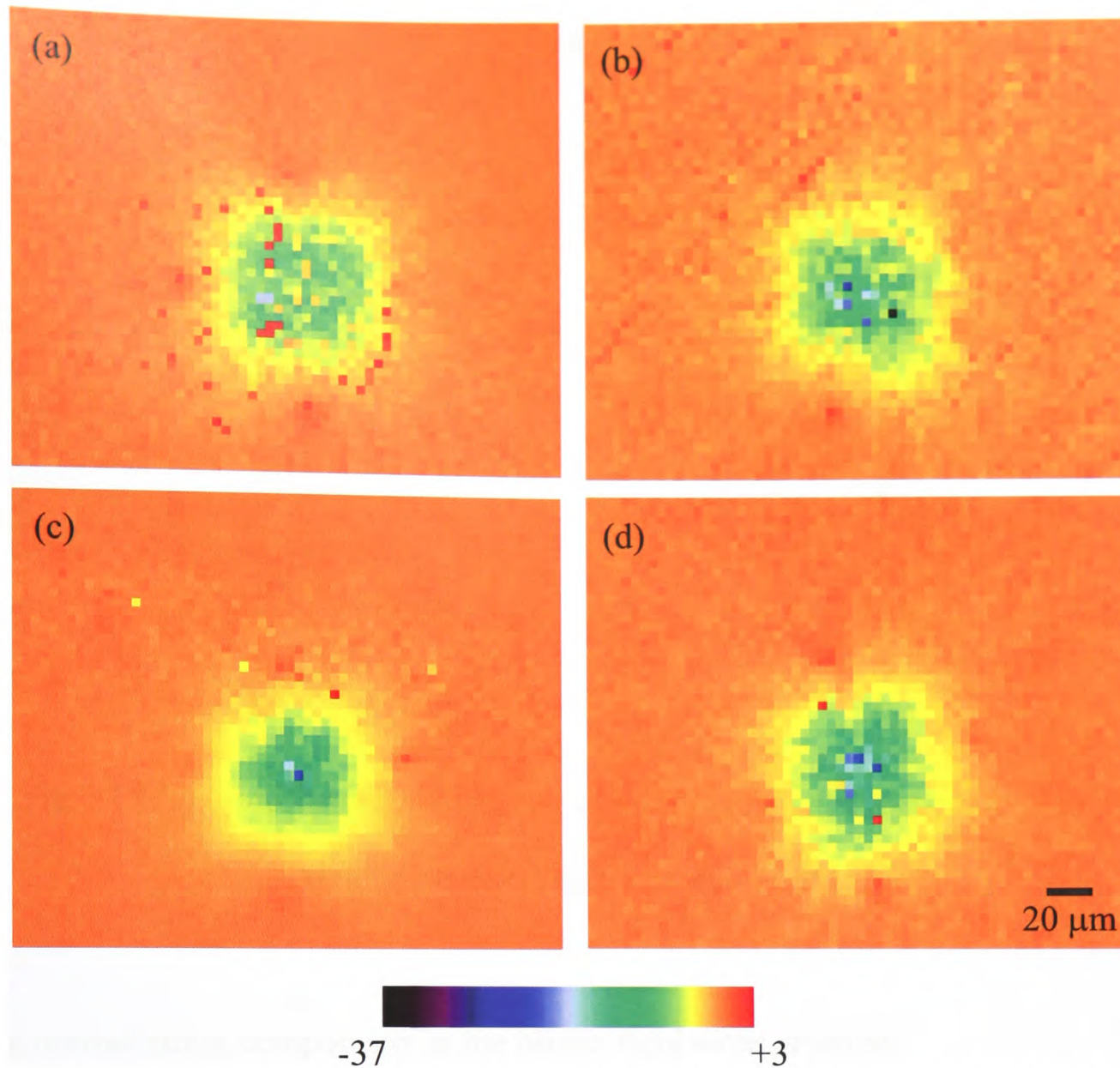


Fig 7.14 Peak position distribution from an area mapping around the indentation on  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/x \text{ vol.}\% \text{ SiC}$  nanocomposite ( $x=2, 5, 10$ ), the unit is  $\text{cm}^{-1}$  shift.

(a)  $\text{Al}_2\text{O}_3$ ; (b) 2 vol.% SiC; (c) 5 vol.% SiC; (d) 10 vol. % SiC

## 7.4 Discussion

### 7.4.1 Yoffe's Model

In Chapter 2, Yoffe's analytical model to predict the elastic stress outside the plastic zone was introduced [12]. The elastic residual stress outside the plastic zone could be described by the "blister" field. The experimental results from the line scans were compared to Yoffe's model in the following sections, to check whether the stresses would be proportional to  $1/r^3$  as predicted. For convenience, the coordinate system and the assumed shape of the plastic zone used by Yoffe were redrawn in Fig 7.15.



