

Nuclear Orientation Studies of Isotopes
Far from Stability



Austyn Glyn Griffiths
Hertford College

A thesis submitted in partial fulfillment of the requirements
for the degree of Doctor of Philosophy
in the University of Oxford

Michaelmas term 1989

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Abstract

Low Temperature Nuclear Orientation (LTNO) is an important technique in the study of nuclei far from stability. The theory of LTNO and its applications to the measurement of static nuclear moments and other quantities of spectroscopic interest are reviewed.

Among the nuclei off the line of stability those in the $A \sim 75$ region are of considerable interest, exhibiting large quadrupole deformations, triaxiality and shape coexistence. LTNO measurements performed on neutron deficient bromine isotopes have yielded the static magnetic dipole moments of the nuclei $^{72g,72m,74m,75,76,77}\text{Br}$, spectroscopic information on $^{72-77}\text{Se}$ and also identified the ground state spin of ^{73}Br as $1/2^-$.

Existing odd A and odd-odd particle-rotor computer codes have been extended in order to include a Variable Moment of Inertia (VMI) asymmetric rotational core. The formalism necessary for this modification is developed.

The measured magnetic moments are interpreted within the framework of this particle - VMI rotor model. It is shown that the systematic reduction in the moments of the odd A nuclei $^{75-81}\text{Br}$ characterizes the transformation of the *prolate* ground state configuration from largely $\pi[301]3/2$ in $^{79,81}\text{Br}$ to almost pure $\pi[312]3/2$ in $^{75,77}\text{Br}$. This trend is fully consistent with the increase in deformation towards the lower masses suggested by the known electric quadrupole moments. In contrast, the ground state spin of ^{73}Br can only be interpreted in terms of an *oblate* nuclear shape. This is the first evidence for the predicted prolate-oblate shape transition in the bromine nuclei. In addition the magnetic moment of $^{74}\text{Br}^m$, by identifying a $\pi[431]3/2\nu[422]5/2$ configuration, strongly suggests a positive parity assignment for the isomeric state.

Finally, an experiment to search for possible T-violation effects in nuclear gamma decay is described. Using coincidence techniques, a measurement of the T-odd P-even quantity $(\mathbf{I} \cdot \mathbf{k} \times \mathbf{e})(\mathbf{I} \cdot \mathbf{k})(\mathbf{I} \cdot \mathbf{e})$ has lead to a limit on the T-violating phase angle between the E2 and M1 matrix elements associated with the 604 keV gamma transition in ^{192}Pt of $\sin \eta = \pm 11(12) \times 10^{-3}$.

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Chapter 1

Low Temperature Nuclear Orientation

1.1 Introduction

The probability of emission of electromagnetic radiation depends, in general, on the angle θ between the expectation value $\langle \mathbf{J} \rangle$ of the angular momentum vector of the radiating system and the direction $\mathbf{k}$ in which the photons are observed with sharp linear momentum $\mathbf{p} = \omega \mathbf{k}$ and helicity τ .

For a source of radiation consisting of an ensemble of many radiating systems with their $\mathbf{J}$ vectors randomly oriented, $\langle \mathbf{J} \rangle = 0$ and the radiation is isotropic in space. In order to observe an anisotropic radiation pattern the spatial distribution of the angular momentum vectors must favour a definite direction, so that $\langle \mathbf{J} \rangle \neq 0$. The intensity and polarization properties of the emitted radiation are then a function of the angles between the orientation axes and the directions of observation $\mathbf{k}$ and the polarization $\mathbf{P}$ of the radiation.

If the polarization of radiation is not observed then by definition the *directional distribution* $W(\mathbf{k})$ of the radiation is measured. If the radiation is observed with a detector which is sensitive to the polarization properties $\mathbf{P}$ of the radiation then the *polarization distribution* $W(\mathbf{k}, \mathbf{P})$ is measured. The term *angular distribution* is used to refer to both.

Oriented ensembles of nuclei may be prepared by three different methods:

1. Extranuclear static and dynamic orientation. (Low temperature orientation, optical pumping, radiofrequency methods).
2. Orientation by absorption of nuclear radiation of well defined direction and polarization. (Nuclear reactions, Coulomb excitation).
3. Orientation by observing a preceding emitted radiation in a well defined direction and with well defined properties. (Angular correlations, nuclear cascade radiation).

In Low Temperature Nuclear Orientation (LTNO), which forms the main subject of this work, orientation is achieved through the interaction of external fields with either the static magnetic dipole moment or the static electric quadrupole moment of the nucleus. At very low

temperatures the energetically lower states, m -states in the case of magnetic interactions, become preferentially populated according to the Boltzmann distribution. Thus in thermodynamic equilibrium various degrees of orientation are obtained depending on the relative magnitudes of the energy splitting of the nuclear substates and the randomizing thermal energy, kT .

The following sections will deal with how the angular distribution of gamma radiation is related to the various nuclear properties of interest and to the orienting interaction.

1.2 The angular distribution of radiation from an oriented source

1.2.1 The description of mixed quantum states by density matrices

A physical system is in a *pure state* if every value of a complete set of commuting observables has been measured. There is then maximum information known about the state under consideration which can thus be described uniquely by a complete set of quantum numbers.

An angular distribution measurement does not result in a complete knowledge of the individual nuclei nor the individual quanta, only ensemble averages are observed. The statistical nature of the the ensemble is involved and maximum information is not available. The quantum mechanical description of such *mixed states* requires *incoherent superpositions* of pure quantum states. Thus an ensemble cannot be described by state vectors, but is instead described by a *density matrix* [1].

Consider a pure state $|\Psi\rangle$, which by selecting a coordinate system in Hilbert space can be described by its projections onto all coordinate axes. Choosing the complete orthonormal set $|\phi\rangle$ for this purpose, then

$$|\Psi\rangle = \sum_{\phi} |\phi\rangle \langle\phi|\Psi\rangle \quad (1.1)$$

where the transformation brackets $\langle\phi|\Psi\rangle$ are of course unitary. The expectation value of any operator a in the state $|\Psi\rangle$ is therefore

$$\langle\Psi|a|\Psi\rangle = \sum_{\phi\phi'} \langle\Psi|\phi'\rangle \langle\phi'|a|\phi\rangle \langle\phi|\Psi\rangle. \quad (1.2)$$

A mixed state consisting of an ensemble of several independent nuclei must then be described by an *incoherent* superposition of the possible pure states $|\Psi_n\rangle$, weighted according to the proportion of nuclei $g(n)$ in those pure states. The expectation value of the operator a in such a mixed state is therefore given by the weighted average of the expectation values $\langle\Psi|a|\Psi\rangle$

$$\langle a \rangle = \sum_{n=1}^N g(n) \langle \Psi_n | a | \Psi_n \rangle / \sum_{n=1}^N g(n). \quad (1.3)$$

It is convenient to normalize the weights $g(n)$ to unity

$$\sum_{n=1}^N g(n) = 1. \quad (1.4)$$

Substituting (1.2) into (1.3) and using (1.4) one can write

$$\langle a \rangle = \sum_{n, \phi, \phi'} \langle \phi | \Psi_n \rangle g(n) \langle \Psi_n | \phi' \rangle \langle \phi' | a | \phi \rangle \quad (1.5)$$

or alternatively

$$\langle a \rangle = \sum_{\phi, \phi'} \langle \phi | \rho | \phi' \rangle \langle \phi' | a | \phi \rangle \quad (1.6)$$

where

$$\langle \phi | \rho | \phi' \rangle = \sum_{n=1}^N \langle \phi | \Psi_n \rangle g(n) \langle \Psi_n | \phi' \rangle. \quad (1.7)$$

The state of the ensemble is thus completely characterized by the *density matrix* ρ whereas the matrix A , with elements $\langle \phi' | a | \phi \rangle$, depends only upon the operator a and the representation $|\phi\rangle$. With these definitions (1.6) may be rewritten in the form

$$\langle a \rangle = \text{Tr}(\rho A). \quad (1.8)$$

The density matrix can be considered as a representation of the density operator

$$\rho_{op} = \sum_{n=1}^N |\Psi_n\rangle g(n) \langle \Psi_n| \quad (1.9)$$

in the basis $|\phi\rangle$. The density operator completely describes the state of the ensemble, be it mixed (at least two $g(n) > 0$) or pure (only one non-zero $g(n)$).

1.2.2 The statistical tensors

In angular distribution problems the symmetries of the ensemble with respect to rotations are of interest and so it is natural to choose the basis states $|\phi\rangle$ to be the eigenstates $|\varrho jm\rangle$ of the angular momentum operator $\mathbf{J}^2$ and its projection onto the z -axis, J_z . The symbol ϱ represents the other quantum numbers needed to specify the states uniquely. The matrix elements of the density operator in this representation are

$$\rho(\varrho_1 j_1 \varrho_2 j_2) = \langle \varrho_1 j_1 m_1 | \rho | \varrho_2 j_2 m_2 \rangle = \sum_n \langle \varrho_1 j_1 m_1 | \Psi_n \rangle g(n) \langle \Psi_n | \varrho_2 j_2 m_2 \rangle. \quad (1.10)$$

The *grand density matrix* $\rho(\eta)$ which describes an ensemble of nuclei η in all their possible states is a square matrix with $(2j_1 + 1) + (2j_2 + 1) + \dots$ rows and columns, where the j_i are the angular momenta of the different nuclear levels or of the different daughter nuclei and radiation fields into which the nuclei η might decay. The density matrix $\rho(j_1 j_2)$ is just a $(2j_1 + 1) \times (2j_2 + 1)$ submatrix of this grand matrix.

The angular momentum states $|jm\rangle$ transform under a rotation of the quantization axis from $S(xyz)$ to $S(\bar{x}\bar{y}\bar{z})$ according to

$$|j\bar{m}\rangle_{\bar{S}} = \sum_m |jm\rangle_S D_{m\bar{m}}^{j*}(S \rightarrow \bar{S}) \quad (1.11)$$

where the D -matrices are defined in Brink and Satchler [2]. The elements of the density matrix transform under the same rotation according to

$$\langle j_1 \bar{m}_1 | \rho | j_2 \bar{m}_2 \rangle_{\bar{S}} = \sum_{m_1 m_2} D_{m_1 \bar{m}_1}^{j_1}(S \rightarrow \bar{S}) \langle j_1 m_1 | \rho | j_2 m_2 \rangle_S D_{m_2 \bar{m}_2}^{j_2*}(S \rightarrow \bar{S}). \quad (1.12)$$

Making use of the Clebsh-Gordon series of D -matrices [2] then

$$\begin{aligned} \langle j_1 \bar{m}_1 | \rho | j_2 \bar{m}_2 \rangle_{\bar{S}} &= \sum_{m_1 m_2 \lambda} \langle j_1 m_1 | \rho | j_2 m_2 \rangle_S (-1)^{m_2 - \bar{m}_2} (2\lambda + 1) \begin{pmatrix} j_1 & j_2 & \lambda \\ m_1 & -m_2 & q \end{pmatrix} \\ &\times \begin{pmatrix} j_1 & j_2 & \lambda \\ \bar{m}_1 & -\bar{m}_2 & \bar{q} \end{pmatrix} D_{\bar{q}\bar{q}}^{\lambda*}(S \rightarrow \bar{S}). \end{aligned} \quad (1.13)$$

After multiplying both sides of this equation by the factor

$$(-1)^{j_2 + \bar{m}_2} (2\lambda + 1)^{\frac{1}{2}} \begin{pmatrix} j_1 & j_2 & \lambda \\ \bar{m}_1 & -\bar{m}_2 & \bar{q} \end{pmatrix}$$

and summing over $\bar{m}_1$ and $\bar{m}_2$ (keeping $\bar{q}$ fixed) one obtains, because of the orthogonality relations of the 3- j symbols,

$$\left\{ \sum_{\bar{m}_1} \langle j_1 \bar{m}_1 | \rho | j_2 \bar{m}_2 \rangle_{\bar{S}} (-1)^{j_2 + \bar{m}_2} (2\lambda + 1)^{\frac{1}{2}} \begin{pmatrix} j_2 & j_1 & \lambda \\ -\bar{m}_2 & \bar{m}_1 & \bar{q} \end{pmatrix} \right\} = \sum_q \left\{ \sum_{m_1} \langle j_1 m_1 | \rho | j_2 m_2 \rangle_S (-1)^{j_2 + m_2} (2\lambda + 1)^{\frac{1}{2}} \begin{pmatrix} j_1 & j_2 & \lambda \\ -m_2 & m_1 & q \end{pmatrix} \right\} D_{q\bar{q}}^{\lambda*}(S \rightarrow \bar{S}). \quad (1.14)$$

The linear combinations of the elements of $\rho(j_1 j_2)$ enclosed in braces transform like spherical tensors of rank λ . Due to this simple transformation property it is more convenient to use these linear combinations than the matrix elements themselves. Thus it is possible to define the *statistical tensors* $\rho_q^\lambda(j_1 j_2)$ to be

$$\rho_q^\lambda(j_1 j_2) = \sum_{m_1} \langle j_1 m_1 | \rho | j_2 m_2 \rangle (-1)^{j_2 + m_2} (2\lambda + 1)^{\frac{1}{2}} \begin{pmatrix} j_2 & j_1 & \lambda \\ -m_2 & m_1 & q \end{pmatrix}. \quad (1.15)$$

This represents the vector relationship $\lambda = \mathbf{J}_2 - \mathbf{J}_1$ and so the rank of the statistical tensor is an integer in the range $|j_2 - j_1|$ to $j_2 + j_1$.

1.2.3 Oriented nuclear states

If the interacting fields that cause the orientation of the ensemble have an axis of cylindrical symmetry, which in the case of LTNO is that defined by the direction of the external field, then the density operator (1.9) may be expressed in the form

$$\rho_{op} = \sum_{m=-I}^{+I} |Im\rangle g(m) \langle Im| \quad (1.16)$$

where the eigenstates $|Im\rangle$ of the nuclear spin operator I_z are defined with respect to the symmetry z -axis. The corresponding density matrix $\rho(I)$ in the angular momentum representation $|I\bar{m}\rangle$ chosen with respect to some representation-coordinate system $\bar{S}$ is

$$\langle I\bar{m} | \rho | I\bar{m}' \rangle = \sum_m \langle I\bar{m} | Im \rangle g(m) \langle Im | I\bar{m}' \rangle. \quad (1.17)$$

If one chooses $\bar{S}$ to be the symmetry frame defined by the external field then the density matrix becomes diagonal

$$\langle Im | \rho | Im' \rangle = g(m) \delta_{mm'} \quad (1.18)$$

and the weighting factors $g(m)$ are then simply the relative populations of the states $|Im\rangle$ in the ensemble.

An ordinary ensemble of nuclei with spin I at room temperature has in general no fixed orientation axis in space, hence it must be invariant with respect to three dimensional rotations. Thus the statistical tensors describing such an random ensemble must satisfy the relation

$$\rho_q^\lambda(I)_{\bar{S}} = \sum_q \rho_q^\lambda(I)_S D_{q\bar{q}}^{\lambda*}(S \rightarrow \bar{S}) = \rho_{\bar{q}}^\lambda(I)_S \quad (1.19)$$

for any rotation $S \rightarrow \bar{S}$. This is only possible if D^λ is the unit matrix ($q = \bar{q} = \lambda = 0$) or if $\rho_q^\lambda(I)$ vanishes. Hence the statistical tensor in this case is

$$\rho_0^0(I) = (2I + 1)^{-\frac{1}{2}} \quad (1.20)$$

with corresponding density matrix

$$\rho(I) = (2I + 1)^{-1} \mathbf{1} \quad (1.21)$$

expressing the fact that the population is uniformly distributed amongst the various $|Im\rangle$ states.

When orientation is achieved, unequal populations of the $|Im\rangle$ states occur giving rise to statistical tensors $\rho_q^\lambda(I)$ with $\lambda \neq 0$. With the axis of cylindrical symmetry, the z -axis, defined by the applied field it is required that the statistical tensors be invariant with respect to rotations through an arbitrary angle α about this axis. This rotation is represented by

$$D_{q\bar{q}}^{\lambda*}(\alpha, 0, 0) = e^{-iq\alpha} \delta_{q\bar{q}} \quad (1.22)$$

and leads to the condition

$$\rho_{\bar{q}}^\lambda(I) = \sum_q \rho_q^\lambda(I) D_{q\bar{q}}^{\lambda*}(\alpha, 0, 0) = \rho_q^\lambda(I) e^{-iq\alpha} = \rho_q^\lambda(I). \quad (1.23)$$

Since α is arbitrary this equation implies that $q = \bar{q} = 0$. Thus with cylindrical symmetry the statistical tensors can be uniquely described by the series of parameters, the *orientation parameters* $B_\lambda(I)$ [3], where

$$B_\lambda(I) = (2I + 1)^{\frac{1}{2}} \rho_0^\lambda(I) = \sum_m (-1)^{I+m} [(2I + 1)(2\lambda + 1)]^{\frac{1}{2}} \begin{pmatrix} I & I & \lambda \\ -m & m & 0 \end{pmatrix} g(m). \quad (1.24)$$

The orientation parameters are normalized to unity: $B_0(I) = 1$.

The form of the interaction used in LTNO is now introduced explicitly. In principle this could be either the interaction of the nuclear static quadrupole moment with an applied electric field gradient or that of the static nuclear magnetic dipole moment with an applied magnetic

field. In this thesis only the latter interaction is considered. Using the direction of the $\mathbf{B}$ -field as the representation axis, the magnetic energy of the nuclear m -levels is

$$E(m) = -\frac{\mu B}{I}m \quad (1.25)$$

where μ is the nuclear magnetic dipole moment. In accordance with the Boltzmann distribution the populations in thermodynamic equilibrium are

$$g(m) = \langle m | \rho | m \rangle = e^{m\beta} / \sum_m e^{m\beta} \quad (1.26)$$

with

$$\beta = \frac{\mu B}{IkT}. \quad (1.27)$$

Hence the statistical tensors for this magnetically oriented ensemble are given by

$$\rho_0^\lambda(I) = (2I+1)^{-\frac{1}{2}} B_\lambda(I) = \left\{ \sum_m e^{m\beta} \right\}^{-1} \sum_m (-1)^{I+m} (2\lambda+1)^{\frac{1}{2}} \begin{pmatrix} I & I & \lambda \\ -m & m & 0 \end{pmatrix} e^{m\beta}. \quad (1.28)$$

Figure 1.1 illustrates the temperature dependence of the orientation parameters.

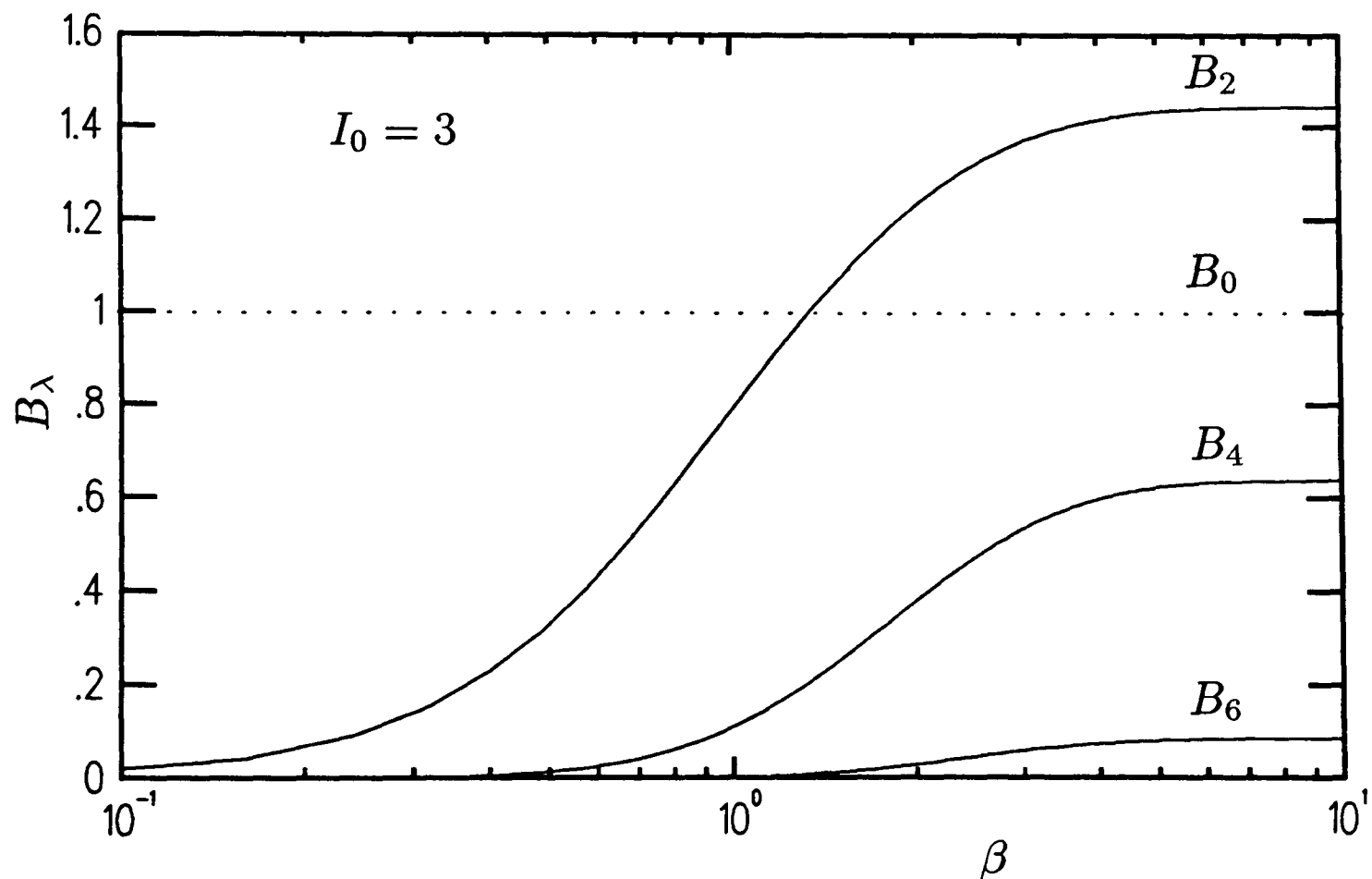


Figure 1.1. The non-zero orientation parameters, B_λ , for a magnetically orientated state of spin 3 as a function of $\beta = \mu B / IkT$.

1.2.4 The efficiency matrices and tensors

The probability of finding a system described by the density matrix ρ in the state $|\phi_n\rangle$ is seen from (1.6) to be given by

$$P(\phi_n) = \langle \phi_n | \rho | \phi_n \rangle. \quad (1.29)$$

Generally it is necessary to calculate this probability when the ensemble is observed by a measuring device which responds to the different pure states with different efficiencies. In direct analogy with the density matrix of the ensemble, such a detector must be described by a weighted *incoherent* superposition of the pure state response characteristics. If the efficiency for detecting the pure state $|\phi_n\rangle$ is denoted by ϵ_n then the result of a measurement with the ensemble in an arbitrary state is

$$W = \sum_n \epsilon_n P(\phi_n) = \sum_n \epsilon_n \langle \phi_n | \rho | \phi_n \rangle \quad (1.30)$$

or

$$W = \text{Tr}(\epsilon \rho) \quad (1.31)$$

where ϵ is the diagonal matrix with elements ϵ_n . By defining the efficiency operator

$$\epsilon_{op} = \sum_n |\phi_n\rangle \epsilon(n) \langle \phi_n| \quad (1.32)$$

so that the efficiency matrix is the efficiency operator represented in some convenient basis $|m\rangle$

$$\langle m | \epsilon | m' \rangle = \sum_n \langle m | \phi_n \rangle \epsilon(n) \langle \phi_n | m' \rangle \quad (1.33)$$

then it is possible to construct the efficiency tensor according to (1.15)

$$\epsilon_q^\lambda(j_1 j_2) = \sum_{m_1} \langle j_1 m_1 | \epsilon | j_2 m_2 \rangle (-1)^{j_2 + m_2} (2\lambda + 1)^{\frac{1}{2}} \begin{pmatrix} j_2 & j_1 & \lambda \\ -m_2 & m_1 & q \end{pmatrix}. \quad (1.34)$$

With these definitions it can be shown [4] that the result of a measurement with a measuring device characterized by efficiency tensor $\epsilon_q^\lambda(j_1 j_2)$ on an ensemble represented by the statistical tensor $\rho_q^\lambda(j_1 j_2)$ is given by

$$W = \sum_{j_1 j_2 \lambda q} \epsilon_q^{\lambda\dagger}(j_2 j_1) \rho_q^\lambda(j_2 j_1). \quad (1.35)$$

Often, as in the case of LTNO, measurements are only made on the emitted radiation implying the existence of, but giving no information on, a final nuclear state. In such cases a summation over the final unobserved states is performed and the measurement is represented by the unit efficiency matrix. If these states have angular momentum j_i then by analogy with (1.20) the corresponding efficiency tensor is

$$\epsilon_q^\lambda(j_1 j_2) = (2j_1 + 1)^{\frac{1}{2}} \delta_{j_1 j_2} \delta_{\lambda 0} \delta_{q 0}. \quad (1.36)$$

Finally, the efficiency matrix for the detection of emitted electromagnetic radiation in helicity states $|\mathbf{p}\tau\rangle$ is considered. Clearly the efficiency operator should be defined in the representation $|\mathbf{p}\kappa\rangle$. Thus

$$\epsilon_{op} = \sum_{\kappa} |\mathbf{p}\kappa\rangle Q(\kappa) \langle \mathbf{p}\kappa| \quad (1.37)$$

where $Q(\kappa)$ is the response of the detector to the plane wave state of pure polarization $\kappa = \pm 1$. The efficiency matrix is therefore

$$\langle \mathbf{p}\tau | \epsilon | \mathbf{p}\tau' \rangle = \sum_{\kappa} \langle \mathbf{p}\tau | \mathbf{p}\kappa \rangle Q(\kappa) \langle \mathbf{p}\kappa | \mathbf{p}\tau' \rangle \quad (1.38)$$

and so for a detector which responds only to either positive or negative helicity states

$$\epsilon(+)=\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \quad \text{or} \quad \epsilon(-)=\begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}. \quad (1.39)$$

1.2.5 The time evolution of the final state density matrix

An ensemble of nuclei η , which at the time $t = 0$ is described by the density matrix $\langle I_i m_i | \rho | I_i m'_i \rangle$, will ultimately decay into a final nuclear state I_f by the emission of radiation with a definite momentum $\mathbf{p}$. This final state is represented by the final state density matrix $\langle \mathbf{p}\tau I_f m_f | \rho | I_f m'_f \mathbf{p}\tau' \rangle$. This subsection will deal briefly with the derivation of this final state matrix. Starting with the time dependent Schrödinger equation

$$i \frac{\partial}{\partial t} |\Psi\rangle = H |\Psi\rangle \quad (1.40)$$

whose general solution may be written

$$|\Psi(t)\rangle = U(t, t_0) |\Psi(t_0)\rangle \quad (1.41)$$

where $U(t, t_0)$ is the unitary time-evolution operator, then the time behaviour of the density operator is given by

$$\rho_{op}(t) = \sum_n U(t, t_0) |\Psi_n(t_0)\rangle g(n) \langle \Psi_n(t_0)| U^\dagger(t, t_0) = U(t, t_0) \rho_{op}(t_0) U^\dagger(t, t_0). \quad (1.42)$$

From (1.40) and (1.41) it is clear that $U(t, t_0)$ must also satisfy the Schrödinger equation

$$i \frac{\partial}{\partial t} U(t, t_0) = H U(t, t_0). \quad (1.43)$$

The total Hamiltonian of the system is

$$H = H_0 + H_{int} \quad (1.44)$$

where H_0 is that part of the Hamiltonian which gives rise to the stationary states and H_{int} induces transitions between the eigenstates of H_0 . Explicitly, for the case of electromagnetic radiation [5]

$$H_{int} = - \int \mathbf{j}(\mathbf{r}') \mathbf{A}(\mathbf{r}', t) dv'. \quad (1.45)$$

Performing the unitary transformation to the Heisenberg representation leads to the operators

$$H_H(t) = e^{(\frac{iH_0 t}{\hbar})} H_{int} e^{(-\frac{iH_0 t}{\hbar})} \quad (1.46)$$

and

$$U_H(t, t_0) = e^{(\frac{iH_0 t}{\hbar})} U(t, t_0) e^{(-\frac{iH_0 t}{\hbar})}. \quad (1.47)$$

The expression corresponding to (1.43) in this representation may be solved to give to first order

$$U_H(t, t_0) = 1 - i \int_{t_0}^t H_H(t') dt' + \dots \quad (1.48)$$

Combining the second term in this expansion with the transformed analogue of (1.42) and using first-order perturbation theory leads to

$$\begin{aligned} \langle \mathbf{p} \tau I_f m_f | \rho(t) | I_f m'_f \mathbf{p} \tau' \rangle &= \langle \mathbf{p} \tau I_f m_f | e^{(-\frac{iH_0 t}{\hbar})} \left\{ \int_0^t e^{(\frac{iH_0 t'}{\hbar})} H_{int} e^{(-\frac{iH_0 t'}{\hbar})} dt' \right\} \\ &\times \rho(0) \left\{ \int_0^t e^{(\frac{iH_0 t'}{\hbar})} H_{int}^\dagger e^{(-\frac{iH_0 t'}{\hbar})} dt' \right\} e^{(\frac{iH_0 t}{\hbar})} | I_f m'_f \mathbf{p} \tau' \rangle. \end{aligned} \quad (1.49)$$

The statevector

$$e^{(\frac{iH_0 t}{\hbar})}|I_f m'_f \mathbf{p} \tau'\rangle = |I_f m'_f \mathbf{p} \tau'\rangle_H \quad (1.50)$$

is the time independent eigenvector of H_0 in the Heisenberg representation. Since the initial grand density matrix $\rho(0)$ has only the elements $\langle I_i m_i | \rho(0) | I_i m'_i \rangle$, then (1.49) can be written

$$\begin{aligned} \langle \mathbf{p} \tau I_f m_f | \rho(t) | I_f m'_f \mathbf{p} \tau' \rangle &= \langle \mathbf{p} \tau I_f m_f | H_{int} | I_i m_i \rangle \langle I_i m_i | \rho(0) | I_i m'_i \rangle \\ &\times \langle I_i m'_i | H_{int}^\dagger | I_f m'_f \mathbf{p} \tau' \rangle \left| \int_0^t e^{i(\omega + \frac{E_f - E_i}{\hbar})t'} dt' \right|^2 \end{aligned} \quad (1.51)$$

where the arguments of the exponential arise from the action of $e^{(\frac{iH_0 t}{\hbar})}$ on $\langle \mathbf{p} \tau |$ and $\langle I_f m_f |$ and of $e^{(-\frac{iH_0 t}{\hbar})}$ on $|I_i m_i \rangle$ respectively. In accordance with the conservation of energy the integral is only appreciably different from zero if $\hbar\omega = \hbar\omega_0 = E_i - E_f$. In the Heisenberg representation the transition matrix elements of (1.51) are time independent

$$\begin{aligned} \langle \mathbf{p} \tau I_f m_f | H_{int} | I_i m_i \rangle &= \int \langle I_f m_f | \mathbf{j}(\mathbf{r}') | I_i m_i \rangle \mathbf{A}_{\mathbf{p}\tau}(\mathbf{r}') dv' \\ &= \sqrt{\frac{2\pi}{L^3\omega}} \langle I_f m_f | \int \mathbf{j}(\mathbf{r}') \mathbf{e}_\tau^* e^{-i\frac{\mathbf{p}\mathbf{r}'}{\hbar}} dv' | I_i m_i \rangle \end{aligned} \quad (1.52)$$

since the time dependence of $\mathbf{j}(\mathbf{r}', t)$, namely $e^{\frac{i(E_f - E_i)}{\hbar}t}$, is exactly compensated by that of $\mathbf{A}(\mathbf{r}, t)$, namely $e^{i\omega t}$. By integrating over ω and multiplying by the density of states, the last term in (1.51) results in a factor $L^3\omega_0^2 t d\Omega / (2\pi)^2$. This is correct for values of t much smaller than the mean life time $1/\lambda_\gamma$, where λ_γ is the total transition probability. For times larger than this, t should be replaced by the radioactive decay factor $(1 - e^{-\lambda_\gamma t})/\lambda_\gamma$. Thus the elements of the final state density matrix are

$$\begin{aligned} \langle \mathbf{p} \tau I_f m_f | \rho(t) | I_f m'_f \mathbf{p} \tau' \rangle &= \frac{d\Omega}{2\pi} \omega \frac{(1 - e^{-\lambda_\gamma t})}{\lambda_\gamma} \langle I_f m_f | \int \mathbf{j}(\mathbf{r}') \mathbf{e}_\tau^* e^{-i\frac{\mathbf{p}\mathbf{r}'}{\hbar}} dv' | I_i m_i \rangle \\ &\times \langle I_i m_i | \rho(0) | I_i m'_i \rangle \langle I_i m'_i | \int \mathbf{j}(\mathbf{r}') \mathbf{e}_\tau e^{i\frac{\mathbf{p}\mathbf{r}'}{\hbar}} dv' | I_f m'_f \rangle. \end{aligned} \quad (1.53)$$

Introducing the absolute transition amplitudes $\gamma(\pi L, I_i \rightarrow I_f)$ [4] which are proportional to the multipole expansions of (1.52) and again using the Clebsh-Gordan series for the product of two D -matrices gives

$$\begin{aligned}
 \langle \mathbf{p} \tau I_f m_f | \rho(\infty) | I_f m'_f \mathbf{p} \tau' \rangle &= \frac{d\Omega}{8\pi\lambda_\gamma} \sum_{m, m'_i} (2I_i + 1) \sum_{LL'\lambda} [(2L + 1)(2L' + 1)]^{\frac{1}{2}} \\
 &\times (2\lambda + 1) \begin{pmatrix} I_f & L & I_i \\ m_f & M & -m_i \end{pmatrix} \begin{pmatrix} I_f & L' & I_i \\ m'_f & M' & -m'_i \end{pmatrix} \\
 &\times \begin{pmatrix} L & L' & \lambda \\ M & -M' & q \end{pmatrix} \begin{pmatrix} L & L' & \lambda \\ \tau & -\tau' & \mu \end{pmatrix} \langle I_i m_i | \rho(0) | I_i m'_i \rangle \\
 &\times [\gamma(EL) + \tau\gamma(ML)][\gamma^*(EL') + \tau'\gamma^*(ML')] D_{q\mu}^{\lambda*}(\mathbf{e}_z \rightarrow \mathbf{k})
 \end{aligned} \tag{1.54}$$

for times $\lambda_\gamma t \sim \infty$. The diagonal elements of this final state density matrix expression give the absolute probability that a photon with helicity τ is emitted into the solid angle $d\Omega$ in the direction $\mathbf{p}$ from the initial state $\rho(0)$ to form the final nuclear state $|I_f m_f\rangle$.

From this expression the final state statistical tensor $\rho_{qf}^{\lambda_f}(I_f, \mathbf{p})$ may be constructed in accordance with (1.15). This is a 2×2 matrix in the helicity space τ , whose four elements in $|Im\rangle$ space are

$$\begin{aligned}
 \langle \tau | \rho_{qf}^{\lambda_f}(I_f, \mathbf{p}) | \tau' \rangle &= \frac{d\Omega}{8\pi\lambda_\gamma} \sum_{\lambda, \lambda_{q_i}, q, LL'} (-1)^{\lambda_i - q_i} (2\lambda + 1)^{\frac{1}{2}} \left(\frac{2I_i + 1}{2I_f + 1} \right)^{\frac{1}{2}} \\
 &\times \begin{pmatrix} \lambda_f & \lambda & \lambda_i \\ q_f & q & -q_i \end{pmatrix} \rho_{q_i}^{\lambda_i}(I_i) D_{q\mu}^{\lambda*}(\mathbf{e}_z \rightarrow \mathbf{k}) [\gamma(EL) + \tau\gamma(ML)] \\
 &\times [\gamma^*(EL') + \tau'\gamma^*(ML')] \frac{\begin{pmatrix} L & L' & \lambda \\ \tau & -\tau' & \mu \end{pmatrix}}{\begin{pmatrix} L & L' & \lambda \\ 1 & -1 & 0 \end{pmatrix}} F_{\lambda}^{\lambda_f \lambda_i}(LL' I_f I_i)
 \end{aligned} \tag{1.55}$$

where the $F_{\lambda}^{\lambda_f \lambda_i}(LL' I_f I_i)$ are the generalized F -coefficients defined by

$$\begin{aligned}
 F_{\lambda}^{\lambda_f \lambda_i}(LL' I_1 I_0) &= (-1)^{L' + \lambda_1 + \lambda_0 + 1} [(2I_0 + 1)(2I_1 + 1)(2L + 1)(2L' + 1) \\
 &\times (2\lambda_0 + 1)(2\lambda_1 + 1)(2\lambda + 1)]^{\frac{1}{2}} \begin{pmatrix} L & L' & \lambda \\ 1 & -1 & 0 \end{pmatrix} \begin{Bmatrix} I_1 & L & I_0 \\ I_1 & L' & I_0 \\ \lambda_1 & \lambda & \lambda_0 \end{Bmatrix}.
 \end{aligned} \tag{1.56}$$

If one of the oriented states I_0 or I_1 is random, so that either λ_0 or λ_1 is zero, then the generalized F -coefficients reduce to the ordinary F -coefficients $F_{\lambda}(LL' I_1 I_0)$.

1.2.6 The angular distribution of gamma radiation from LTNO sources

Into (1.55), which is perfectly general being derived only from the assumption of the invariance of interactions under three dimensional rotations, the conditions specific to nuclear orientation problems are now introduced. Firstly, the statistical tensor of the initial state can be replaced by the orientation parameters (1.24). Secondly, the final nuclear state is *not* observed and so its efficiency tensor is given by (1.36). Only the radiation quanta are observed, in the direction $\mathbf{k}$ using a detector described by the efficiency matrix ϵ . Again it will be assumed that the observed quanta are of electromagnetic origin, although the formalism applies also to alpha and beta radiation. Thus using (1.35) the probability of detecting the photon is

$$W(\mathbf{k}) = \sum_{\tau\tau'} \langle \tau | \rho_0^0(I_f) | \tau' \rangle \langle \tau' | \epsilon | \tau \rangle (2I_f + 1)^{\frac{1}{2}} \quad (1.57)$$

where $\langle \tau | \rho_0^0(I_f) | \tau' \rangle$ is given by (1.55).

Considering the detection of circularly polarized radiation with helicity τ by an ideal detector with efficiency matrix (1.39), then $\tau = \tau'$ and so

$$W(\theta, \tau) = \frac{d\Omega}{8\pi\lambda_\gamma} \sum_{\lambda q LL'} \tau^\lambda \tau^{L+L'} B_\lambda(I_i) F_\lambda(LL' I_f I_i) \times [\gamma(EL) + \tau\gamma(ML)][\gamma^*(EL') + \tau\gamma^*(ML')] P_\lambda(\cos\theta) \quad (1.58)$$

where use has been made of the fact that [2]

$$D_{00}^{\lambda*}(\phi, \theta, 0) = P_\lambda(\cos\theta). \quad (1.59)$$

The angles ϕ, θ are the polar angles of the observation direction $\mathbf{k}$ with respect to coordinate system xyz in which the initial state statistical tensor is represented. With the inclusion of the parity selection rules for electromagnetic transitions: $L + L'$ even (odd) if both components are electric or magnetic (one component electric while the other magnetic), the *circular polarization distribution* is obtained as

$$W(\theta, \tau) = \frac{d\Omega}{8\pi} \sum_{\lambda} \tau^\lambda B_\lambda(I_i) A_\lambda P_\lambda(\cos\theta) \quad (1.60)$$

where the *angular distribution coefficients*, A_λ , are given by

$$A_\lambda = \sum_{L\pi L'\pi'} F_\lambda(LL' I_f I_i) \gamma(\pi L) \gamma^*(\pi' L') / \sum_{L\pi} |\gamma(\pi L)|^2 \quad (1.61)$$

with the normalization $A_0 = 1$; and by definition

$$\lambda_\gamma = \sum_{L\pi} |\gamma(\pi L)|^2. \quad (1.62)$$

For two mixed multipoles L and $L' = L + 1$ with amplitude mixing ratio $\delta = \gamma(\pi' L + 1)/\gamma(\pi L)$ the angular distribution coefficients can be written in the form

$$A_\lambda = \{F_\lambda(LLI_f I_i) + 2\delta F_\lambda(LL + 1I_f I_i) + \delta^2 F_\lambda(L + 1L + 1I_f I_i)\} / \{1 + \delta^2\}. \quad (1.63)$$

Figure 1.2 shows an example of the δ dependence of the angular distribution coefficients.