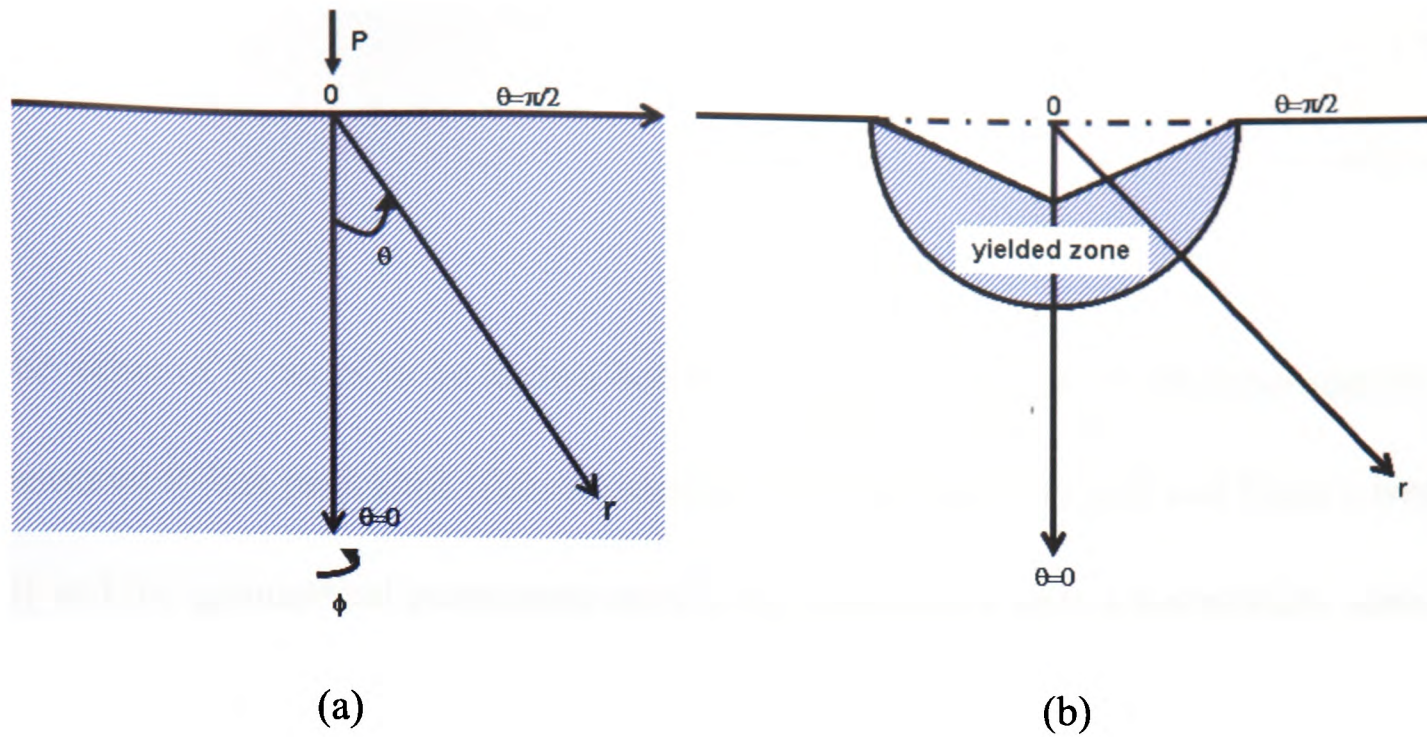


Fig 7.15 (a) Coordinate systems  $(r, \theta, \phi)$  and  $(\rho, \phi, z)$ ; (b) schematic diagram of yielded zone in Yoffe's model (after [12])

The normal stress components in the blister field were given as:

$$\sigma_r = \frac{B}{r^3} 4[(5 - \nu) \cos^2 \theta - (2 - \nu)] \quad (7.1 \text{ a})$$

$$\sigma_\theta = -\frac{B}{r^3} 2(1 - 2\nu) \cos^2 \theta \quad (7.1 \text{ b})$$

$$\sigma_\phi = \frac{B}{r^3} 2(1 - 2\nu)(2 - 3 \cos^2 \theta) \quad (7.1 \text{ c})$$

where  $r$  is the distance from the indentation center and  $\theta$  is the angle between the  $r$  axis and the loading direction as defined in Fig 7.15;  $\nu$  is the Poisson ratio,  $\nu=0.24$  for alumina materials [13];  $B$  is a constant that measures the strength of the field.

The line scan was carried out on the surface where  $\theta = \pi/2$ . Equation 7.1 then becomes:

$$\sigma_r = -\frac{7.04B}{r^3} \quad (7.2 \text{ a})$$

$$\sigma_\theta = 0 \quad (7.2 \text{ b})$$

$$\sigma_\phi = \frac{2.08B}{r^3} \quad (7.2 \text{ c})$$

The value of  $B$  needs to be known before relating the stresses to the experimentally measured R-line peak shift. The calculation of  $B$  was based on Cook and Pharr's work [3] and the geometrical parameters used in the calculation were schematically shown in Fig 7.16.

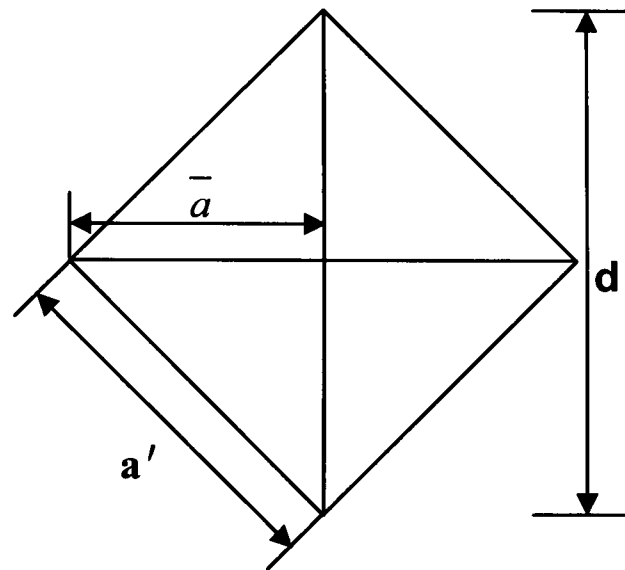


Fig 7.16 Geometrical parameters for Vickers indentation

$$\Delta V = \frac{5\pi B}{6E} \quad (7.3)$$

$$\Delta V = 0.068f \left( \frac{P}{H} \right)^{3/2} \quad (7.4)$$

$$H = \frac{P}{a'^2} \quad (7.5)$$

$$a' = \sqrt{2}\bar{a} = \frac{\sqrt{2}}{2}d \quad (7.6)$$

where  $\Delta V$  is the volume displaced by the indentation;  $E$  is the Young's modulus;  $P$  is



the load;  $H$  is the hardness of the material, defined by Equation 7.5;  $f$  is the factor accounting for the densification process during the indentation process.  $f$  was assumed to be 1 here, i.e. no densification of the material occurred during indentation;  $a$  is the length of the indent edge;  $d$  is the diagonal length of the indent and  $\bar{a}$  is half of  $d$ .

From Equations 7.3, 7.4, 7.5 and 7.6:

$$B = 9.192 \times 10^{-3} E d^3 \quad (7.7)$$

For  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites,  $E \approx 400$  GPa [14],  $d \approx 30$   $\mu\text{m}$ , so from Equation 7.7,  $B = 9.93 \times 10^4$  GPa $\cdot\mu\text{m}^3$ .

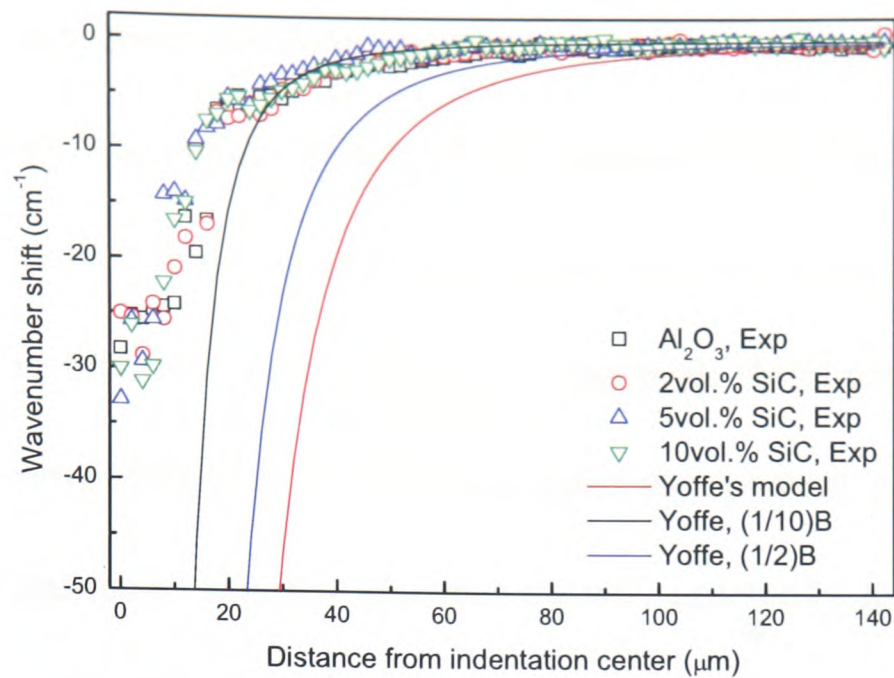
$$\begin{aligned} \Delta \nu_{R1} &= \frac{2.56 + 3.50 + 1.53}{3} (\sigma_r + \sigma_\theta + \sigma_\phi) \\ &= 2.53 \left( -\frac{7.04B}{r^3} + \frac{2.08B}{r^3} \right) \\ &= -\frac{1.25 \times 10^6}{r^3} \end{aligned} \quad (7.8)$$

where  $\Delta \nu$  has units of  $\text{cm}^{-1}$ , and  $r$  is measured in  $\mu\text{m}$ .