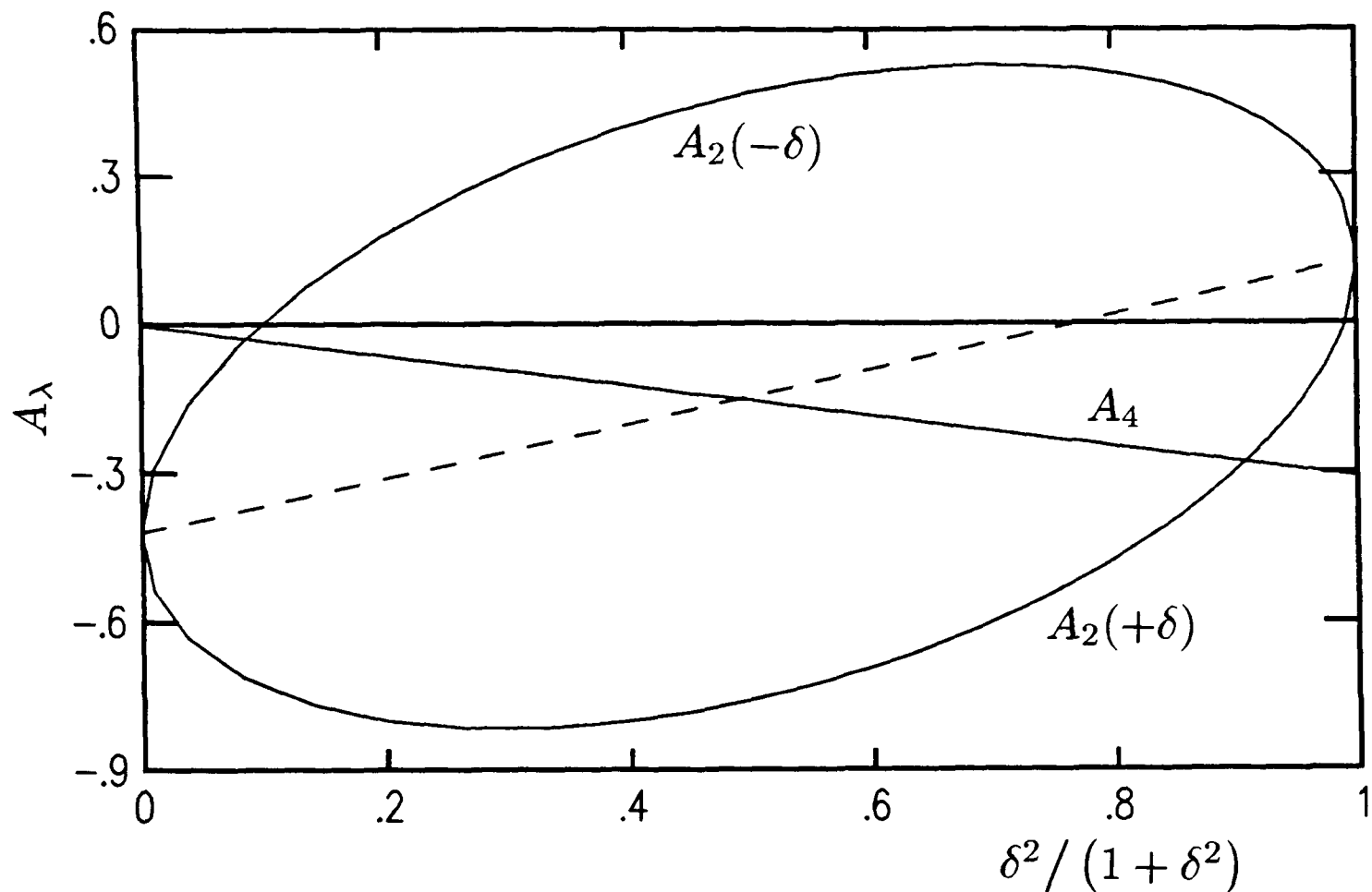


Figure 1.2. The non-zero angular distribution coefficients, A_λ , as a function of δ for gamma radiation in a mixed $2 \rightarrow 2$ transition.

The *directional distribution* of gamma radiation is now obtained by summing (1.60) over both helicity states which yields a factor of 2(0) if λ is even (odd). Thus

$$W(\theta) = \frac{d\Omega}{4\pi} \sum_{\lambda=\text{even}} B_\lambda(I_i) A_\lambda P_\lambda(\cos \theta). \quad (1.64)$$

1.2.7 Unobserved intermediate radiations

Generally in LTNO measurements the detected gamma radiation quantum will be preceded by a cascade of unobserved intermediate radiation. In order to construct the final state statistical tensor, the procedure outlined in subsection 1.2.5 must be performed in turn for each emitted photon. Since only the last of these is observed it is necessary to integrate the final state statistical tensor over solid angle for each unobserved quantum. In this way it can be shown [4] that the final state statistical tensor $\rho_{q_n}^{\lambda_n}(I_n)$ that describes the ensemble after the emission of an unobserved gamma radiation γ_n from an initial state $\rho_{q_{n-1}}^{\lambda_{n-1}}(I_{n-1})$ is obtained from the relation

$$\rho_{q_n}^{\lambda_n}(I_n) = U_{\lambda_n}(\gamma_n) \rho_{q_{n-1}}^{\lambda_{n-1}}(I_{n-1}) \delta_{\lambda_n \lambda_{n-1}} \delta_{q_n q_{n-1}} \quad (1.65)$$

where the *de-orientation coefficients*, U_λ , have been introduced which are given by

$$U_\lambda = \sum_{L\pi} U_\lambda(I_i I_f L) |\gamma(\pi L)|^2 / \sum_{L\pi} |\gamma(\pi L)|^2 \quad (1.66)$$

or in terms of δ

$$U_\lambda = \{U_\lambda(I_i I_f L) + \delta^2 U_\lambda(I_i I_f L + 1)\} / \{1 + \delta^2\} \quad (1.67)$$

with the *de-orientation factor* defined by

$$U_\lambda(I_i I_f L) = (-1)^{I_i + I_f + \lambda + L} [(2I_i + 1)(2I_f + 1)]^{\frac{1}{2}} \begin{Bmatrix} I_i & I_i & \lambda \\ I_f & I_f & L \end{Bmatrix}. \quad (1.68)$$

The de-orientation factors are normalized to unity: $U_0 = 1$. Hence, for a cascade of n unobserved photons the final state statistical tensor is

$$\rho_q^\lambda(I) = U_\lambda(\gamma_n) U_\lambda(\gamma_{n-1}) \cdots U_\lambda(\gamma_1) \rho_q^\lambda(I_0). \quad (1.69)$$

In particular, the directional distribution of a gamma radiation γ_{n+1} that is emitted from a state I_n , formed by the emission of n unobserved quanta from an axially oriented state I_0 , is

$$\begin{aligned} W(\theta) &= \sum_{\lambda=\text{even}} B_\lambda(I_0) U_\lambda(\gamma_1) U_\lambda(\gamma_2) \cdots U_\lambda(\gamma_n) A_\lambda(\gamma_{n+1}) P_\lambda(\cos \theta) \\ &= \sum_{\lambda=\text{even}} B_\lambda(I_0) U_\lambda^{\text{cascade}} A_\lambda P_\lambda(\cos \theta). \end{aligned} \quad (1.70)$$

where the solid angle factor has been dropped for simplicity. If several different cascades are possible between the oriented level and the observed radiation then an effective de-orientation

coefficient is obtained as the average of the $U_{\lambda}^{cascade}$, weighted by the appropriate branching intensity (which should be normalized to unity).

The formalism of this subsection also applies without modification to alpha, beta and electron capture decay. In the LTNO experiments to be described in this thesis, the gamma cascade resulting in the observed photon is preceded by the lepton quantum which initiates the nuclear decay. In this case the term $U_{\lambda}(\gamma_1)$ in (1.70) refers to the lepton field rather than to the electromagnetic field. The appropriate leptonic de-orientation factor is calculated from (1.66) with $\gamma(L)$ understood to be the amplitude of the L^{th} multipole component in the lepton field and L the total angular momentum carried away by both the electron and neutrino.

In as much as the observed directional distribution in LTNO is dependent upon nuclear factors alone, (1.70) represents the final expression for its angular dependence and in principle is an infinite series. However the tensor nature of its coefficients, manifest in their dependence upon the n - j symbols [2], imposes limitations upon its maximum rank λ . By inspection these can be seen to be:

$$\begin{aligned} B_{\lambda}(I_0) &\equiv 0 \quad \text{if} \quad \lambda > 2I_0 \\ U_{\lambda}(LI_i I_f) &\equiv 0 \quad \text{if} \quad \lambda > 2I_i \quad \text{or} \quad \lambda > 2I_f \\ A_{\lambda}(LL' I_i I_f) &\equiv 0 \quad \text{if} \quad \lambda > L + L' \quad \text{or} \quad \lambda > 2I_i. \end{aligned} \tag{1.71}$$

Since the gamma transition probability greatly reduces as the multipole order increments, transitions with $L > 2$ are experimentally rare. Thus in the majority of cases and in the absence of further selection rules the directional distribution series may be truncated to

$$W(\theta) = 1 + B_2 U_2 A_2 P_2(\cos \theta) + B_4 U_4 A_4 P_4(\cos \theta) \tag{1.72}$$

where use has been made of the fact that the zero rank coefficients are normalized to unity.

1.3 Non-nuclear contributions to the angular distribution in LTNO

In the derivations of the previous section only the “nuclear” contributions to the angular distribution have been considered explicitly. Attention will now be given to those aspects of the LTNO system which might be termed “non-nuclear” in origin.

1.3.1 The solid angle correction factors

The directional distribution of (1.70) applies only to detection of nuclear radiation at a single angle θ . Owing to the finite solid angle subtended by the detector, the observed angular distribution will differ somewhat from this ideal distribution expected for “point” detectors. The proper interpretation of precision angular distribution measurements thus depends on the accurate knowledge of this difference.

To perform this correction it is necessary to integrate (1.70), weighted by the detector efficiency, over the subtended solid angle. Consider, therefore, the case of an axially symmetric detector centered along the radius vector with polar angle θ_0 . Using this radius vector as the initial line the polar angles β , such that $\theta = \theta_0 + \beta$ and $\Delta\beta$, such that the detector subtends a right cone of semi-angle $\Delta\beta$ at the origin, may be defined. In this co-ordinate system the useful absorption of gamma radiation incident onto the detector is proportional to

$$\left\{1 - e^{-\tau(\gamma)x(\beta)}\right\} \quad (1.73)$$

where $\tau(\gamma)$ is the full energy absorption coefficient and $x(\beta)$ is the path length through the active volume of the detector.

The measured directional distribution will therefore be given by

$$\overline{W(\theta)} = \frac{\int W(\theta) \left\{1 - e^{-\tau(\gamma)x(\beta)}\right\} d\Omega}{\int \left\{1 - e^{-\tau(\gamma)x(\beta)}\right\} d\Omega}. \quad (1.74)$$

Separating out the angular terms from this expression it is seen that the quantities of interest are the integrals

$$I_\lambda = \int P_\lambda(\cos(\theta_0 + \beta)) \left\{1 - e^{-\tau(\gamma)x(\beta)}\right\} \sin \beta d\beta d\phi. \quad (1.75)$$

Following the method of Rose ^[6] the spherical harmonic addition theorem

$$P_\lambda(\cos(\theta_0 + \beta)) = P_\lambda(\cos \theta_0)P_\lambda(\cos \beta) + \dots \quad (1.76)$$

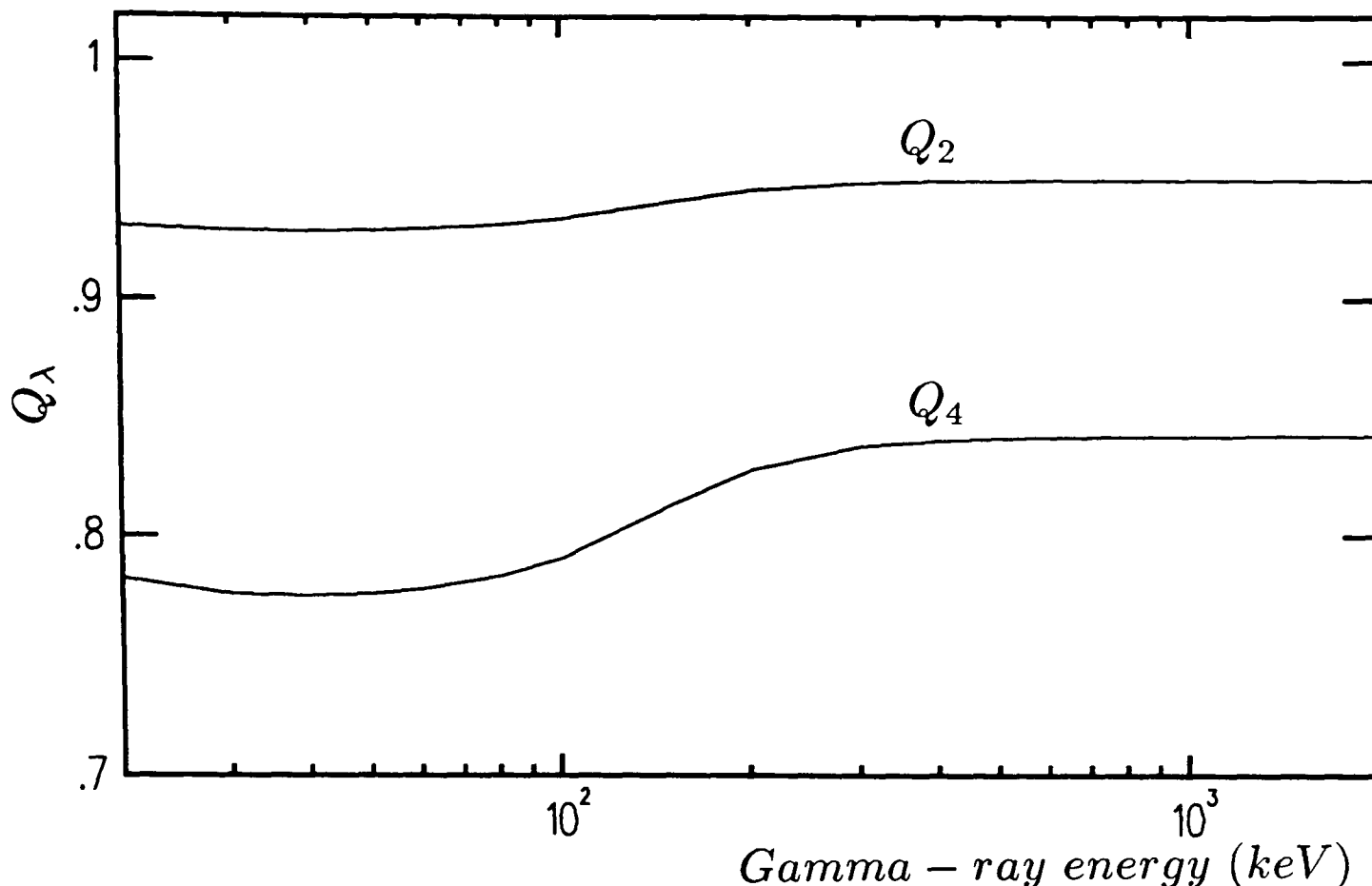


Figure 1.3. The dependence of the solid angle correction factors, Q_λ , on gamma ray energy for a Ge(Li) detector of typical dimensions at 8 cm from the source.

is applied, where the ellipses indicate azimuthal terms. These do not contribute to the integration over ϕ and so (1.75) reduces to

$$I_\lambda = 2\pi P_\lambda(\cos \theta_0) \int_0^{\Delta\beta} P_\lambda(\cos \beta) \left\{ 1 - e^{-\tau(\gamma)x(\beta)} \right\} \sin \beta d\beta. \quad (1.77)$$

Introducing the *solid angle correction factors*, Q_λ , defined by

$$Q_\lambda P_\lambda(\cos \theta_0) = \frac{I_\lambda}{I_0} \quad (1.78)$$

enables the corrected directional distribution (1.74) to be written in the form

$$\overline{W(\theta)} = \sum_{\lambda} B_\lambda U_\lambda A_\lambda Q_\lambda P_\lambda(\cos \theta_0). \quad (1.79)$$

The evaluation of the integrals I_λ requires a knowledge of both the detector geometry and the functional form of absorption coefficients $\tau(\gamma)$. These are calculated from the theoretical photoelectric, Compton and pair production cross sections of the detecting material using Monte Carlo simulations [7]. By this method, correction factors have been calculated by Yates [8] for Na(I) detectors and by Camp and Van Lehn [9] and Krane [10] for Ge(Li) detectors. The correction factors used in the present work are calculated using the method of Krane.

Typical dependences of the solid angle correction factors on gamma ray energy and detector to source distance are illustrated in figures 1.3 and 1.4.

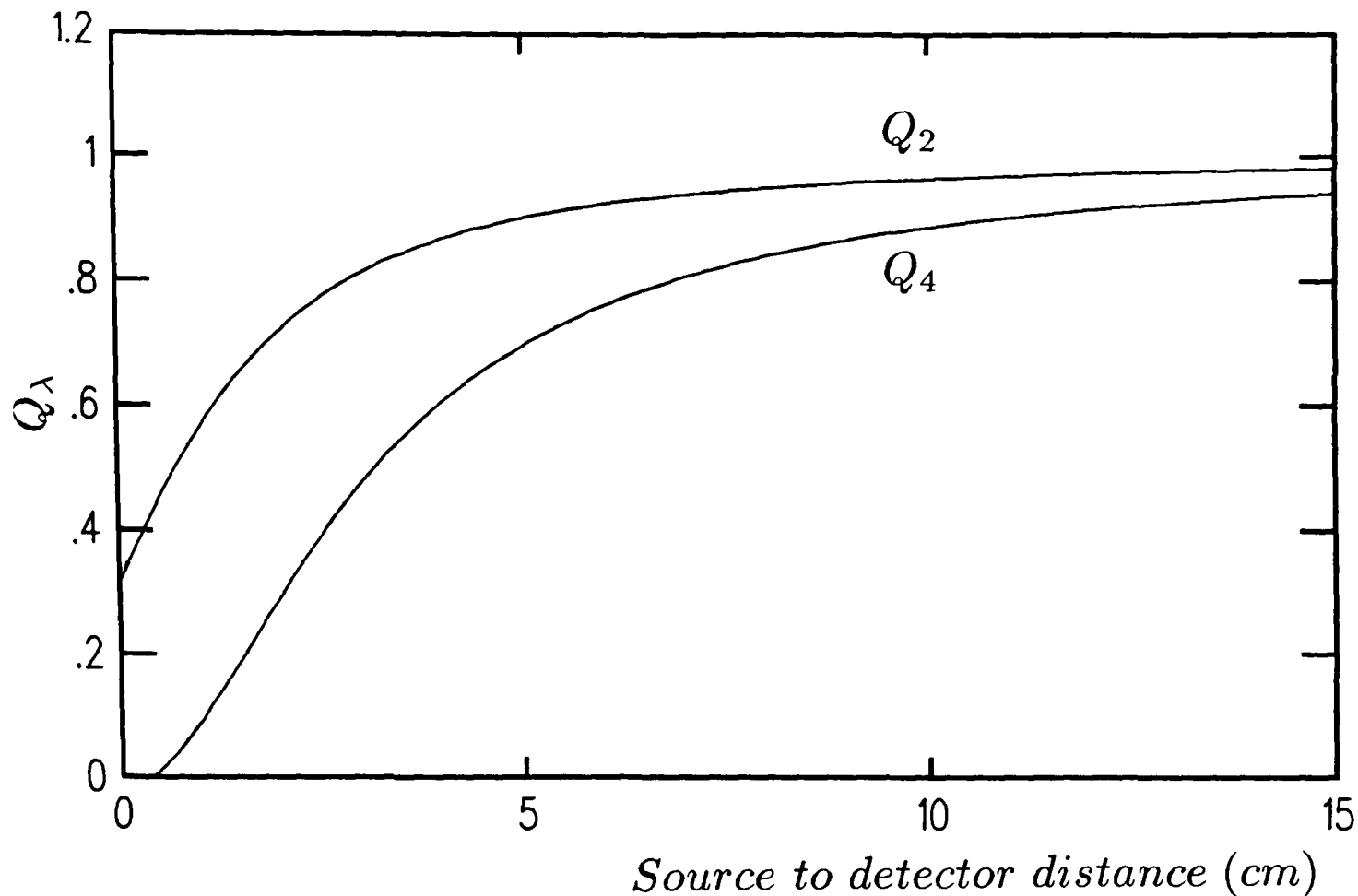


Figure 1.4. The dependence of the solid angle correction factors, Q_λ , on source to detector distance for a Ge(Li) detector of typical dimensions at a gamma ray energy of 500 keV.

1.3.2 The magnetic hyperfine interaction

In subsection 1.2.3 the orienting interaction was considered to be that between the static nuclear magnetic (electric) moment with an externally applied magnetic field (electric field gradient). In the magnetic case, the degree of nuclear orientation is determined largely by the parameter β of (1.27). From (1.26) it can be seen that an appreciable alignment is characterized by $\beta \sim 1$. For a nucleus with a gyromagnetic ratio of unity and taking 5 mK as a lower limit to the continuously attainable temperature of the present generation of $^3\text{He} - ^4\text{He}$ dilution refrigerators, then a magnetic field of at least 15 T would be required for successful orientation. The attainment of such a high field whilst simultaneously maintaining low temperatures is not readily achieved. Thus the “brute force” method, although simple in principle and applicable to all types of nuclei, is not always practical.

Most of the successful orientation experiments have made use of the hyperfine interaction in solids to avoid the need for such high values of B_{app}/T . All experiments described in this thesis have been performed on atomic nuclei implanted into a polycrystalline iron host. The origins of the magnetic hyperfine field ^[11] will now be briefly discussed.

Considering first the case of a free atom, an electronic current density $\mathbf{J}_e d\mathbf{r}$ at a point $\mathbf{r}$ in a coordinate system centered at the nucleus will produce a magnetic field at the nucleus

$$\mathbf{B} = \frac{\mu_0}{4\pi} \frac{\mathbf{r} \times \mathbf{J}_e}{r^3} d\tau. \quad (1.80)$$

Writing $\mathbf{J}_e d\tau = \mathbf{v} dq$, where $\mathbf{v}$ is the velocity of the charge element dq and noting that the electronic angular momentum is $\mathbf{l} = m_e \mathbf{r} \times \mathbf{v}$, then for each electron i

$$\mathbf{B}_{l_i} = -\frac{\mu_0}{4\pi} 2\mu_B \mathbf{l}_i \langle r_{l_i}^{-3} \rangle. \quad (1.81)$$

Performing the summation over i , the contributions from closed subshells disappear and in the LS coupling limit

$$\mathbf{B}_L = -\frac{\mu_0}{4\pi} 2\mu_B \mathbf{L} \langle r_L^{-3} \rangle. \quad (1.82)$$

Similarly the electron spin will also give rise to a magnetic field, but since there is now a finite probability of electrons with non-zero spin existing at the nucleus it is necessary to consider separately the contributions from electrons “inside” and “outside” of the nucleus. Electrons outside the nucleus give rise to a simple dipole sum field

$$\mathbf{B}_s = -\frac{\mu_0}{4\pi} g_s \mu_B \sum_i [3(\mathbf{s}_i \cdot \hat{\mathbf{r}}) \hat{\mathbf{r}} - \mathbf{s}_i] \langle r_{si}^{-3} \rangle \quad (1.83)$$

while those inside give rise to the Fermi contact term, which can be written in the LS coupling limit as

$$\mathbf{B}_F = -\frac{\mu_0}{4\pi} \frac{8\pi}{3} g_s \mu_B |\psi_s(0)|^2 \mathbf{S}. \quad (1.84)$$

In the solid state there are two mechanisms which give rise to such a contact field. In a ferromagnet the s - d exchange interaction between the magnetic $3d$ -electrons and the s -like conduction electrons removes the degeneracy of the $s^\uparrow$ and $s^\downarrow$ conduction bands. This leads to a net surplus of spin-up electrons and consequently a “conduction electron polarization” Fermi contact term of the form

$$\mathbf{B}_F = -\frac{\mu_0}{4\pi} \frac{8\pi}{3} g_s \mu_B \{ |\psi_s^\uparrow(0)|^2 - |\psi_s^\downarrow(0)|^2 \} \mathbf{S}. \quad (1.85)$$

Secondly, due to the overlap between the polarized conduction electrons of the magnetic host with the inner s -electrons of the non-magnetic impurity, it is possible for filled orbitals with spin parallel to the net spin to have a different density distribution from those same orbitals with antiparallel spin. The result of such exchange correlation effects, which for example can

be incorporated into Hartree-Fock calculations by relaxing the condition that the radial wavefunction is independent of the spin orientation, is that a closed s -subshell can exhibit a net spin without losing its spherical symmetry. These core s -electrons then contribute to the hyperfine field via an “exchange core polarization” contact term of the form (1.85). Such contact terms are important for all systems with non-zero resultant spin. However their significance is greatest for d -electron atoms for which in many cases the strong ligand field present in the lattice serves to quench the orbital contribution. Further, in a system of cubic symmetry such as iron the “outside” spin term vanishes.

These mechanisms give rise to the large magnetic fields, typically 10–1000 T for impurities in an iron host, required at the nucleus. It only remains to apply a modest external field B_{app} to polarize the host lattice and produce the axis of quantization. Fields of only ~ 0.7 T are sufficient to almost completely ($> 99\%$) align the domains in iron. B_{app} is subject to both the Knight shift and demagnetizing corrections. The former is usually small ($\sim 1\%$), while the latter is also small for a thin foil magnetized in its plane. Since also $B_{hyp} \gg B_{app}$, it is usually adequate to neglect these effects and take the effective magnetic field at the nucleus to be simply $|\mathbf{B}_{hyp} + \mathbf{B}_{app}|$.

The hyperfine interaction experienced by an impurity atom is of course highly sensitive to the nature of the surrounding lattice. It is not necessarily the case that the sites occupied by impurity atoms in the host lattice are unique and so they will be subject to a range of different hyperfine fields. An NMR experiment on oriented nuclei (NMR/ON) would in principle detect separately all of these different interactions. A static LTNO experiment on the other hand detects only the average hyperfine interaction for the system. If all the possible implantation lattice sites are labelled i , with associated magnetic hyperfine fields B_{hyp}^i and population probabilities f_i , then the observed directional distribution of gamma radiation in a static LTNO experiment will be

$$W(\theta) = \sum_{\lambda i} f_i B_{\lambda}(B_{hyp}^i) U_{\lambda} A_{\lambda} Q_{\lambda} P_{\lambda}(\cos \theta) \quad (1.86)$$

or in its truncated form

$$W(\theta) = 1 + \sum_i f_i \left\{ B_2(B_{hyp}^i) U_2 A_2 Q_2 P_2(\cos \theta) + B_4(B_{hyp}^i) U_4 A_4 Q_4 P_4(\cos \theta) \right\} \quad (1.87)$$

where use has been made of the fact that $\sum_i f_i = 1$. These are the final forms of the directional distribution expressions and will be used throughout this thesis.

Finally the magnetic hyperfine anomaly is discussed. This does not influence the form of the angular distribution formula, but is included here for the sake of completeness.

Once determined for one isotopic implant of a given element, the hyperfine field being atomic in its nature should be the same for all other members of that isotopic chain which are implanted under similar conditions. However, implicit in the Fermi contact term (1.84) and the lack of an “inside” orbital contribution is the assumption of a point nucleus. In reality the nucleus is an extended body implying that (1.84) should be integrated over the nuclear volume and that an “inside” orbital contribution should also be considered since electrons of non-zero orbital angular momentum exist within the nucleus. This provides a nucleus dependent perturbation to the hyperfine interaction energy $-\mu \cdot \mathbf{B}$, which can be considered as a nucleus dependent correction to an otherwise atomic hyperfine field. The hyperfine anomaly is related to the fractional change in the hyperfine interaction when the nuclear perturbation is “switched on” and typically being only $\sim 1\%$ is neglected in this thesis. Therefore in chapter 4 it is assumed that the magnetic hyperfine field as determined for one nucleus may be applied without correction to all other nuclei in the same isotopic chain.

1.3.3 The nature of the lattice site occupation

Clearly, the interpretation of static LTNO data can only proceed once the parameters B_{hyp}^i and f_i of (1.86) are known. In the bromine experiments described in chapter 4 samples were prepared by implanting the radioactive nuclei emerging from an isotope separator at energies of ~ 60 keV into the iron host matrix. Monte Carlo simulations ^[12] show that the entrant nucleus produces a series of primary lattice recoils which can go on to initiate independent subcascades. The resulting vacancies and interstitials typically cover a volume of order $(100 \text{ lattice spacing})^3$. The local temperatures produced in these cascades can reach 5000 K and last for about 10 ps ^[13] during which time a degree of self-annealing can occur. Frequently this leaves the implant in an undamaged area of the lattice, with residual vacancies and interstitials several atomic spacings away. Thus, provided that the implanted dose is not so great that the entire lattice is significantly damaged, it can be expected that a fair proportion of the implanted nuclei will come to rest in undamaged substitutional sites. This critical dose may be taken to be $\sim 10^{14}$ ions cm^{-2} or ~ 1 atomic percent. By comparison a typical LTNO dose is only $\sim 0.001 - 0.1$ atomic percent.

The actual fraction of nuclei which occupy substitutional sites and also the nature of any alternative sites depends on their “solubility” in the host lattice. For “soluble” implants it is

reasonable to make the simple assumption that the system contains only two sites, the substitutional site with magnetic hyperfine field B_{sub} and associated population f and a second site with zero field and population $1 - f$, corresponding to incompletely implanted activity on the surface of the lattice. Experimentally this is generally found to be a satisfactory assumption with values of f approaching 100%, especially where the lattice has been annealed after implantation. In this regime the truncated directional distribution is simply

$$W(\theta) = 1 + f \left\{ B_2(B_{sub}) U_2 A_2 Q_2 P_2(\cos \theta) + B_4(B_{sub}) U_4 A_4 Q_4 P_4(\cos \theta) \right\}. \quad (1.88)$$

For “insoluble” implants the situation is potentially very different. If the implanted ion is much smaller than the host then the lattice will contract locally and will therefore trap interstitials. In the reverse situation, implants will tend to accumulate vacancies. Thus, provided that the enthalpy of solution is sufficient to bind the interstitials or vacancies, the favoured sites will not necessarily be substitutional. This will be especially the case if the temperature is above 140 K or 200 K, the temperatures at which interstitials and vacancies respectively achieve full mobility in iron ^[14]. Thus it is expected that the proportion of insoluble implants in substitutional sites will be larger for cold implantation (< 4 K) than for room temperature implantation. In general therefore, insoluble implantations will exhibit a substitutional site and one or more sites associated with interstitials or vacancies, together with sites experiencing small or zero net hyperfine fields. In this complicated regime it is necessary to retain the general directional distribution expression (1.87).

This subject will be discussed further, with specific reference to bromine ions, in chapter 4.

1.4 Electric quadrupole orientation

In addition to the magnetic hyperfine field it is also possible to exploit the large electric field gradients in solids to achieve orientation and hence a measurement of the nuclear electric quadrupole moment ^[15]. This can only be done in a host lattice of non-cubic symmetry where the quadrupole interaction is non-vanishing. Also the requirement of a uniquely defined direction for the electric field gradient necessitates the use of a single crystal host.

Since iron is a cubic lattice the electric interaction disappears for implants occupying substitutional sites. For non-substitutional sites this symmetry is destroyed and a composite magnetic

and electric interaction must be considered. In a polycrystalline lattice the contribution to the total orientation arising from the electric quadrupole interaction is greatly reduced since the principal axis of each microcrystal is randomly directed in space. Nevertheless, the integrated effect of the individual electric interactions leads to an attenuation of the observed orientation when compared to that expected from the magnetic interaction alone. The magnitude of this attenuation depends upon the relative strengths of the magnetic and electric interactions as well as the temperature and oriented spin [16].

With regard to the experiments described in this thesis, all were performed on nuclei implanted into polycrystalline iron foils. In the case of the bromine isotopes studied in chapter 4 this resulted in a significant proportion of nuclei ($\sim 30\%$) coming to rest in non-substitutional lattice sites. However, the resulting electric quadrupole attenuations are expected to be small, typically a few per cent. On the other hand, the iridium sample used in the time reversal experiment discussed in chapter 8 was melted with iron leading to an almost complete occupation of substitutional sites. With full cubic symmetry thus maintained, the electric quadrupole interaction vanishes completely.

Since the role of the electric quadrupole interaction is either small or non existent it is not considered in this work.

1.5 Nuclear spin-lattice relaxation

To conclude this brief survey of the principles of LTNO the situation that arises when the assumption of thermodynamic equilibrium fails will be considered. In such cases it is necessary to include the time dependence of the approach to equilibrium into the calculation of the orientation parameters [17]. It will be assumed that the spin system is highly dilute so that impurities are coupled only to the host lattice and mutual interactions within the nuclear spin system are negligible. In this limit the nuclei relax independently and so cannot be described by a temperature. If this relaxation proceeds via an exchange of energy with the degenerate Fermi gas formed by the conduction electrons of the (metallic) host lattice then the spin-lattice interaction is given by the Hamiltonian

$$H_{S-L} = \mathbf{AI} \cdot \mathbf{S} = AI_z S_z + \frac{1}{2}(S_+ I_- + S_- I_+) \quad (1.89)$$

where $\mathbf{I}$ is the nuclear spin operator and $\mathbf{S}$ the effective lattice spin operator which is related to the orbital and spin operators of the conduction electrons. Transitions induced by the non-diagonal elements link states differing by one unit in the magnetic quantum number. The corresponding transition probabilities, derived from Fermi's golden rule, are

$$W_{m,m+1} = \frac{\Delta E}{2kC_k} \frac{[I(I+1) - m(m+1)]}{e^{\Delta E/kT} - 1} \quad (1.90)$$

and

$$W_{m+1,m} = W_{m,m+1} + \frac{\Delta E}{2kC_k} [I(I+1) - m(m+1)]. \quad (1.91)$$

Here ΔE is the magnetic substate energy splitting and C_k , the Korringa constant, is a system dependent constant arising from the integral over electron states. Under the conditions that the nuclear spin system is weakly coupled to the lattice and has a much smaller heat capacity, it can be shown that the spin density matrix remains diagonal at all times [18]. These diagonal elements, which correspond to the sublevel populations p_m , obey a gain-loss equation of the form

$$\frac{dp_m}{dt} = \sum_n (W_{nm}p_n - W_{mn}p_m) \quad (1.92)$$

or in matrix notation

$$\frac{d\mathbf{p}}{dt} = \mathbf{R}\mathbf{p} \quad (1.93)$$

where $\mathbf{p}$ is a $(2I+1)$ dimensional column vector and $\mathbf{R}$ is the time dependent relaxation matrix. Under the equivalence transformation $\mathbf{D}$, where $D_{mn} = \delta_{mn}e^{-E_m/2kT}$, the matrix $\mathbf{R}^S = \mathbf{D}^{-1}\mathbf{R}\mathbf{D}$ becomes symmetric. Hence (1.93) is solved by the diagonalization of $\mathbf{R}^S$ which leads to

$$\mathbf{p}(t) = \mathbf{D}\mathbf{U}\mathbf{e}^{\mathbf{K}t}\mathbf{U}^{-1}\mathbf{D}^{-1}\mathbf{p}(0) \quad (1.94)$$

where $\mathbf{U}$ and $\mathbf{K}$ are the matrices of the eigenvectors and eigenvalues of $\mathbf{R}^S$ respectively. Thus the time evolution of the populations is a multi-exponential function which is completely determined by the Korringa constant and the initial populations, $p_m(0)$.

Experimentally it is convenient to define a single effective time constant τ_{slr} to approximate the time dependence of (1.94) such that

$$\ln \left[\frac{B_2(t = \infty) - B_2(0)}{B_2(\infty) - B_2(\tau_{slr})} \right] = 1. \quad (1.95)$$

The initial populations $B_2(0)$ could be either semi-oriented, if they were produced by decay from an oriented precursor, or random. For the case of interest to the present work (see subsection 4.4.4) the parent state is produced directly by on-line implantation so that the latter assumption is appropriate and $B_2(0) = 0$. A simple empirical estimate for τ_{slr} can then be made from

$$\tau_{emp} = \min(\tau_{htl}, \tau_{ltl}) \quad (1.96)$$

where

$$\tau_{htl} = \frac{4}{3} \frac{C_k}{T} \quad \text{and} \quad \tau_{ltl} = \frac{3.3}{(I + \frac{1}{2})} \frac{C_k}{\beta T}. \quad (1.97)$$

The labels *htl* and *ltl* refer to the high and low temperature limits, as determined by the conditions $\beta \ll 1$ and $\beta \gg 1$ respectively. Apart from the region where the two limits overlap, i.e. where $\beta \sim 5/(2I+1)$, the agreement with τ_{slr} as deduced from an exact calculation of (1.94) is good. This is illustrated in figure 1.5 for the case of spin 5.

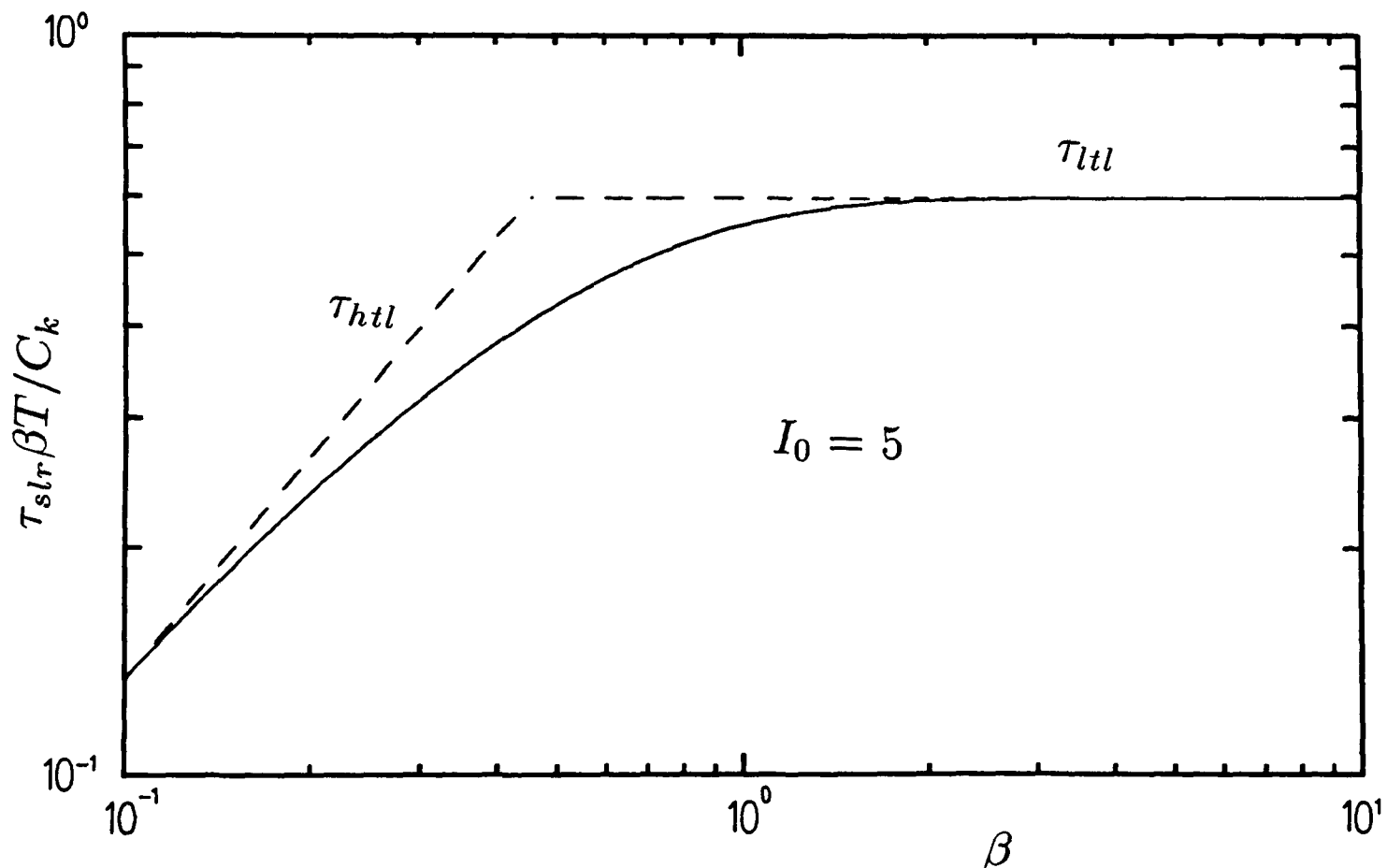


Figure 1.5. The ratio $\tau_{slr}\beta T/C_k$ as a function of β for an oriented state of spin 5. The dashed line represents τ_{emp} .