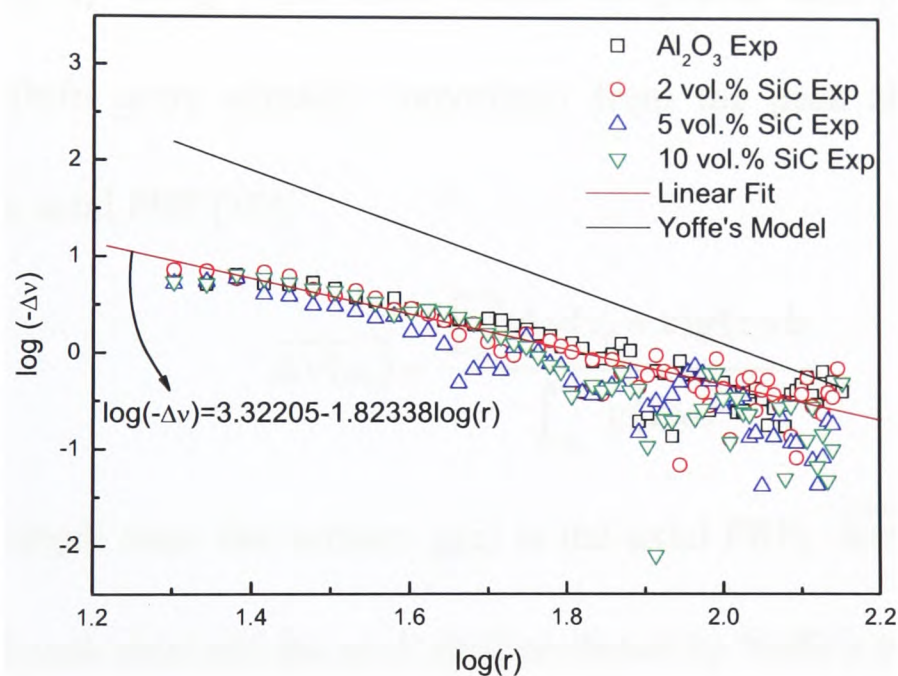
The comparison of the raw experimental results to Yoffe's model prediction for  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites is shown in Fig 7.17.

Seen from Fig 7.17, Yoffe's model successfully predicted the qualitative trend of the peak shifts along the distance to the indentation center, but the predicted peak shifts were much bigger than the experimental results, especially when  $r$  is small. The peak shifts and the distance to the indentation center followed the  $1/r^n$  rule well as shown in

Fig 7.17 (b), but  $n$  was about 1.8, significantly smaller than that proposed by Yoffe ( $n=3$ ). There are at least three possible reasons for the discrepancy: first, the effect of the  $B$  value. According to Equation 7.7,  $B$  is a function of  $E$  and  $d$ .  $d$  was measured experimentally and  $E$  came from the literature. Seen from Fig 7.17 (a), the  $B$  value significantly affected the fitting when  $r$  is small and smaller  $B$  (e.g.,  $B/10$ ) resulted a better fitting, particularly outside the plastic zone. The  $B$  value was probably overestimated in Equations 7.3-7.7 because the plastic zone was assumed to be rigid. In reality it will relax elastically under the constraint of the surrounding un-yielded material. The relaxation of indentation shape might also affect the  $B$  value (it is assumed to have the shape of the indenter as described in Equation 7.4). However, it is unrealistic that the error from these sources would vary the  $B$  value by more than a factor of 10 which is the change needed to approximate the experimental results. Second, the experimental peak shifts on the surface were actually volume averaged and most of the collected signals came from underneath the surface where the compressive stress predicted by Yoffe's model is less. This was proved in Fig 7.13: below the surface the peak shifts were smaller than those on the surface. Third, in Yoffe's model the stress relaxation due to cracks was not taken into account and this might also explain the lower compressive stresses measured, and also the  $1/r^{1.8}$  rather than the  $1/r^3$  dependence of stress.



(a)



(b)

Fig 7.17 Comparison between the raw experimental results and the prediction from Yoffe's model (Equation 7.8) on the peak shifts along a line scan on  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/x$  vol.% SiC nanocomposite ( $x=2, 5, 10$ ). (a) and (b) are with linear and logarithmic scales, respectively. In (a), three  $B$  values were used for Yoffe's model:  $B=9.93 \times 10^4 \text{ GPa} \cdot \mu\text{m}^3$ ,  $(1/10) B$  and  $(1/2) B$ . The smaller  $B$  values provided a better fitting to the raw experimental results but still overestimate the peak shift close to the plastic zone; In (b),  $r$  starts from outside the plastic zone ( $r \geq 20 \mu\text{m}$ ).

### 7.4.2 Convolution

As discussed above, the lower compressive stress measured from experiments might be due to the sampling volume effect, i.e, the measured peak shifts are actually the results of convolution from the real peak shifts with the probe response function (PRF). Since the PRF is known (Chapter 4), the peak shifts predicted by Yoffe's model can be convoluted with the PRF to predict the apparent peak shifts that are measured experimentally. This also provides a good opportunity to check the validity of Yoffe's model to predict the real peak shifts.

More specifically, by doing depth scans outside the plastic zone (see Fig 7.18), the apparent peak shifts were actually convoluted from the peak shift on the depth direction and the axial PRF [15]:

$$\overline{\Delta v(z_0)} = \frac{\int_{z_0}^{-z_0} \Delta v(z_0 + z)g(z)dz}{\int_{z_0}^{-z_0} g(z)dz} \quad (7.9)$$

where  $z_0$  is the depth from the surface;  $g(z)$  is the axial PRF;  $\overline{\Delta v(z_0)}$  and  $\Delta v(z_0)$  are the averaged peak shift and the peak shift predicted by Yoffe's model, respectively;  $t$  is the specimen thickness.