Provided that the relaxation proceeds via electron-hole pair excitations and magnetic dipole interactions (neglecting hyperfine anomalies) then it is possible to derive the scaling property

$$\left(\frac{\mu}{I}\right)^2 C_k = \text{constant} \quad (1.98)$$

for different isotopes of a given element in the same host. Thus C_k values and hence relaxation times may be estimated for a whole range of isotopes provided that at least one nucleus of the same element has been measured.

The significance of relaxation in LTNO comes when the radioactive decay is considered. If $\tau_{s,lr}$, which can vary from 10^{-3} to 10^4 s for impurities in iron, is significant in comparison with the nuclear lifetime then an appreciable number of nuclei decay from incompletely oriented states. This results in a temperature dependent reduction of the observed angular distribution anisotropy via an attenuation in the orientation parameters.

In the case of on-line LTNO the situation is further complicated by the steady stream of incoming warm nuclei. Sublevel populations representing the relaxing and populating states and allowances for the radioactive decay of the relaxing sublevels must then be included into (1.93). On the assumption that N nuclei are implanted per second and that these are equally distributed among the relaxing sublevels then the modified analogue of (1.92) may be written as

$$\frac{dp_m}{dt} = \sum_n (W_{nm}p_n - W_{mn}p_m) - \frac{\ln 2}{t_{\frac{1}{2}}} p_m + \frac{N}{(2I+1)}. \quad (1.99)$$

The sublevel populations will then attain secular equilibrium, given by the condition $dp/dt = 0$. From this the sublevel populations and hence the orientation parameters may be deduced.

Figure 1.6 shows the ratio $B_2(\text{secular})/B_2(\text{equilibrium})$ as a function of β for a range of $t_{\frac{1}{2}}\beta T/\ln 2C_k$ values. The latter expression is essentially a measure of the ratio of the nuclear lifetime to the relaxation time. It is clear that for $t_{\frac{1}{2}}\beta T/\ln 2C_k \ll 1$, $B_2/B_2(\text{eq}) \rightarrow 0$ since the nuclei decay from the unrelaxed warm state, while for $t_{\frac{1}{2}}\beta T/\ln 2C_k \gg 1$, $B_2/B_2(\text{eq}) \rightarrow 1$ full relaxation having occurred before decay.

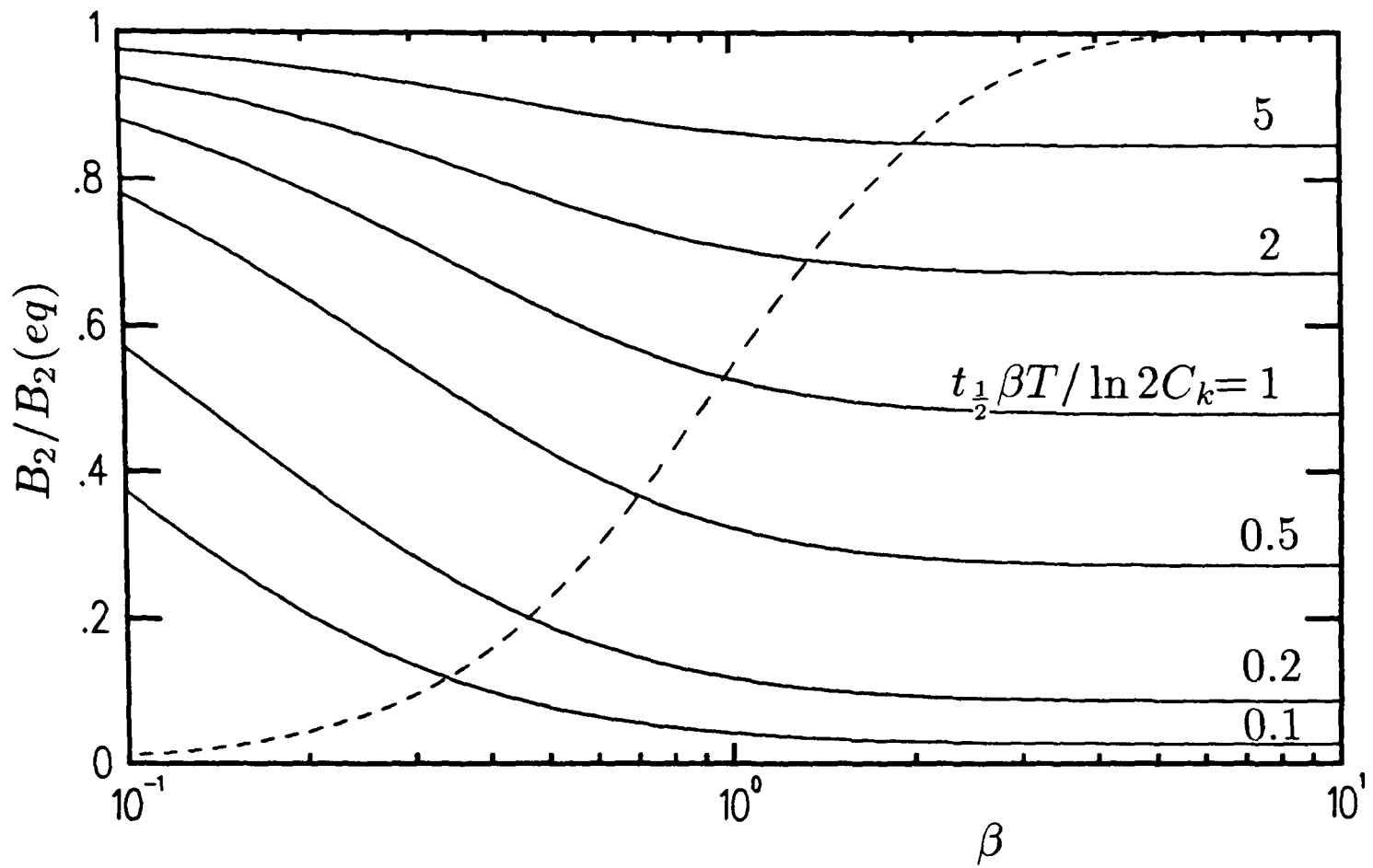


Figure 1.6. The ratio $B_2/B_2(eq)$ for a magnetically oriented state of spin 3 as a function of β for a range of values of $t_{\frac{1}{2}}\beta T / \ln 2C_k$. The dashed line represents $B_2(eq)$, normalized to its saturation value.

References

- [1] . U. Fano, Phys. Rev. **90** (1953) 577
- [2] . D.M. Brink and G.R. Satchler, Angular Momentum (Oxford, 1968)
- [3] . R.J. Blin-Stoyle and M.A. Grace, in Encycl. of Phys. Vol. XLII (Springer-Verlag, 1957)
- [4] . R.M. Steffen and K. Alder, in The Electromagnetic Interaction in Nuclear Spectroscopy, ed. W.D. Hamilton (North Holland, 1975)
- [5] . A. Messiah, Quantum Mechanics (North Holland, 1961)
- [6] . M.E. Rose, Phys. Rev. **C91** (1953) 610
- [7] . E. Storm and H.I. Israel, Los Alamos Scientific Laboratory Report LA-3753 (1967)
- [8] . M.J.L. Yates, in Alpha-, Beta- and Gamma-Ray Spectroscopy, ed. K. Siegbahn (North Holland, 1965)
- [9] . D.C. Camp and A.L. Van Lehn, Nucl. Instr. and Meth. **76** (1969) 192
- [10] . K.S. Krane, Nucl. Instr. and Meth. **98** (1972) 205
- [11] . N.J. Stone, in Low Temperature Nuclear Orientation, eds. N.J. Stone and H. Postma (North Holland, 1987)
- [12] . J.R. Beeler and M.F. Beeler, in Fundamental Aspects of Radiation Damage in Metals, CONF-751006-P1 (1976) 28
- [13] . R.S. Averbach *et al.*, Nucl. Instr. and Meth. **B33** (1988) 693
- [14] . A. Tuross *et al.*, Nucl. Instr. and Meth. **B19** (1987) 123
- [15] . E. Hagn, in Low Temperature Nuclear Orientation, eds. N.J. Stone and H. Postma (North Holland, 1987)
- [16] . R. Haroutunian and M. Meyer, Phys. Rev. **C17** (1978) 292
- [17] . E. Klein, in Low Temperature Nuclear Orientation, eds. N.J. Stone and H. Postma (North Holland, 1987)
- [18] . A. Sher and H. Primakoff, Phys. Rev. **119** (1960) 178

Chapter 2

Experimental apparatus and techniques

2.1 Introduction

The experiments described in this thesis have been performed either off-line, at the Oxford ^3He - ^4He dilution refrigerator used in conjunction with a PrNi_5 demagnetization stage, or at the Daresbury On-Line Isotope Separator-Cryogenic On-Line Device (DOLIS-COLD). The design and performance of these systems will now be outlined.

2.2 The ^3He - ^4He dilution refrigerator

For cooling significantly below 0.3 K the only practical method available until the early 1970's was adiabatic demagnetization of a suitable paramagnetic salt. This technique, however, is basically a single cycle method of cooling; after demagnetization the cryostat begins to warm up owing to the presence of heat leaks. For this reason the method is unsuitable for experiments requiring reasonably continuous low temperature operation in the presence of power sources.

After 1965 the first prototypes of the ^3He - ^4He dilution refrigerator were made operational following the principles originally suggested by London *et al.* ^[1]. These allowed the continuous cooling denied to the demagnetization techniques.

2.2.1 The ^3He - ^4He cooling process

The operation of the ^3He - ^4He dilution refrigerator relies on the special properties of the ^3He - ^4He mixture at low temperatures. At temperatures below the tricritical point (0.86 K) this mixture separates into two phases. One, the *concentrated* phase, is rich in ^3He (99.997 % pure at 0.1 K) while the other, the *dilute* phase, is rich in ^4He (7.0 % ^3He at 0.1 K). The principal of the dilution refrigerator depends on the fact that the proportion of ^3He in the dilute phase remains finite and rather large (6.4 %) even at absolute zero. Through the act of pumping on the dilute phase, ^3He is preferentially removed by virtue of its higher vapour pressure. In order to maintain the ^3He - ^4He equilibrium concentration, ^3He is drawn across the phase boundary from the concentrated phase. At temperatures below 0.5 K, ^4He is effectively a Bose-Einstein condensate and is both thermally and hydrodynamically inert, while ^3He can be regarded as a normal Fermi-Dirac liquid. This process can therefore be considered as the “evaporation” of ^3He atoms from the concentrated phase into the “vacuum” provided by the dilute phase, thus leading to cooling.

A full discussion of the properties of ^3He - ^4He mixtures is given by Lounasmaa [2].

2.2.2 The dilution refrigerator design

A schematic view of the main features of a conventional ^3He - ^4He *dilution unit* is shown in figure 2.1. In stable operation, the concentrated-dilute phase boundary lies in the *mixing chamber* which is the coldest part of the refrigerator. The lower half of the mixing chamber, which contains the dilute phase, is connected to the *still* via a series of heat exchangers. The still is pumped by a large diffusion pump backed by a sealed rotary pump, preferentially removing ^3He ($\sim 95\%$) despite its low ($\sim 7\%$) concentration in the dilute phase. As has already been noted, this leads to the migration of ^3He across the phase boundary and consequently cooling. For optimal circulation rates an electric heater maintains the still at ~ 0.8 K. The ^3He emerging from the rotary pump exhaust re-enters the cryostat via liquid nitrogen *cold traps* which remove any air that may have leaked into the system. Once inside the cryostat, the ^3He is pre-cooled to 1.2 K by contact with the pumped ^4He *pot* and then liquefied in the *condenser*. A flow impedance keeps the pressure of the ^3He sufficiently high for condensation to occur. The ^3He is further cooled to 0.8 K by contact with a wrap around heat exchanger on the still and then returns to the mixing chamber via a concentric tube continuous heat exchanger and four sintered silver step heat exchangers, where cooling takes place by contact with the out-going ^3He from

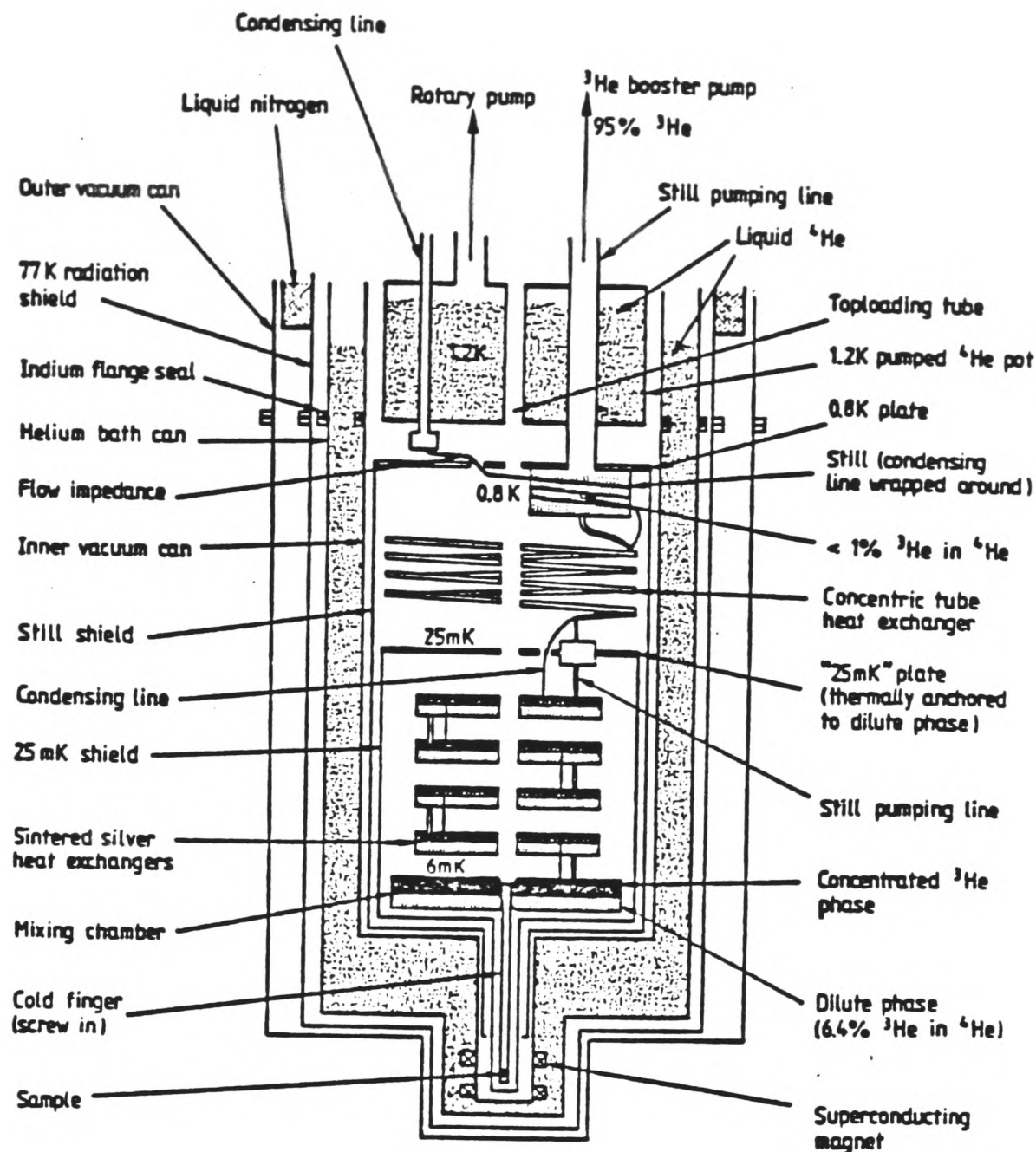


Figure 2.1. A schematic view of a conventional continuous operation ^3He - ^4He dilution refrigerator.

the pumped dilute phase. The efficiency with which the pre-cooling process is performed is one of the limitations on the minimum attainable temperature.

The conventional cryostat design external to the dilution unit consists of the Inner and Outer Vacuum Chambers (IVC and OVC; both evacuated to pressures of $< 10^{-6}\tau$) which enclose the main ^4He bath. Within the OVC lies a radiation shield maintained at 77 K by a liquid nitrogen cooled jacket. The IVC contains the dilution unit, the main helium bath on its outer face acting as a 4.2 K heat shield. Finally, in thermal contact with the dilution unit there is the 1.2 K ^4He *pot* (fed either continuously or intermittently by the main bath), the 0.8 K heat shield (attached to the still) and the 25 mK heat shield.

The experimental samples and thermometers are soldered to a copper cold finger screwed into the base of the mixing chamber.

2.2.3 The operation of the dilution unit

In attaining the base temperature in this continuous mode, a balance must be struck between the extra cooling induced by increasing the rate at which ^3He crosses the phase boundary and the consequent additional heating caused by the greater volume of returning gas and lower heat exchange efficiency at higher circulation rates. For the DOLIS-COLD refrigerator the optimum ^3He circulation rate is $\sim 400\mu\text{moles s}^{-1}$. With the 4 K baffle of the side access tube shut (see subsection 2.5.1) this can lead to a base temperature of $\sim 8\text{ mK}$.

Experimentally in LTNO it is important to be able to vary the temperature of the refrigerator in some manner. In principal this can be done by adjusting the power output of the still heater, thus changing the circulation rate. However this is a rather uncertain and unreliable method. Greater control is achieved by adding known quantities of heat directly into the mixing chamber via a thermally attached resistor. If this power input is denoted by Q then it can be shown [2] that

$$Q = C (T_{mc}^2 - T_{base}^2) \quad (2.1)$$

where T_{mc} is the actual temperature of the mixing chamber and T_{base} the temperature which would be achieved in the absence of Q . From a knowledge of the parameters C and T_{base} it is possible to select the required temperature by supplying the appropriate current to the mixing chamber resistor. For the DOLIS-COLD refrigerator typical values for these parameters are

$$C = 0.019\mu\text{W mK}^{-2} \quad \text{and} \quad T_{base} = 8\text{mK}.$$

The Oxford refrigerator is built to a similar specification, but not having to work against the additional heat load provided by the side access tube can achieve base temperatures of 5 mK.

2.3 Sample preparation and polarization

In this work the host lattices have all been polycrystalline iron foils of at least 99.998% purity. These are prepared by cold-rolling an iron sheet down to a thickness of $\sim 10\mu\text{m}$ and then annealing under an atmosphere of dry hydrogen at a temperature of $\sim 800^\circ\text{C}$ for 24 hours. Finally, just before use, the foil is chemically etched in a solution of 54 vol % H_3PO_4 (80%), 36 vol % H_2O_2 (30%), 8.5 vol % distilled water and 1.5 vol % butoxyethanol [3].

The nuclear orientation thermometers used in this work, namely $^{57}\text{CoFe}$, $^{60}\text{CoCo}(\text{hcp})$ and $^{60}\text{CoNi}$, were all prepared by the high temperature ($900 - 1000^\circ\text{C}$) diffusion of cobalt chloride into the appropriate foil [4]. After etching, 100% of nuclei in full substitutional lattice sites can be consistently achieved owing to the high solubility of cobalt in iron and nickel.

The host foil and thermometer are soldered onto a solid, high purity, copper cold finger which screws into a copper adaptor attached to the mixing chamber of the dilution unit. Both the Oxford and DOLIS refrigerators have a top loading facility which enables the cold finger to be changed whilst the refrigerator is running. This process, which typically takes ~ 3 hours, is often of vital importance to an on-line experiment where the target foil may need to be changed to remove unwanted longer lived activities built up in the sample during implantation.

The loaded sample and thermometer lie at the center of a 1.5 T superconducting split coil magnet which is used to polarize the magnetic domains in the foils. Typically a field of $\sim 0.7\text{ T}$ is applied, which provides not only magnetic saturation but also ensures that the solder is not superconducting and hence that a good thermal contact to the cold finger is achieved. The field produced by the magnet has a homogeneity of 1% over the sample volume.

2.4 The demagnetization stage

As has already been pointed out, adiabatic demagnetization is an important technique in the attainment of low temperatures. Such a PrNi_5 demagnetization stage has been employed for several years in conjunction with the Oxford dilution refrigerator [5]. This has enabled temperatures of as low as 1 mK to be attained for several hours (rising to about 12 mK during periods of remagnetization) and thus has enhanced the study of nuclei with relatively small hyperfine interactions. The demagnetization stage, having similar dimensions to the cold fingers, is fully top-loadable.

2.4.1 Enhanced nuclear magnetism

In the field of cryomagnetism the traditional type of paramagnetic working substance has been one whose susceptibility closely follows Curie's law.

A different class of paramagnets comprise those where the electronic ground state is a singlet, with electronic Zeeman matrix elements linking it to one or more low lying excited singlet states. The ground state singlet can have no permanent magnetic moment, but when a magnetic field is applied these excited states are mixed in, leading to a large Van Vleck paramagnetic susceptibility. This induced moment then interacts with the nuclear magnetic moment through the magnetic hyperfine interaction and leads to a pseudo-Zeeman nuclear interaction which may be orders of magnitude larger than the true Zeeman interaction. Such a class of ions are the odd Z lanthanide nuclei in the tripositive electronic state, including Pr^{3+} . The effect can be illustrated by using the Hamiltonian [6]

$$H = H_x + g_J \mu_B (\mathbf{B} \cdot \mathbf{J}) + A_J (\mathbf{J} \cdot \mathbf{I}) - g_I \mu_B (\mathbf{B} \cdot \mathbf{I}) + H_Q \quad (2.2)$$

where H_x denotes the interaction with the ligand field and the next term the Zeeman interaction for an ion with electronic spin J . The last three terms represent the magnetic hyperfine interaction for a nucleus of spin I , the nuclear Zeeman interaction ($g_I \sim 10^{-3}$ in Bohr magnetons) and the nuclear electric quadrupole interaction respectively. Assuming that H_x produces a singlet ground state $|G\rangle$ and a singlet excited state $|E\rangle$ with energies $\pm \frac{1}{2}\Delta$, then, neglecting H_Q , the Hamiltonian matrix may be written as

$$\begin{pmatrix} +\frac{1}{2}\Delta - g_I \mu_B B_z I_z & \langle E|J_z|G\rangle(g_J \mu_B B_z + A_J I_z) \\ \langle E|J_z|G\rangle(g_J \mu_B B_z + A_J I_z) & -\frac{1}{2}\Delta - g_I \mu_B B_z I_z \end{pmatrix} \quad (2.3)$$

where B_z is the external magnetic field applied along the z -axis. The ground state energy is then

$$W^G = -g_I \mu_B B_z I_z - \frac{1}{2}\Delta \sec \theta \quad (2.4)$$

with

$$\tan \theta = 2\langle E|J_z|G\rangle(g_J \mu_B B_z + A_J I_z)/\Delta. \quad (2.5)$$

The energies of the nuclear magnetic substates may be separated out to give

$$W^N = -g_I \mu_B (B_z + B_E) I_z \quad (2.6)$$

where B_E is the magnetic field at the nucleus generated by the electrons. In the weak field limit, $\langle E|J_z|G\rangle g_J \mu_B B_z \ll \Delta$, this energy may be written

$$W^N = -g_I \mu_B B_z (1 + K_z) I_z \quad (2.7)$$

so that $B_E = K_z B_z$. The enhancement factor K_z is given by

$$K_z = 2 \{ \langle E|J_z|G\rangle \}^2 g_J A_J / g_I \Delta. \quad (2.8)$$

Effectively, the component of the nuclear moment on the z -axis becomes

$$\mu_z^N = -\frac{\partial W^N}{\partial B_z} = g_I \mu_B (1 + K_z) I_z. \quad (2.9)$$

In Pr^{3+} , where the enhancement factor is $K_z = 145$, the effective nuclear moment is therefore raised to the level of almost one Bohr magneton [6].

In the strong field limit, $\langle E|J_z|G\rangle g_J \mu_B B_z \gg \Delta$, the nuclear Zeeman energy levels of the ground state become

$$W^N = -g_I \mu_B B_z I_z - \langle E|J_z|G\rangle A_J I_z. \quad (2.10)$$

Hence B_E reaches a limiting value equal to that given by the hyperfine interaction when the full electronic magnetic moment of the ground state is achieved, i.e. when $\langle E|J_z|G\rangle = J$. This is enormous compared to the applied field B_z , which may thus be ignored. The splitting between successive nuclear levels is then just JA_J . For Pr^{3+} ions, B_E is 337 T and (in thermal units) JA_J is 210 mK. With such enhanced nuclear Zeeman splittings these ions are particularly suited to cooling by adiabatic demagnetization.

2.4.2 Enhanced nuclear cooling

To achieve cooling by adiabatic demagnetization, the PrNi_5 salt is allowed to thermally equilibrate in an applied field of ~ 2.5 T while in thermal contact with the mixing chamber of the dilution unit. In this field the nuclear Zeeman splittings roughly correspond to the strong field limit (2.10) so that, for example, by magnetizing at 50 mK as much as 96% of the Pr^{3+} nuclear entropy may be removed [6]. The demagnetization stage is then thermally decoupled from the dilution unit by allowing an indium heat switch separating the two to become superconducting. The magnetic field is then removed slowly to avoid eddy current heating. In this isentropic process the Boltzmann populations of the Zeeman levels are conserved and so the temperature decreases monotonically with the applied field. The temperature reached on demagnetization to zero field is of the same order as that where the nuclear susceptibility departs from Curie's law. Since the mutual interactions within the nuclear spin system are also enhanced, this limiting temperature is of the order of a mK. However, in practice the applied field is not removed completely since the nuclear specific heat capacity of PrNi_5 is then reduced [7] leading to greater warm up rates.

2.5 The DOLIS-COLD facility

The Daresbury On-Line Isotope Separator - Cryogenic On-Line Device (DOLIS-COLD) makes use of light and medium heavy ion beams from the Nuclear Structure Facility (NSF) Van der Graff generator at the SERC Daresbury laboratory. The NSF is a conventional vertical tandem accelerator which currently operates at terminal voltages of up to 20 MV.

A schematic view of DOLIS is shown in figure 2.2. The beam from the vertical tandem is deflected through $\pi/2$ and focussed by a single magnetic quadrupole doublet onto the target of the DOLIS on-line ion source (both FEBIAD and Thermal Ion Source). The outgoing secondary beam then passes through an extractor electrode, a cylindrical Einzel lens and an accelerating gap. The 30 kV extractor backs off the 100 kV accelerating supply so that the secondary beam energy does not depend on the extractor voltage. In practice, the ions are usually accelerated by only 60 kV. Vertical focussing is provided by a magnetic quadrupole triplet before mass separation occurs in the 60° sector dipole analyzing magnet. The dispersion and resolution is sufficient to separate nuclei with mass numbers up to $A \sim 250$. Following the collection chamber an electrostatic switchyard distributes the beam into one of three lines. The central line leads via

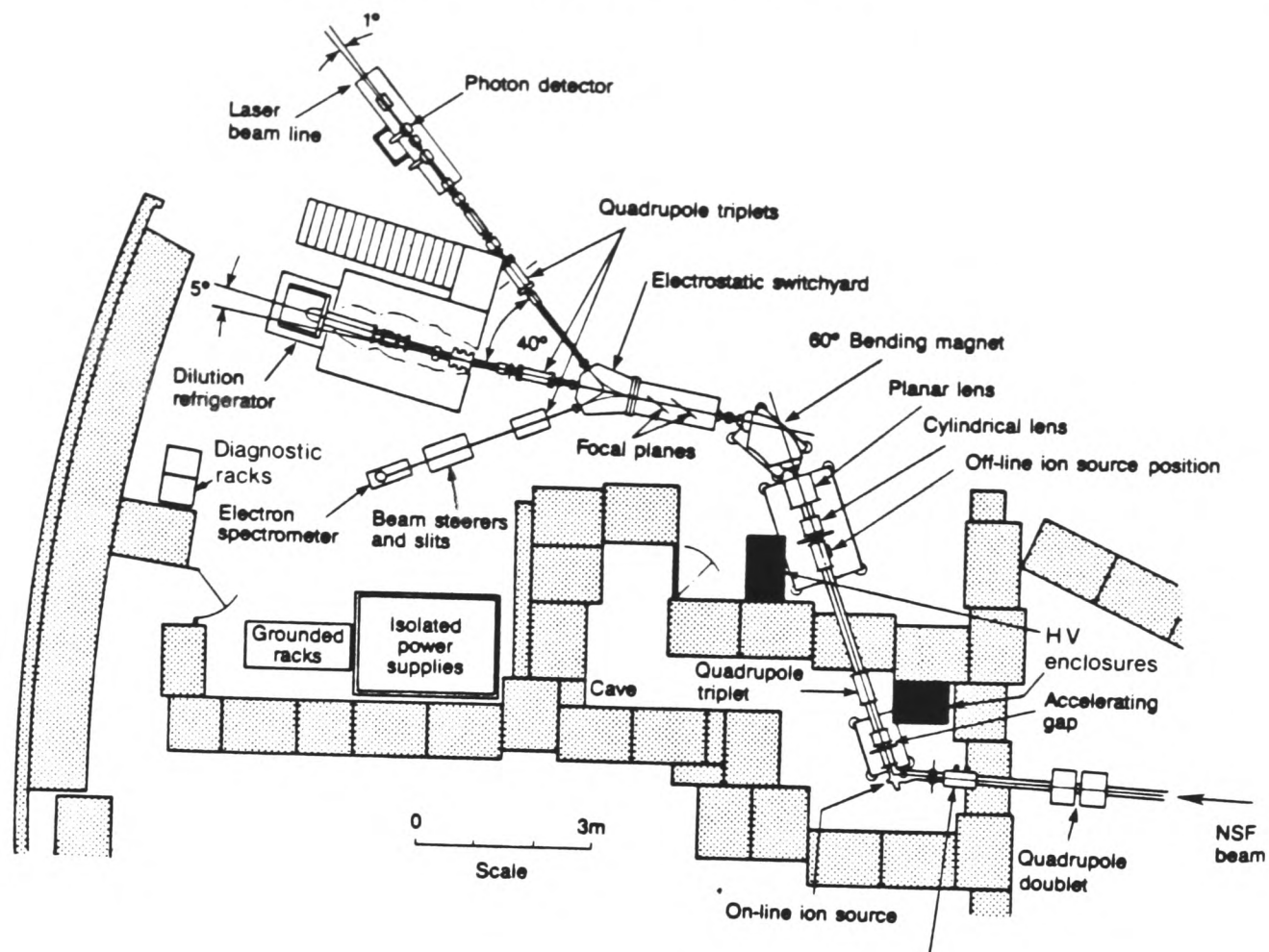


Figure 2.2. A schematic view of the DOLIS facility.

a cooled side access tube to the dilution refrigerator. This incorporates a 5° bend which allows the cold finger of the refrigerator to face a plate cooled to 77 K. The general purpose Mini-Beam Line is at 40° to the central line and in this work has been used for conversion electron studies. The final beam line is used for laser spectroscopy. A full description of the operation of DOLIS has been given by Grant *et al.* [8].

2.5.1 The cooled side access

In an on-line experiment the dilution refrigerator has two extra heat loads placed upon it in addition to the usual heat leaks. These are caused by both the thermal radiation from the incoming beam line and the 60 keV mass separated beam itself. This heat load must be kept at the μW level if temperatures of ~ 10 mK are to be maintained. To this end the refrigerator is supplied with a one metre long side access tube, the inner copper wall of which is cooled to 4 K by thermal contact with the refrigerator's main helium bath. The side access tube and refrigerator are illustrated in figure 2.3 .

One metre from the cold finger there is a variable diameter iris at 77 K and at 40 cm a fixed aperture lead plug, maintained at 4 K by contact with the wall of the side access tube. These not only act as radiation baffles, but also prevent scattered beam from striking the target.

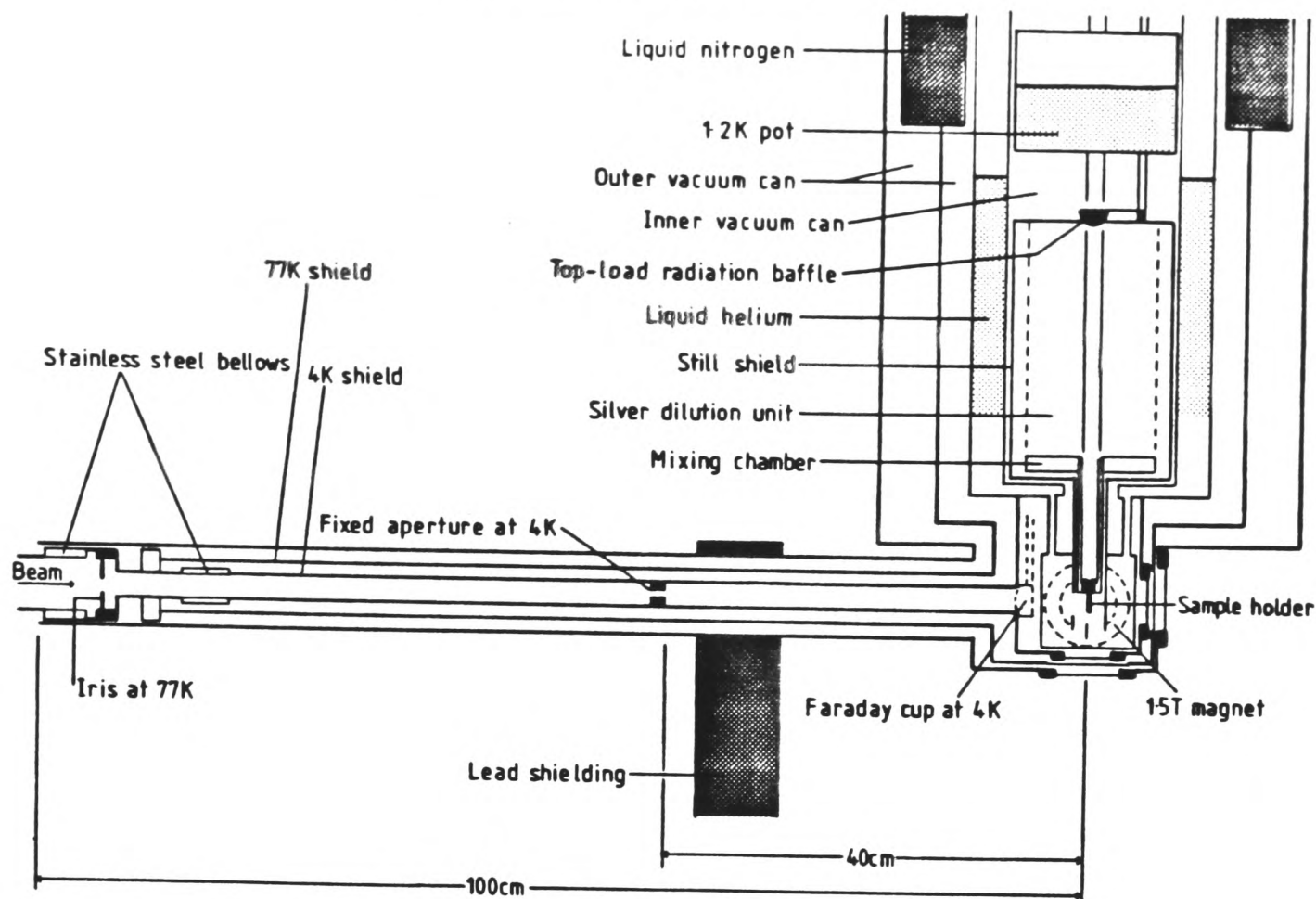


Figure 2.3. The Daresbury dilution refrigerator and side access tube.

Finally, at 7 cm from the sample position, there is a closeable 4 K radiation baffle which has been modified to allow ion current measurements. This can be fully closed for off-line work or for beam diagnostics. The detectors around the refrigerator are shielded against any activity accumulated on the lead plug by an 8 cm wall of lead bricks. As beam transmission rates of $> 90\%$ are consistently achieved, this level of background radiation is never very great.

2.5.2 The Mini-Beam line

The Mini-Beam Line is a general purpose line which in the past has mainly been used for electron conversion studies, allowing the measurement of gamma ray multipolarities and facilitating the search for $E0$ transitions.

The beam line terminates in two cubes. The separated beam enters the upper cube directly and is collected on a movable 6.4 mm wide cassette tape. Activity may thus be transported down to the lower cube. In this way longer lived isotopes can be studied in the absence of shorter lived varieties and vice versa. Both cubes have re-entrant covers to allow the detectors to be positioned as close to the tape as possible for coincidence measurements. Electron spectroscopy is carried out using a mini-orange magnetic filter in conjunction with a cooled Si(Li) detector. The electrons are focussed onto the face of the Si(Li) detector (which is shielded from direct

gamma rays by a lead plug in the axis of the magnet) while positrons are defocussed and therefore not detected.

Spectroscopic studies which can be performed on the mini-beam line include:

1. $\gamma - \gamma$ coincidence
2. $\gamma - \beta^-$ coincidence
3. conversion electron spectroscopy
4. half-life measurements.

2.6 Data acquisition

For all the work to be described, gamma rays have been observed using Ge(Li) detectors which combine reasonable photopeak efficiency (typically $\sim 25\%$ relative to a $3'' \times 3''$ NaI detector at 25 cm from the source) with adequate resolution (typically ~ 2.5 keV at 1333 keV). For directional distribution measurements these are positioned axially ($\theta = 0$) and equatorially ($\theta = \pi/2$) with respect to the polarization axis and detect gamma events in *singles* mode. The Daresbury configuration allows for two axial and two equatorial detectors while at Oxford only one axial and one equatorial detector is used. The detector to source distance is usually ~ 8 cm. A block diagram illustrating singles mode data acquisition is shown in figure 2.4 .

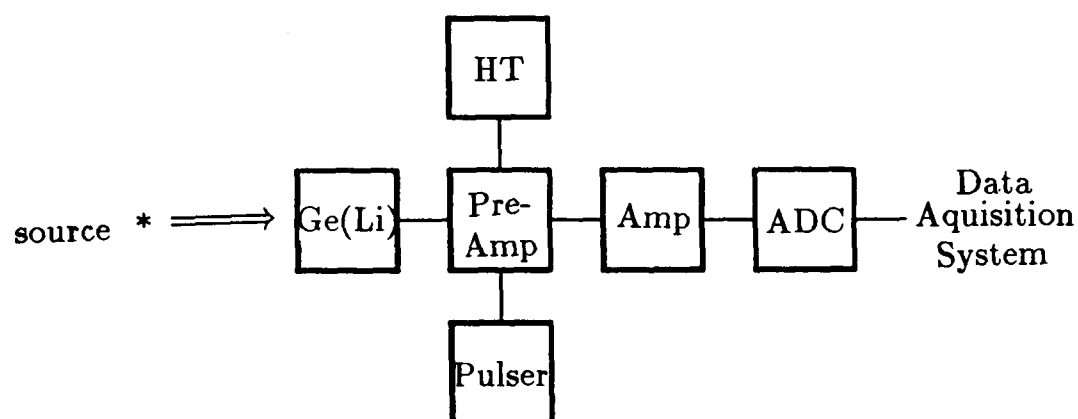


Figure 2.4. A typical singles mode data acquisition arrangement.