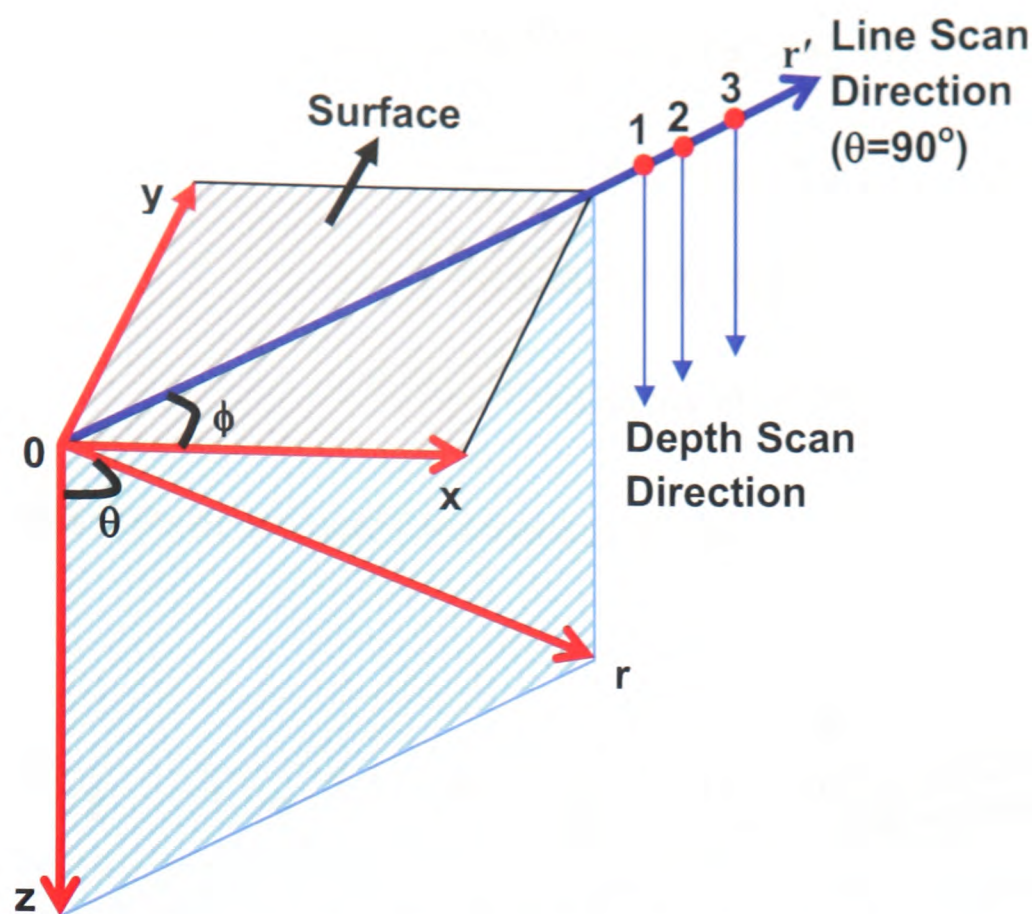


Fig 7.18 Schematic of the depth scans through positions on the route of the line scans (results given in Fig 7.13), in both (x, y, z) and (r, θ, ϕ) coordinates.

$\Delta v(z_0)$  was deduced from Yoffe’s model. In Fig 7.18, for a depth scan through a position with a distance to the indentation center of  $r'$  μm, and at a depth of  $z$  μm to the surface:

$$\cos \theta = \frac{z}{\sqrt{r'^2 + z^2}}$$

(7.10)

$$r = \sqrt{r'^2 + z^2}$$

(7.11)

For Al<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>/SiC nanocomposites:



$$\begin{aligned}
\Delta \nu_{R1} &= \frac{\Pi_a + \Pi_m + \Pi_c}{3} (\sigma_a + \sigma_m + \sigma_c) \\
&= \frac{\Pi_a + \Pi_m + \Pi_c}{3} (\sigma_r + \sigma_\theta + \sigma_\phi) \\
&= \frac{2.53B}{r^3} (14.88 \cos^2 \theta - 4.96) \\
&= \frac{2.51 \times 10^5}{r^3} (14.88 \cos^2 \theta - 4.96)
\end{aligned} \tag{7.12}$$

where  $\Delta \nu$  has units of  $\text{cm}^{-1}$ , and  $r$  is measured in  $\mu\text{m}$ .

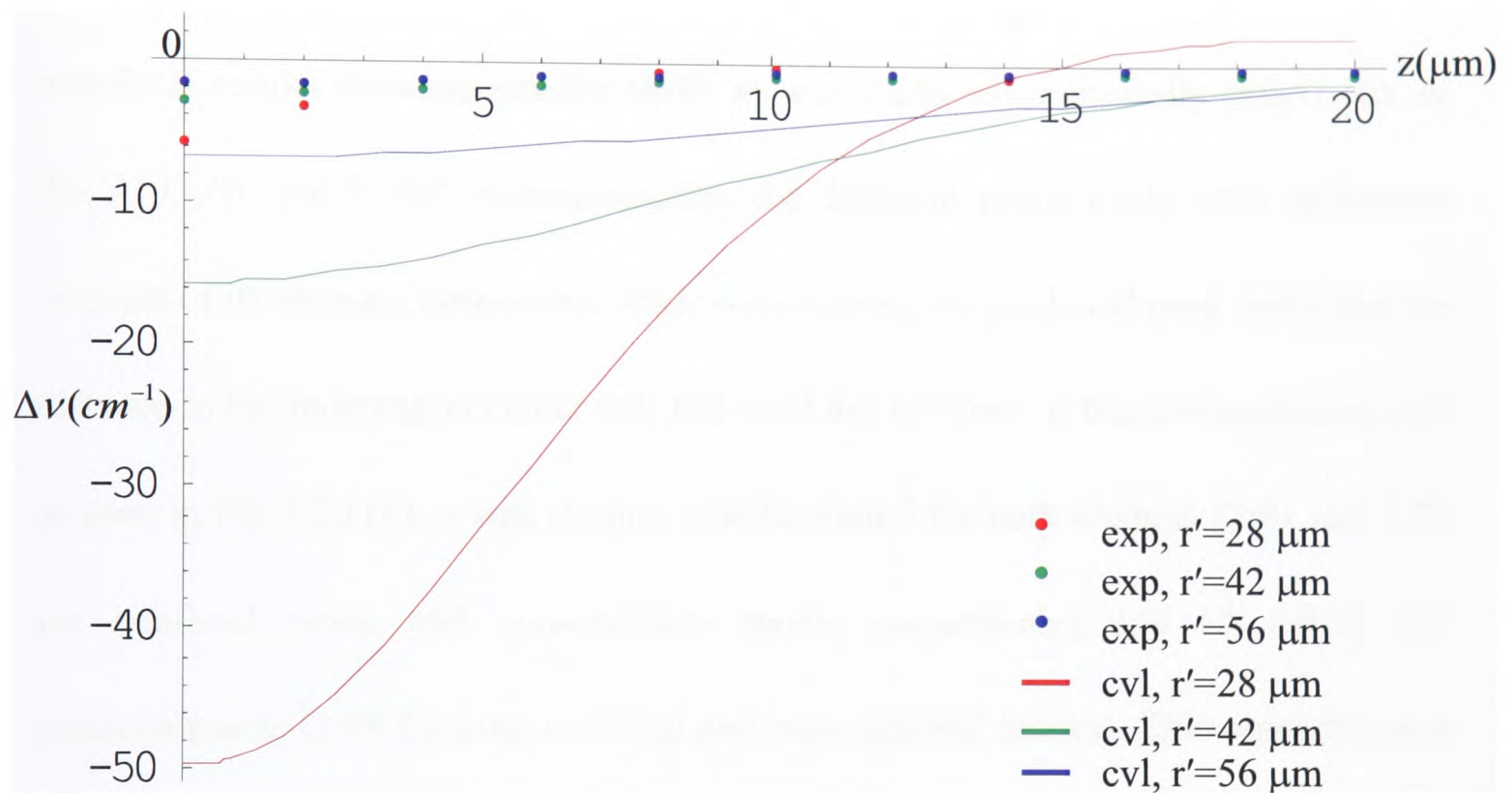


Fig 7.19 Comparison between the experimental results (denoted by *exp*) on the  $\text{Al}_2\text{O}_3$  specimen under the confocal mode, and the convolution results (denoted by *cvl*) based on Equation 7.9

From Equations 7.9, 7.10, 7.11 and 7.12, the peak shifts through a position on the surface with a distance to the indentation center of  $r'$   $\mu\text{m}$  could be predicted. The convolution was done numerically using the Mathematica (Wolfram Research, USA) software. In Fig 7.19, the experimental results on  $\text{Al}_2\text{O}_3$  obtained under confocal mode were compared with the prediction. The predicted peak shifts were much more



negative than the experimental results, as in Fig 7.17 (a).

Similarly, the convolution was carried out for depth scan results for  $\text{Al}_2\text{O}_3$  obtained under non-confocal mode, and for results of  $\text{Al}_2\text{O}_3/10$  vol.% SiC as well. The predicted peak positions on the surface ( $z=0$ ) along the distance to the indentation (as in the line scan) are given in Fig 7.20. Although the level of the predicted peak shift is still much higher than the experimental results, it clearly reflected the difference made by the different probe (confocal and non-confocal mode) in alumina, with the non-focal results showing smaller shifts as was found experimentally (Fig 7.12). In the  $\text{Al}_2\text{O}_3/10$  vol.% SiC nanocomposite, the different probe made little difference because of its stronger absorption. After convolution, the predicted peak shifts and the distance to the indentation center still followed the  $1/r^n$  ( $r=r'$  at this circumstance) rule as seen in Fig 7.20 (b).  $n$  was slightly smaller than 3 for both alumina (2.91 and 2.75 for confocal mode and non-confocal mode, respectively) and 10 vol.% SiC nanocomposite (2.98 for both confocal and non-confocal modes). This reduction in  $n$  is not sufficient to explain the experimental value ( $\sim 1.8$ ). Since the discrepancy between the experimental results and Yoffe's model cannot be explained by the volume averaging (convolution) effect, nor by the  $B$  value (see section 7.4.1), it seems that the stress relaxation by cracking is mainly responsible.

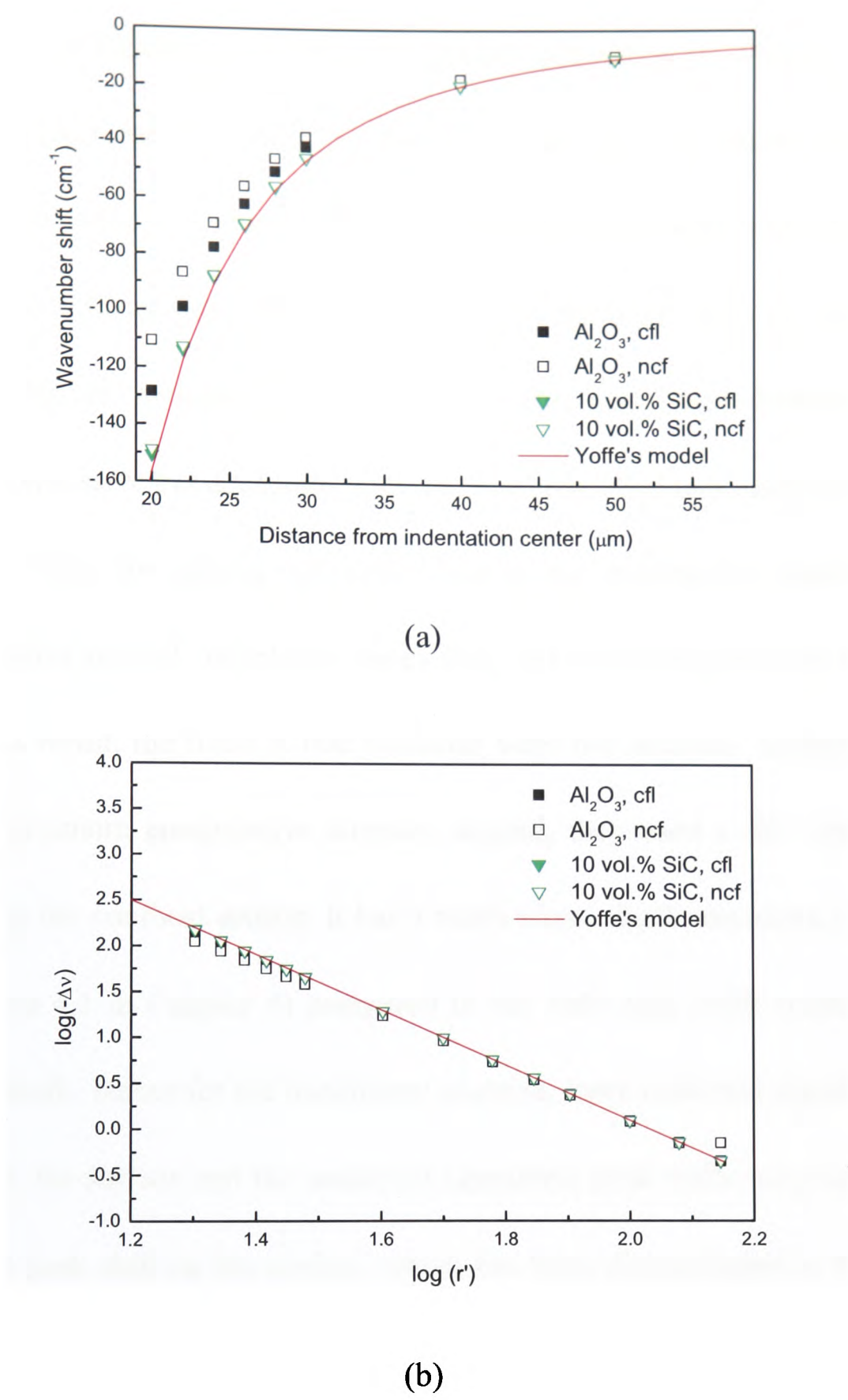
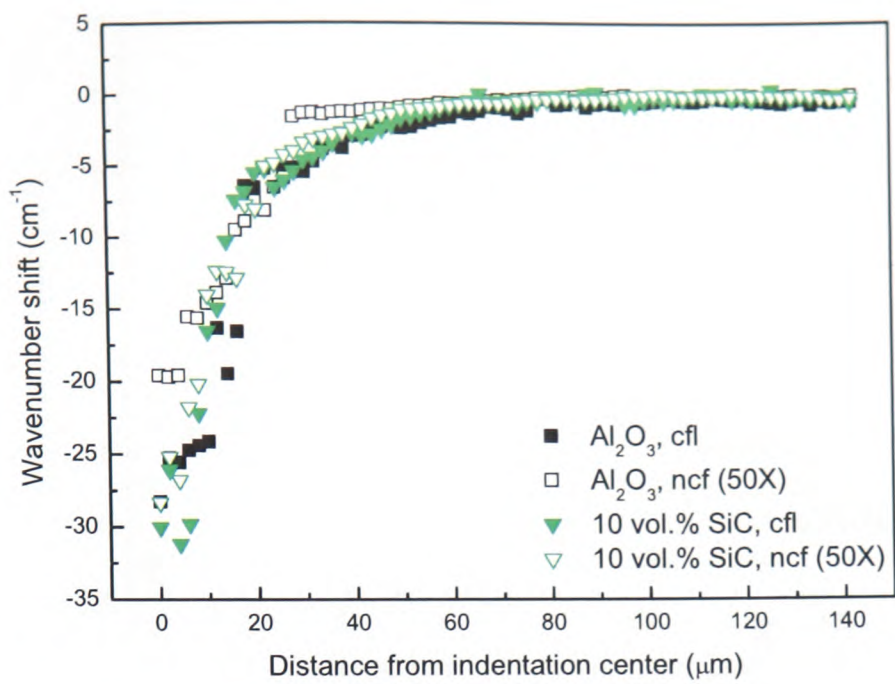