In order to correct for dead-time losses in the system (see subsection 3.3) a pulse generator is connected to the test input of the detector pre-amplifier. The pulser frequency is normally set to ~ 10 Hz and has an amplitude such that it is placed at a convenient position at the top

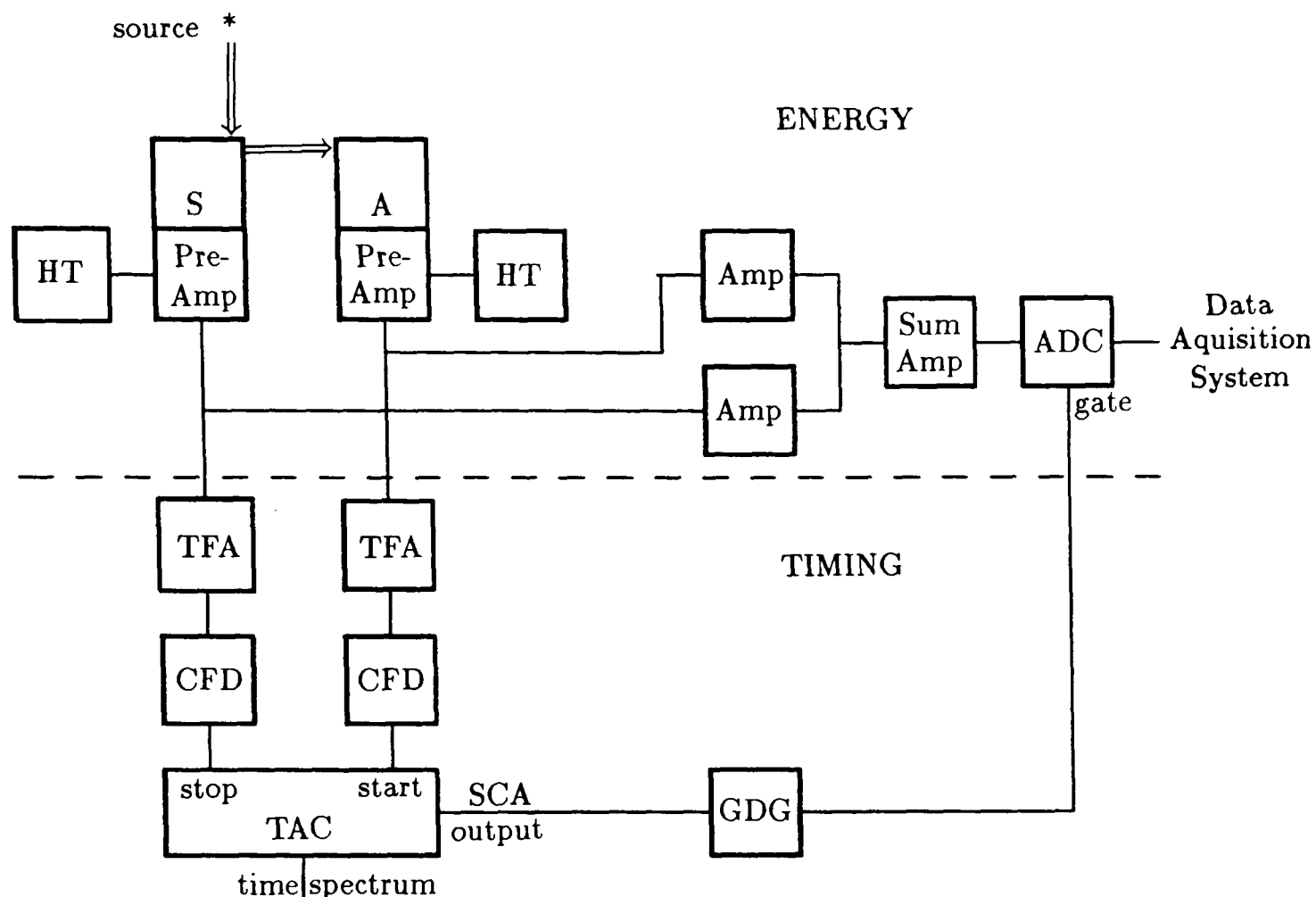


Figure 2.5. A coincidence mode data acquisition system. Energy and timing signals are separated by the dashed line.

of the gamma ray spectrum. The pre-amplifier signals are amplified and directed into standard Analogue to Digital Converters.

In addition to simple directional distribution measurements, polarization measurements can also be performed. Here *coincidence* is required between partially detected gamma ray events in a central detector placed in the polarimeter axis and the subsequent detection of the scattered gamma ray events in one of the onlooking polarimeter detectors. The energies of the two partial gamma ray events are then summed, thereby reconstructing the energy of the original gamma ray. Figure 2.5 shows the main features of such a coincidence data collection system. The scattering detector (also used for directional distribution measurements in singles mode) and one of the analyzing polarimeter detectors are labelled **S** and **A** respectively. The circuitry is divided into energy handling and timing, both taking signals from the same pre-amplifier outputs. The two energy signals are amplified, summed and the passed on to a *gated* ADC. The two timing signals are shaped and digitized by passage through Timing Filter Amplifiers and Constant Fraction Discriminators before entry into the Time to Amplitude Converter as the start/stop pulses. Since the analyser **A**, which in principle only detects previously scattered events from the scatterer **S**, has by far the smaller count rate, it is prudent to use its timing signals as the TAC start pulse and those of the scatterer for the stop pulse. This of course reverses the

true time ordering of the two signals necessitating the addition of appropriate electronic delays. The TAC produces an output whose amplitude is proportional to the time delay between the start/stop signals. This “time spectrum” is characterized by a constant background of events due to random coincidences, together with a peak corresponding to the time delay between true coincidences. The time spectrum is then “cut” so that the Single Channel Analyzer output of the TAC only produces a signal if the start/stop time differential lies within this true coincidence window. The SCA output, after passage through a Gate and Delay Generator, is used to provide the gating pulse for the ADC. In this way true coincidence events are selected for post ADC processing and data storage.

2.7 Thermometry

In any nuclear orientation experiment accurate measurement of the low temperature is of the utmost importance. The natural approach for such measurements is to capitalize upon the LTNO effect itself. An experimentally viable LTNO thermometer needs to satisfy several important conditions, namely:

1. the hyperfine interaction is well known and a 100% occupation of full substitutional sites in the host lattice can be relied upon,
2. the decay scheme is simple with accurately known $U_\lambda A_\lambda$ coefficients,
3. the nuclear spin-lattice relaxation time is short.

Further experimental considerations of less thermometric importance include long half lives and small radioactive heating.

The useful temperature range of a thermometer can be defined in terms of the sensitivity function $T\partial W(\theta, T)/\partial T$. The maximum of the sensitivity function (for axial measurements) is denoted T_{max} . The upper and lower limits of the useful temperature range, T_u and T_l , define the region where the sensitivity function is greater than 5%. These properties for some of the most frequently used LTNO thermometers are shown in table 2.1 [4]. Not surprisingly, there is little difference between T_{max} and that temperature for which the Boltzmann parameter β (1.27) is unity.

Table 2.1. The useful temperature ranges of the more commonly used LTNO thermometers.

Thermometer	E_γ (keV)	βT (mK)	T_l (mK)	T_{max} (mK)	T_u (mK)
$^{54}\text{Mn}\underline{\text{Fe}}$	834.8	- 9.2	1.8	7.2	51
$^{54}\text{Mn}\underline{\text{Ni}}$	834.8	-13.1	2.6	10.4	74
$^{57}\text{Co}\underline{\text{Fe}}$	122.1	-14.2	6.5	16.3	40
	136.5		2.9	12.4	90
$^{57}\text{Co}\underline{\text{Ni}}$	122.1	- 5.9	2.8	6.8	17
	136.5		1.2	5.2	37
$^{60}\text{Co}\underline{\text{Co}}(\text{hcp})$	1173.2/1332.5	- 6.1	1.3	6.9	50
$^{60}\text{Co}\underline{\text{Fe}}$	1173.2/1332.5	- 8.0	1.8	9.0	66
$^{60}\text{Co}\underline{\text{Ni}}$	1173.2/1332.5	- 3.3	0.8	3.7	27
$^{166}\text{Ho}^m\underline{\text{Ho}}(\text{hcp})$	711.7	137	57	300	1250
	810.3		32	137	750

The experiments described in this thesis were performed in two temperature ranges. Those which employed the dilution refrigerator alone lead to temperatures of 8 – 40 mK, for which $^{57}\text{Co}\underline{\text{Fe}}$ and $^{60}\text{Co}\underline{\text{Co}}(\text{hcp})$ thermometers are appropriate. Where the demagnetization stage was used in conjunction with the dilution refrigerator, leading to temperatures of 2 – 12 mK, a $^{60}\text{Co}\underline{\text{Ni}}$ thermometer was employed. The sensitivity functions and decay schemes for ^{57}Co and ^{60}Co are shown in figures 2.6 and 2.7 respectively [4].

The accuracy of this method of thermometry has previously been put to the test by a direct comparison of a $^{60}\text{Co}\underline{\text{Co}}(\text{hcp})$ thermometer against a Josephson junction noise thermometer [9]. In the temperature range 10 – 50 mK, the results show only a 0.5% difference in the implied temperatures from the two devices. Thus the measurement of temperature by the LTNO method with accuracies in the region of 1 – 2% is relatively straightforward and unambiguous.

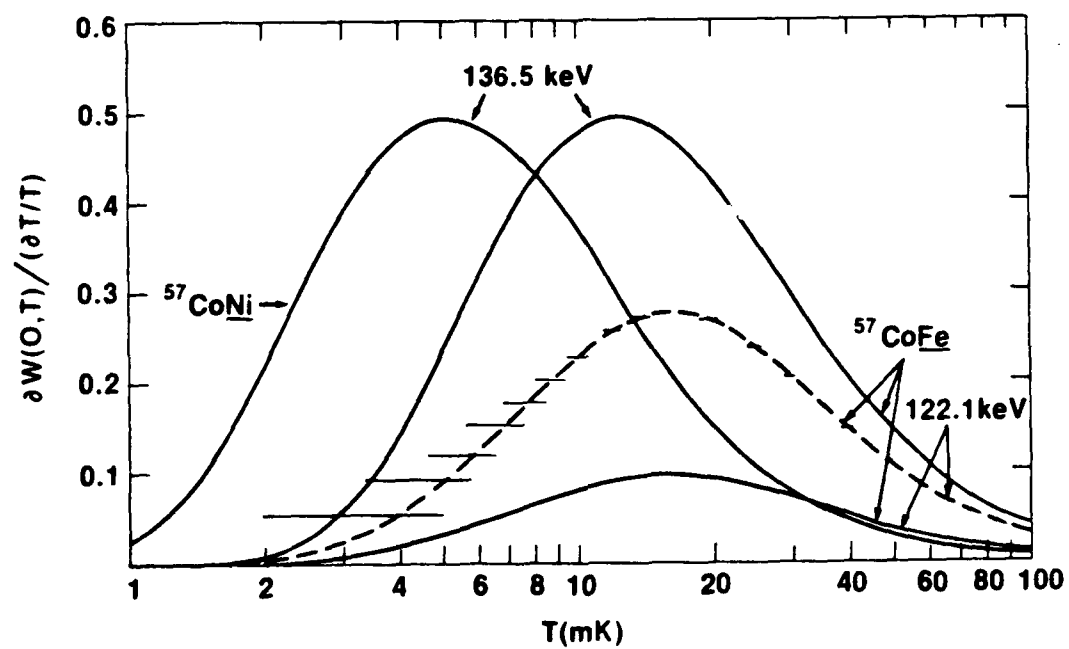
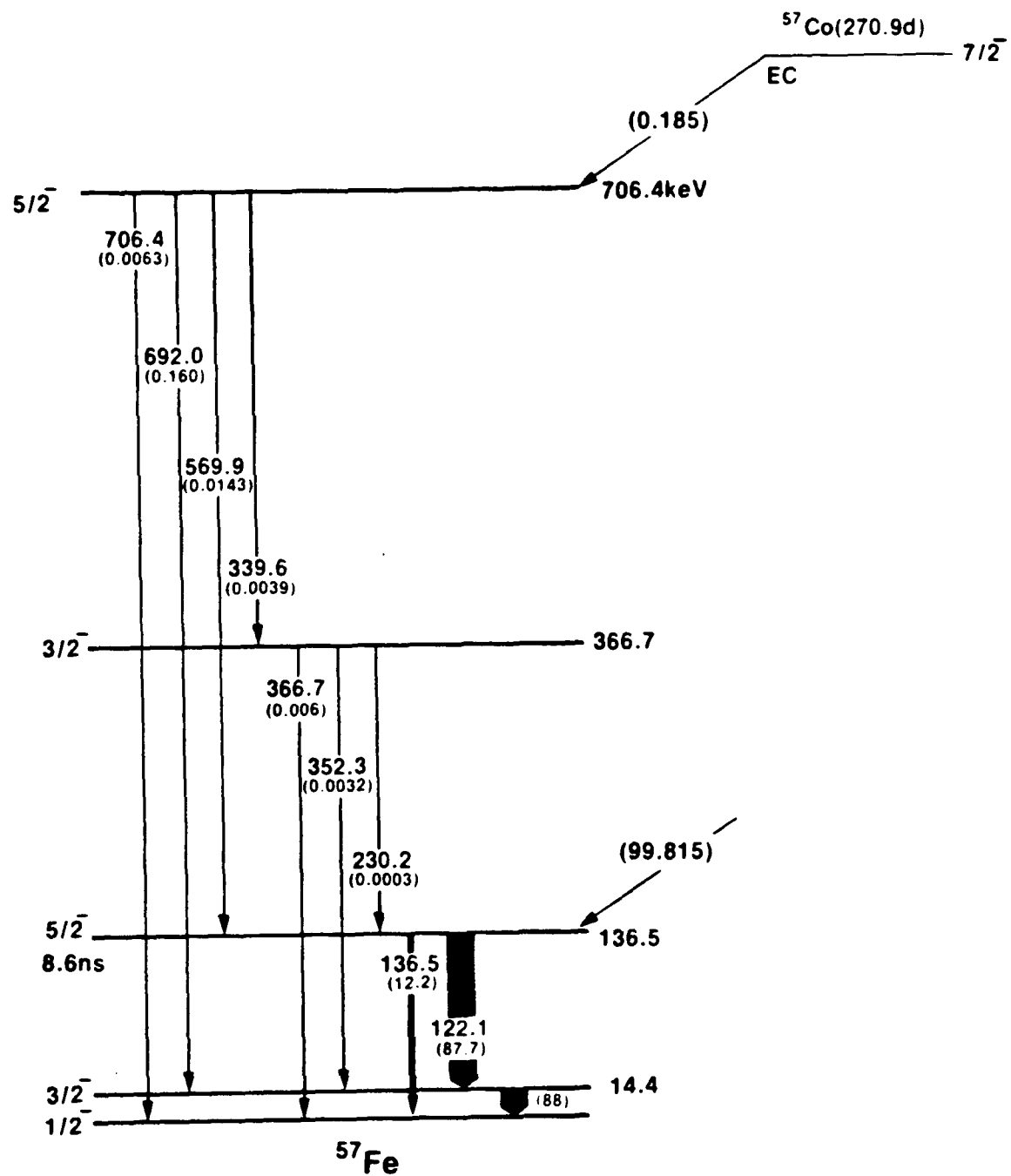


Figure 2.6. The decay scheme and sensitivity functions of ^{57}Co in Fe and Ni. The dashed line represents the effective, intensity normalized, sensitivity function of the 122.1 keV transition while the horizontal bars represent the effects of uncertainties in its mixing ratio.

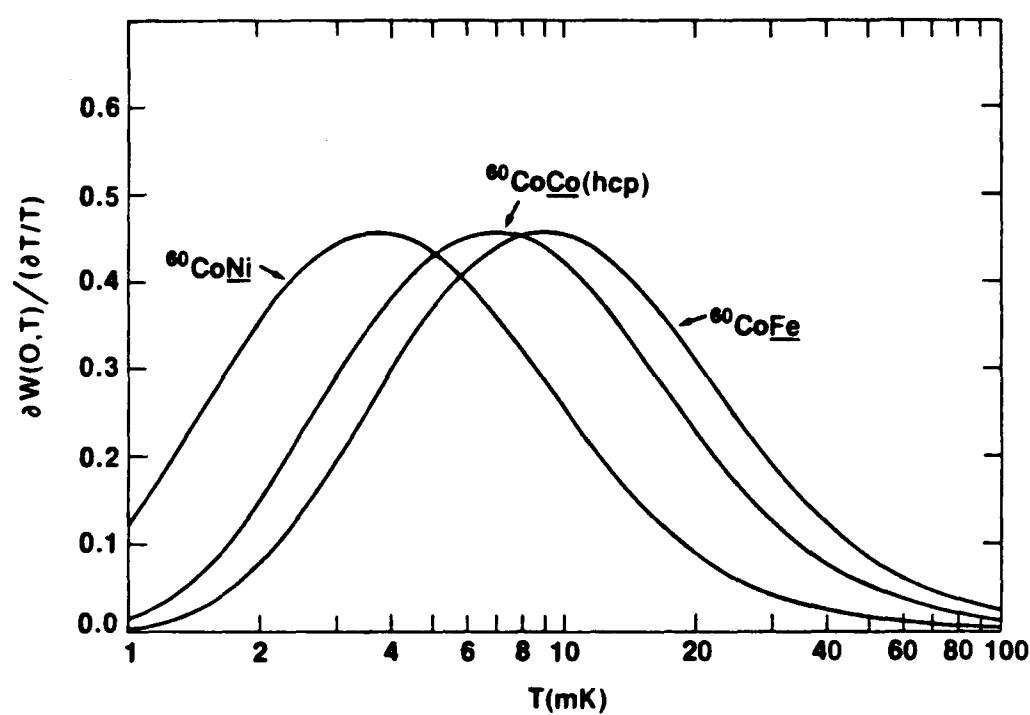
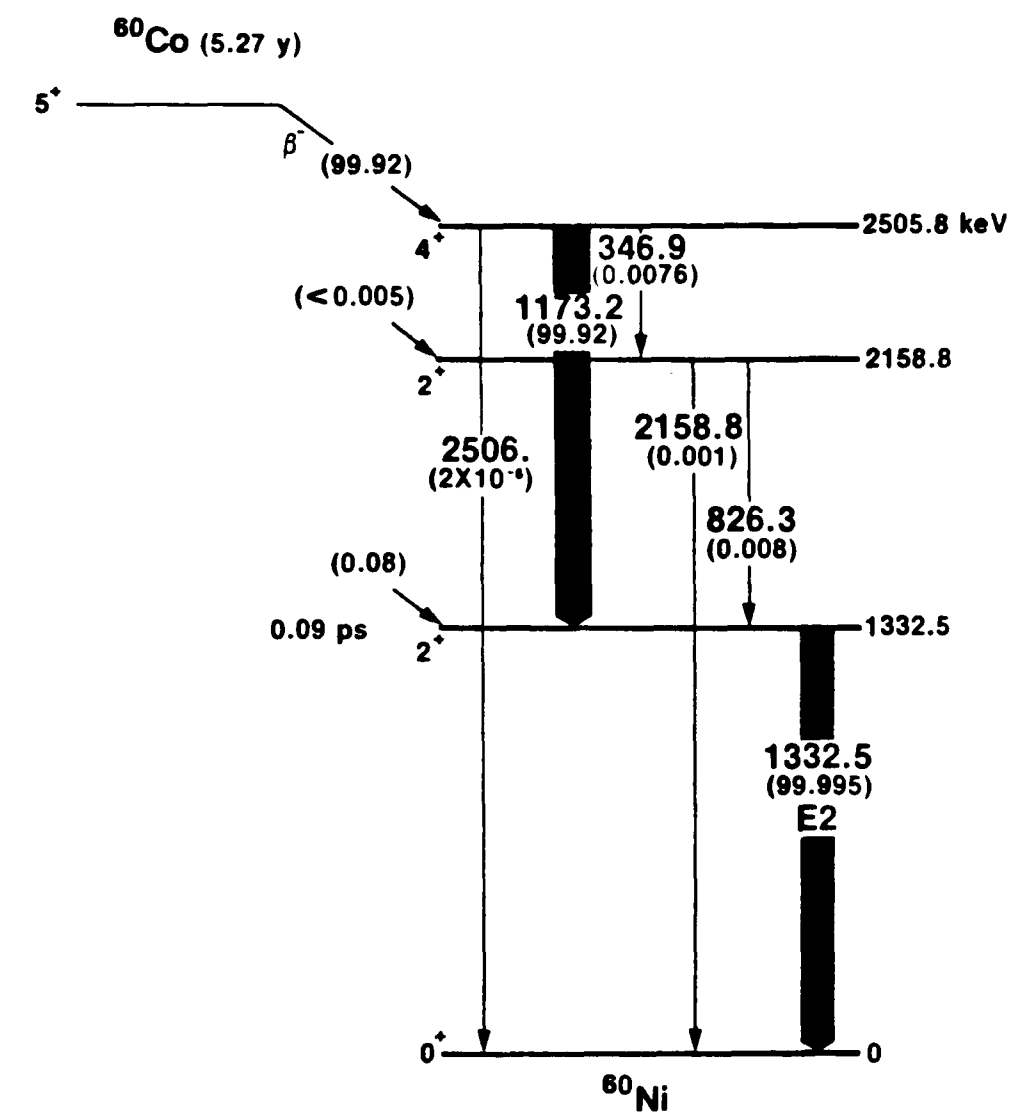


Figure 2.7. The decay scheme and sensitivity functions of ^{60}Co in Co, Fe and Ni.