Fig 7.20 The predicted peak position distribution (convoluted) along the line scan direction through the indentation center. *cfl* and *ncf* denote confocal and non-confocal modes. (a) and (b) are with linear and logarithmic scales, respectively. In both plots,  $r'$  start from outside the plastic zone ( $r' \geq 20 \mu\text{m}$ ).

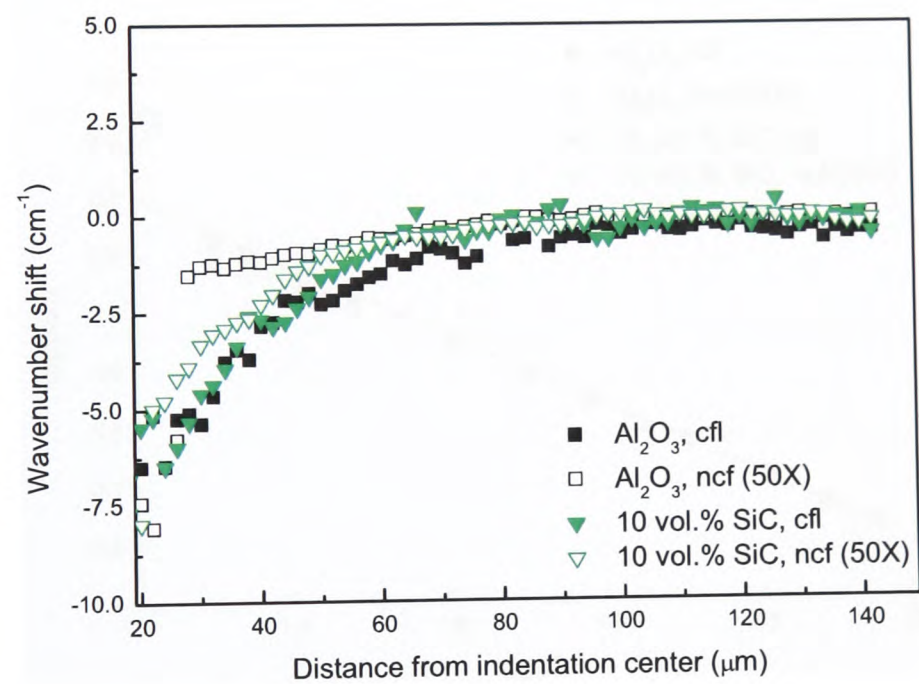
Recently, Wu et al. [16] used  $\text{Cr}^{3+}$  fluorescence microscopy technique to investigate the residual stress distribution across a 3N loaded indentation for alumina and  $\text{Al}_2\text{O}_3/x$  vol.% SiC nanocomposites ( $x=1, 5, 10$ ). They found that the maximum peak shifts appeared at the center of the indentation and the maximum peak shift was greater in nanocomposites. Away from the indentation center, the peak shifts for alumina were nearly zero; for the nanocomposites, the peak shifts away from indentation center were still compressive but decreased with distance. There are two main problems with their results: first, for spectra collected close to the indentation center (sampling volume includes part of the plastic zone) they did not distinguish the two sets of R-lines. As a result, the fitted R-line positions were not accurate, neither were their calculated maximum compressive stresses; second, they used a 40 $\times$  objective lens without using the confocal setting. It has a much worse depth resolution ( $\text{FWHM} > 10 \mu\text{m}$ , see Table 4.1 in Chapter 4) compared to the 100 $\times$  lens (with confocal setting) used in this work. Hence for the translucent alumina, more collected signal came from deeper below the surface and the measured (apparent) peak shifts were much smaller than the real peak shift on the surface, which has been demonstrated in Fig 7.21 and 7.22. The depth resolution has a smaller effect on the nanocomposites due to their stronger absorption. Consequently, the measured difference in peak shifts (at the same distance from the indentation center) between alumina and nanocomposites were enormously exaggerated due to the poor spatial resolution.

This work proved that Wu et al.'s measurement results are wrong as they did not

account for the sampling volume or the convolution effect. In reality, the results from this work show that there is a small difference between the alumina and the  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites. This was also exemplified in Fig 7.12, which showed that the peak shifts were very similar between alumina and nanocomposites if measured with high resolution. It also suggested that the  $100\times$  lens used in this work together with the confocal microscope setting proved to be capable of measuring the peak shifts that were close to the real peak shifts without being volume averaged.

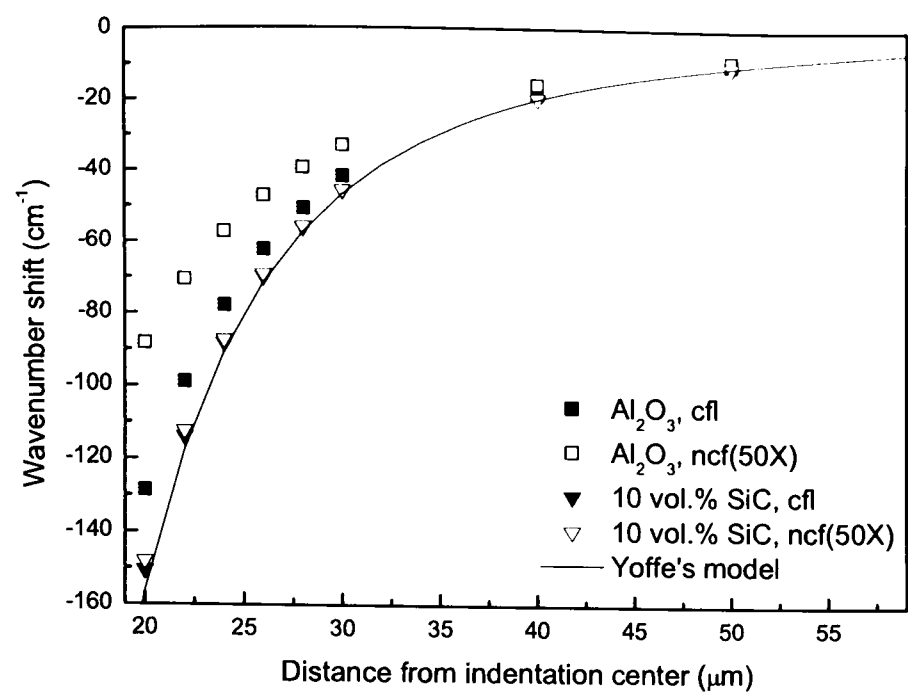


(a)

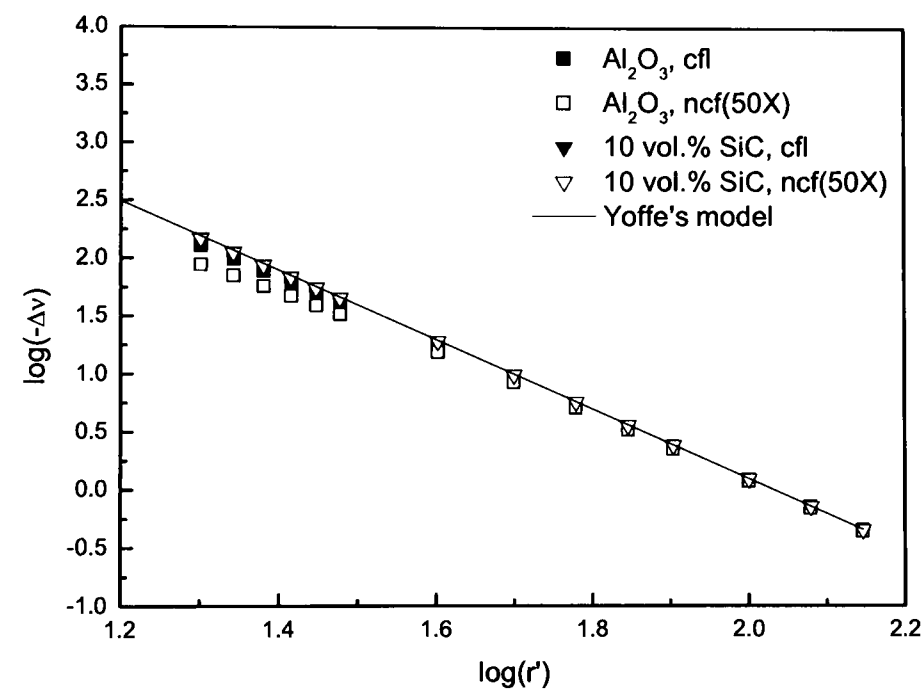


(b)

Fig 7.21 (a) Comparison of peak shift distribution along a line scan through the indentation center on Al<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>/ 10 vol.% SiC nanocomposite specimens, using non-confocal (50× lens) (denoted by *ncf* (50×)) and confocal mode (denoted by *cfl*), respectively. In (b), the X scale was expanded to start from outside the plastic zone.



(a)



(b)

Fig 7.22 The predicted peak position distribution (convoluted) along the line scan direction through the indentation center. *cfl* and *ncf* (50×) denote confocal and non-confocal (50× lens) modes. (a) and (b) are with linear and logarithmic scales, respectively. In both plots,  $r'$  start from outside the plastic zone ( $r' \geq 20 \mu\text{m}$ ).



## 7.5 Summary

1) Peak fitting for spectra close to the indentation center provides an effective method to estimate the size of the plastic zone. The broad and narrow peaks separated from the spectra are physically meaningful, representing the plastically deformed zone and the elastically deformed region outside the plastic zone.

2) Yoffe's model qualitatively predicts the trend of the peak shifts along the distance from the indentation center, but quantitatively overestimates the stress level by at least an order of magnitude and predicts a  $1/r^3$  stress dependence instead of the  $1/r^2$  dependence that was experimentally measured. The lack of quantitative agreement was explained mainly by the cracking induced stress relaxation.

3) The peak shifts measured from line scans across the indentation center were similar between  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites when high resolution was achieved. Poor spatial resolution resulted in the measured peak shifts in alumina being averaged within a large depth due to its transparency and hence deviating much from the real peak shift on the surface, thus exaggerating the difference between  $\text{Al}_2\text{O}_3$  and the  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites, in which the depth of collected signal was limited by the opaque nature of the material.

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## Chapter 8

### Conclusions and Future Work

#### 8.1 Conclusions

In this work, grinding/polishing and indentation induced surface residual stresses were investigated using confocal  $\text{Cr}^{3+}$  fluorescence microscopy. The high spatial resolution of this technique enabled the measurement of local stress variation rather than the mean stress average over a large sampling volume as is measured by other techniques.

The main focus of this work was to understand where the collected signal came from. Due to the translucency of the alumina materials, a substantial amount of fluorescence signal comes from underneath the surface even with the high resolution. As a result, the experimentally measured stresses (actually measured indirectly by the R-line shifts) were actually averaged over a finite volume. The probe response function (PRF) has to be taken into account to recover the real stress. In this work, the PRF in the depth direction was proposed and it was used to convolute with the stress functions in the grinding/polishing and indentation conditions to predict the apparent stresses that were measured experimentally.

One of the main questions this work aimed to answer is: how different are the residual stresses and the plastic deformation between alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites?

Is the difference real or simply because of the measurement technique?

The main conclusions from this work are summarized below.

### 8.1.1 Spatial Resolution

The axial resolution of the microscope was measured from the FWHM of the intensity-defocus distance profiles from different materials including silicon wafer, sapphire and ruby. The best axial resolution reaches  $\sim 2 \mu\text{m}$  when the  $100\times$  objective lens and the confocal settings are used; the best lateral resolution is about  $1.5 \mu\text{m}$ .

Mathematical descriptions for the probe response in the depth direction were proposed for different materials including sapphire, ruby, polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites, taking into account refraction, absorption, and scattering. They described the intensity-defocus distance profiles reasonably well.

### 8.1.2 Grinding and Polishing of Polycrystalline Alumina

The residual stresses on the ground and polished polycrystalline aluminas were found to depend greatly on the local environment, and particularly whether plastically deformed, ground regions of the surface were examined or those with surface fracture and pullouts. Both tensile and compressive stresses exceeding 1 GPa were present in the ground surfaces of aluminas, and compressive stresses were present in the pullout region.

Separation of the broad peaks and narrow peaks by the curve fitting for spectra

collected near the ground/polished surfaces enabled separation of the stresses in the ground region into two physically distinct regions: broad peaks representing the plastically deformed surface layer, and narrow peaks representing the elastically deformed material beneath.

A residual stress model taking into account the pullout was proposed using an array of dislocations. The experimental results supported the main features of the model when the PRF is taken into account.

The stress level and the thickness of the stressed layer were obtained from the fitting of the prediction based on the convolution of the stress function with the PRF, to the experimental results. The depth of plastic deformation was found to be deeper in grinding than in the polished condition for the same grain sized alumina. Tensile stresses that were present in the ground condition were produced by the pullouts and they would lead to crack growth and strength degradation; however, in the polished condition, due to the lack of pullouts, the tensile stresses were prevented while the compressive stresses remained. The residual stress level in the ground condition is slightly higher than that in the polished condition.

### **8.1.3 Grinding of $\text{Al}_2\text{O}_3/\text{SiC}$ Nanocomposites**

Tensile stresses were found on ground surfaces of alumina and  $\text{Al}_2\text{O}_3/ 2 \text{ vol. \% SiC}$  nanocomposite, but not on the 5 and 10 vol.% SiC nanocomposites, mainly due to



their being fewer pullouts in the latter materials.

The depths of deformation of alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites were similar ( $\sim 1 \mu\text{m}$ ). The maximum compressive stresses in the 5 and 10 vol.% SiC nanocomposites were higher than those in alumina and 2 vol.% SiC nanocomposite but this was again due to the reduction in pullout. The main difference between the ground alumina and ground  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites is not the plastic deformation but the brittle fracture behavior.

#### 8.1.4 Indentation

Curve fitting for spectra close to the indentation is crucial to measure the peak shifts that characterize the residual stress, and it provides an effective method to estimate the size of the plastic zone based on the R1/R2 peak area ratio. The broad and narrow peaks separated from the spectra are physically meaningful, representing the plastically deformed zone and the elastically deformed region outside the plastic zone.

Yoffe's analytical model qualitatively predicts the trend of the peak shifts along the distance from the indentation center but quantitatively overestimates the stress level by at least an order of magnitude, and predicts a  $1/r^3$  stress dependence instead of the  $1/r^2$  dependence that was experimentally measured. The quantitative discrepancy was attributed mainly to the cracking that relaxed the stresses.

The peak shifts measured from line scans across the indentation center were similar between  $\text{Al}_2\text{O}_3$  and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites when the best confocal resolution was achieved. Poor spatial resolution resulted in the measured peak shifts in the translucent alumina being averaged within a large depth and hence deviating much from the real peak shift on the surface, thus exaggerating the difference between  $\text{Al}_2\text{O}_3$  and the  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites, in which the depth of the collected signal was limited due to the strong absorption.

## 8.2 Suggestions for Future Work

### 8.2.1 Improvement of the PRF

In Chapters 5 and 6, the fitting of to the theoretical prediction (convolution of the stress function with the PRF) to the narrow peaks from the ground region and peaks from the pullout region was not satisfactory in some of the results. This was attributed to the tail part of the proposed PRFs (section 5.6.1). There are several possible reasons for this and further work looking into them to improve the PRF would be worthwhile:

- 1) The PRFs proposed in Chapter 4 were essentially modified Lorentzian functions. However, as seen from Fig 4.2 the Lorentzian function does not describe the probe response for the silicon wafer very well when the laser is focused above the surface. The solution to the tail problem might start by modifying this basic function.

- 2) The assumption of the second Lorentzian function to compensate for the

“scattering” effect was not fully justified. Some unrealistic assumptions were also made to simplify the mathematical problem, such as the use of the same coefficients for absorption and scattering and the same light path. Although overall the double-Lorentzian function describes the probe response curves in the polycrystalline alumina and  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites very well, these assumptions might be the reasons behind the unsatisfactory fitting in the tail part.

3) Degradation of resolution (broadening of the width with depth) was not taken into account in the double-Lorentzian function. This was not true for polycrystalline aluminas as they are translucent and the penetration depths of the laser in them are not negligible. This should have a less significant effect in the more opaque  $\text{Al}_2\text{O}_3/\text{SiC}$  nanocomposites but seen from the fitting results in Chapter 6, the tail problem still exists.

### **8.2.2 Shape and Size of the Plastic Zone**

In Chapter 7, the R1/R2 peak area ratio was used to estimate the size of the plastic zone. This needs further justification and the shape and size of the plastic zone can be estimated by other methods. Apichart Limpichaipanit used the sequential polishing method [1] to map (two dimensionally) the stress distribution at different depths of the indentation (starting from surface and finally to a depth underneath the surface where no residual stress could be detected) and determined the shape and size of the compressive plastic zone and the less compressive elastic regions based on the false

color maps of the peak shift, similar to those in Fig 7.14. However, the false color maps are tricky and might be deceptive and it should be borne in mind that in the plastic zone the peak shifts shown in the area mapping come from two-peak fittings only and are hence not accurate. Apart from the peak shifts, the peak broadening is also a good indicator of the plastic zone as the stress variation inside the plastic zone is more complicated than that outside it and the complexity of the stress variation is reflected on the peak width. The peak broadening was not analyzed in this work and might be worth further investigation.

### 8.2.3 Beyond Yoffe's Model

Yoffe's model is simple and analytical in predicting the stress distribution around the indentation. However, it can not be used to predict the stress field in the plastically deformed region. As a result, the peak shifts inside the plastic zone in Chapter 7 that came from the four-peak fitting cannot be understood very well. They are important as they determine the maximum compressive stress in the plastic zone. Modeling of the stress field suitable for the plastic zone is thus required. In addition, Yoffe's model does not account for the stress relaxation by the cracks. The experimentally measured  $1/r^2$  instead of  $1/r^3$  stress dependence on distance (Chapter 7) needs theoretical explanation.

### 8.2.4 Cathodoluminescence

Although compared to other residual stress measurement techniques optical

fluorescence microscopy has the advantage of better resolution, its best resolution is still limited to a micrometer scale. The resolution is limited by the wavelength of the visible light. This limitation can be overcome by using electrons rather than photons to stimulate the fluorescence, i.e., using scanning electron microscope (SEM)-based cathodoluminescence (CL) [2]. Todd et al. experimentally measured the spatial resolution of CL to be 550 nm with an acceleration voltage of 10 kV [3]. A submicron scale measurement is then possible using the CL technique. In addition, the high resolution stress measurements will be correlated with direct SEM observation of the finer microstructural details than could be seen under the optical microscope.

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