References

- [1] . H. London *et al.*, Phys. Rev. **128** (1962) 1992
- [2] . O.V. Lounasmaa, Experimental Principles and Methods Below 1 K (Academic Press, 1974)
- [3] . D. Visser, Ph.D. Thesis (University of Groningen, 1981)
- [4] . H. Marshak, in Low Temperature Nuclear Orientation, eds. N.J. Stone and H. Postma (North Holland, 1987)
- [5] . D.I. Bradley *et al.*, Hyp. Int. **35** (1987) 1033
- [6] . B. Bleaney, Physica **69** (1973) 317
- [7] . M. Kubota *et al.*, Phys. Rev. Lett. **45** (1980) 1812
- [8] . I.S. Grant *et al.*, Nucl. Instr. and Meth. **26B** (1987) 95
- [9] . R.J. Soulen and H. Marshak, Cryogenics **7** (1980) 408

Chapter 3

Data Analysis

3.1 Introduction

As has been shown in chapter 1, the angular distribution of gamma radiation from an oriented source is the combined effect of a great many properties of the nuclear system. This chapter will deal with the methods by which these nuclear properties (hyperfine interaction, oriented spin, daughter spins and multipole mixing ratios) are extracted from experimental measurements.

3.2 Spectrum analysis

Once acquired, the gamma ray spectra are analyzed using the Oxford GENeral DATA analysis program, GENDAT †. This package offers two methods of extracting the gamma ray peak areas from these spectra. The first of these, CEN, integrates the counts in the region of interest and subtracts a background linearly interpolated between lower and upper background regions. An example of such a CEN peak fit, with its associated region defining markers, is shown in figure 3.1. Markers $0 \rightarrow 1$ define the lower background region, $2 \rightarrow 3$ the region of interest and $4 \rightarrow 5$ the upper background region.

The second method, FIT, performs a multiple Gaussian peak fit, allowing unresolved multiplets to be analyzed. FIT invokes a least squares fit to the function

$$y(x) = b_0 + b_1x + b_2x^2 + \sum_{p=1}^N A_p \exp \left\{ -\frac{1}{2} \left(\frac{x - c_p}{\sigma_p} \right)^2 \right\} \quad (3.1)$$

where x is the spectrum channel number. The parameters $b_{0,1,2}$ define the background, while c_p and σ_p are the centroids and Full Width Half Maxima of the N peaks labelled by p . Initial

† D. Sinclair and T. Fox, Nuclear Physics Laboratory, Oxford (1983)

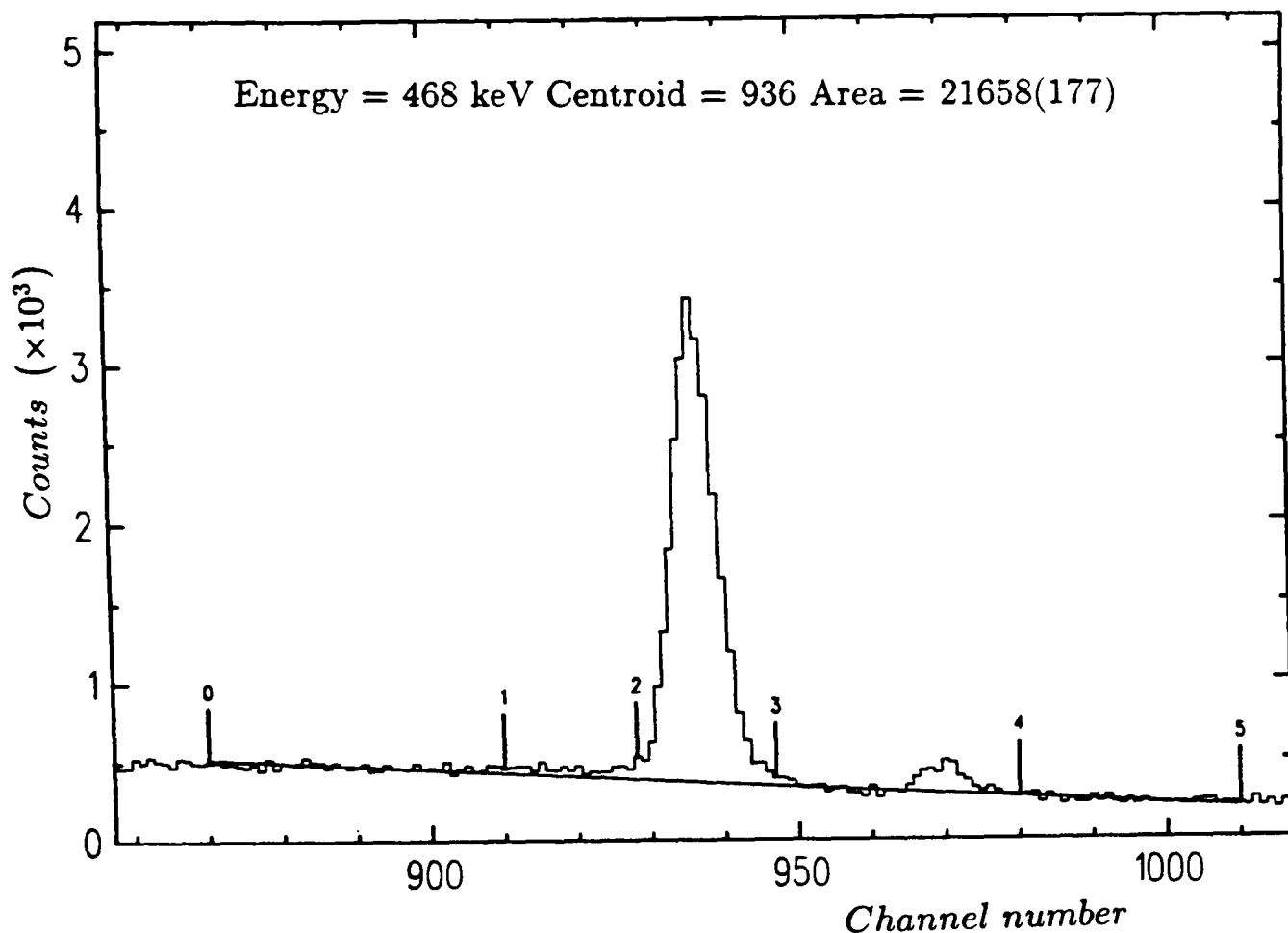


Figure 3.1. The region defining markers appropriate for a CEN peak area fit. The interpolated background is also shown.

guesses for both c_p and σ_p are required. The former is provided by the placement of markers $1 \rightarrow N$, the markers 0 and $N + 1$ being used to define the beginning and end of the region to be fitted. These are illustrated for the case of a triplet in figure 3.2. Initial values for the σ_p are obtained on the assumption that the FWHM is linearly dependent on the gamma ray energy, or more appropriately the centroid c_p . The parameters of this linear function are determined from the fitted centroids and FWHM of two single peaks, one near the beginning and one near the end of the spectrum.

Since the peak shapes need not necessarily be truly Gaussian, the non-peak fitting CEN procedure is used in preference to FIT, unless the use of FIT is forced by the inadequate resolution of multiplet peaks.

Frequently, the background under the peak of interest is discontinuous. In these cases, the 122 keV peak of ^{57}Co being a good example, the behavior of the background under the peak is not clearly defined. GENDAT allows for this by enabling peak markers to be “shifted” from one energy calibration to another. In this way it is possible to apply the *same* markers to *all* spectra, including those of different detectors. Thus although both CEN and FIT may subtract a discontinuous background in a non-unique manner, this is to a large extent compensated by the fact that each peak and its associated background is treated consistently in all spectra.

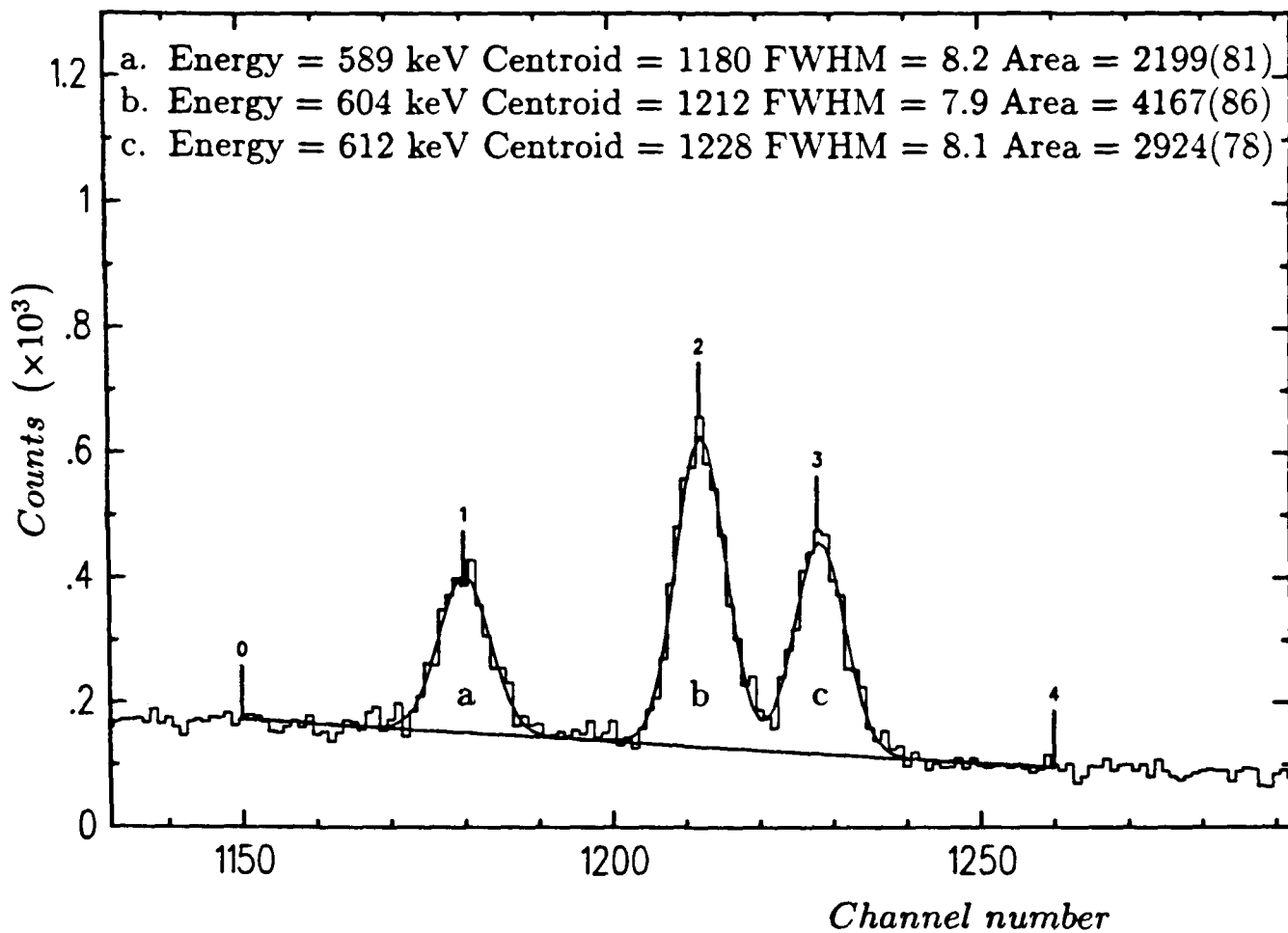


Figure 3.2. The region defining markers appropriate for FIT. The fitted triple Gaussian and the interpolated background are both shown.

In the same vein it is possible to gain shift entire spectra to one control calibration. This is of great use where the detection system suffers from changes in amplification and allows spectra to be added (for improved statistics and hence peak shapes) without fear of a reduction in resolution.

3.3 Experimental determination of $W(\theta)$

In chapter 1 the directional distribution $W(\theta)$ of gamma rays from a LTNO source (1.86) was derived. For a particular gamma ray this can be related to the observed peak area $N_\gamma(\theta)$ by the expression

$$N_\gamma(\theta) = W_\gamma(\theta) I_\gamma \epsilon_\gamma \Omega D \quad (3.2)$$

where I_γ is the gamma ray intensity per parent decay and D denotes the number of parent decays during the counting period. The detector is assumed to have a gamma ray detection efficiency ϵ_γ and to subtend a solid angle Ω at the source.

To clarify the arguments which follow in this section, D will now be evaluated explicitly for the case where the sample decays according to the usual exponential decay law, with decay

constant λ . Thus, for a spectrum whose counting period is of length t_{real} and commences at the time t_{start}

$$\begin{aligned} D &= \int_{t_{start}}^{t_{start}+t_{real}} e^{-\lambda t} dt \\ &= e^{-\lambda t_{start}} \{1 - e^{-\lambda t_{real}}\} \end{aligned} \quad (3.3)$$

where constant coefficients have been neglected for simplicity. It is clear that the factors D depend on the counting period of the spectrum. However, it is necessary to distinguish between the period for which the ADC was actually available for the processing of signals, t_{live} , and the total counting period, t_{real} . Since in the course of registering a signal the ADC is unable to receive further data, these two times will differ by an amount equal to the dead time, t_{dead} . Typically this constitutes 5 – 25% of the counting time, depending on the count rate. This can be accounted for by introducing a corrective factor, t_{live}/t_{real} . Thus

$$D = t_{live} R \quad (3.4)$$

where the rate R is given by

$$R = \frac{e^{-\lambda t_{start}}}{t_{real}} \{1 - e^{-\lambda t_{real}}\}. \quad (3.5)$$

Experimentally it is necessary to have some means of determining the live time, the real time being pre-defined by the ADC. This is achieved by the pulser method [1], in which an artificial peak is introduced into the measured gamma ray spectrum by a stable pulse generator whose signals are fed into the pre-amplifier of the detector (see section 2.6). These undergo the same pulse processing as the nuclear events and are subject to almost the same counting losses. Therefore the area of the pulser peak divided by the constant repetition rate yields the live time.

However, there is a systematic error associated with such a correction. Namely the fact that the pulser signals come more or less evenly spaced in time and thus they are able to interact with the gamma ray signals, which are randomly distributed in time, but not with themselves. As a result the pulser signals can only correct for the partial gamma ray deadtime, not the total dead time to which they themselves contribute. For the most part these errors can be avoided if the pulser frequency is sufficiently low that the ratio of its dead time to that of the gamma rays is kept below 10% [2].

If p denotes the pulser peak area and f its repetition rate, then from (3.4) one obtains

$$D = \frac{p}{f} R. \quad (3.6)$$

Generally peak areas from several spectra j acquired under similar conditions are summed. Thus

$$\sum_j \{N/p\}_{j,\gamma}(\theta) = \frac{1}{f} W_\gamma(\theta) I_\gamma \epsilon_\gamma \Omega \sum_j R_j. \quad (3.7)$$

In order to eliminate the pulser repetition rate, relative intensity, efficiency and solid angle terms, “warm” counts are taken at a sufficiently high temperature, usually ~ 1 K, so that the directional distribution becomes isotropic ($W(\theta) = 1$). Therefore by taking the “cold” to “warm” ratio and rearranging

$$\begin{aligned} W(\theta) &= \frac{\sum_j \{N/p\}_j(\theta)}{\sum_j R_j} \times \frac{\sum_k R_k}{\sum_k \{N/p\}_k(\theta)} \\ &= \sum_{\lambda i} f_i B_\lambda(B_{hyp}^i) U_\lambda A_\lambda Q_\lambda P_\lambda(\cos \theta) \end{aligned} \quad (3.8)$$

where the subscript γ has been dropped for simplicity and the labels j and k are understood to refer to summation over “cold” and “warm” spectra respectively. From this expression, the fundamental measurement in LTNO, the *anisotropy*, can be defined to be $(W(\theta) - 1) \%$.

Thus far only one specific mode of source decay has been considered. If the source is fed by one or more decaying parent nuclei, then D can be derived from (3.3) in an identical manner using instead a multi-exponential integrand. Where the source is produced continuously on-line, then the explicit time dependence of the integrand is unknown as a result of the variable production rate. However, the arguments leading to (3.4), (3.6), (3.7) and (3.8) are still valid, even though R is unknown.

LTNO experiments generally detect the gamma radiation directional distribution at two angles, axial ($\theta = 0$) and equatorial ($\theta = \pi/2$). The data acquisition procedure is such that the counting period begins and ends simultaneously in all detectors. Corresponding axial and equatorial spectra will therefore have common values of R . Applying (3.8) to the ratio of axial and equatorial directional distributions, with the proviso that the summations over the two directions are identical, yields

$$\begin{aligned} \frac{W(0)}{W(\frac{\pi}{2})} &= \frac{\sum_j \{N/p\}_j(0)}{\sum_k \{N/p\}_k(0)} \times \frac{\sum_k \{N/p\}_k(\frac{\pi}{2})}{\sum_j \{N/p\}_j(\frac{\pi}{2})} \\ &= \frac{\sum_{\lambda i} f_i B_\lambda(B_{hyp}^i) U_\lambda A_\lambda Q_\lambda P_\lambda(1)}{\sum_{\lambda i} f_i B_\lambda(B_{hyp}^i) U_\lambda A_\lambda Q_\lambda P_\lambda(0)}. \end{aligned} \quad (3.9)$$

In this way the observed gamma ray peak areas (with pulser normalization) can be directly related to the theoretical angular distribution without making any assumptions as to the nature of the source decay.

The terms on the right hand sides of (3.8) and (3.9) are, in the absence of *a priori* knowledge, multiparameter expressions. Assuming the truncated directional distribution (1.87) leads to just two unknown parameters, $\sum_i f_i B_2 U_2 A_2$ and $\sum_i f_i B_4 U_4 A_4$. In order to calculate these separately it is necessary to retain the two independent axial and equatorial anisotropies (3.8), thus allowing the separation of the two terms by Gaussian elimination. In this way and in the case where the axial and equatorial solid angle correction factors are identical one obtains

$$\sum_i f_i B_2(B_{hyp}^i) U_2 A_2 Q_2 = \frac{1}{7} \left\{ 3(W(0) - 1) - 8(W(\frac{\pi}{2}) - 1) \right\} \quad (3.10)$$

and

$$\sum_i f_i B_4(B_{hyp}^i) U_4 A_4 Q_4 = \frac{4}{7} \left\{ (W(0) - 1) + 2(W(\frac{\pi}{2}) - 1) \right\}. \quad (3.11)$$

Where the series is confined to $\lambda = 2$ by any of the conditions of (1.71), or if *a priori* knowledge allows the two parameters to be related, then there is no loss of information incurred by considering only axial/equatorial ratios.

Finally, it should be noted that the R factors can be removed from (3.8) without calculation by normalizing to a gamma ray peak that is known to exhibit no anisotropy. Presumably this would be by virtue of it having arisen from a level of spin zero or one half and, needless to say, it must belong to the nucleus under consideration in order to achieve the decay correction. In this way separate axial and equatorial anisotropies may be obtained even in the case of on-line sources. However, since a suitable transition may either not be available or may be statistically poor, this is not universally applicable *unlike* the axial/equatorial method.

3.4 Temperature determination

The temperature is determined from the anisotropy of an isotope whose gamma rays have well known orientation coefficients. Since these thermometers are invariably off-line sources, for which the R are always calculable, the temperatures can be extracted from either axial, equatorial or axial/equatorial anisotropies. A more complete discussion of thermometry sources is given in section 2.7.

3.5 Experimental determination of the LTNO parameters

3.5.1 The hyperfine interaction

The extraction of the hyperfine interaction generally proceeds from a least squares fit to the temperature dependence of the experimental anisotropies. Let these anisotropies be denoted by E_l with errors σ_l and correspond to the inverse temperature $1/T_l$. If the appropriate angular distribution function (axial, equatorial or axial/equatorial) to be fitted to these data is $E(1/T_l, \alpha_m)$ which depends on the parameters α_m , then the reduced χ^2 quality of fit is defined as

$$\chi^2 = \sum_l \frac{1}{\sigma_l^2} \{E(1/T_l, \alpha_m) - E_l\}^2. \quad (3.12)$$

Optimum values for the parameters α_m are then found such that χ^2 is minimized in the parameter space. The error σ_{α_n} on the parameter α_n is determined from the condition [3]

$$\chi^2(\alpha_n \pm \sigma_{\alpha_n}) = \chi_{min}^2 + 1 \quad (3.13)$$

where the term on the left hand side is minimized with respect to the remaining parameters $\alpha_{m \neq n}$, while the first term on the right hand side is the absolute minimum of χ^2 with respect to all parameters. This procedure is accomplished by means of the minimizing package MINUIT [4] from the CERN library.

In the case of directional distributions, the parameters α_m refer to the f_i , the hyperfine interaction (electric and magnetic), the U_λ and the A_λ (or the multipole mixing ratio δ). Thus by performing a fit to the temperature dependent data the hyperfine interaction may be deduced. In principle, since the orientation parameters depend on both the Boltzmann parameter β and the oriented parent spin I_0 , it is also possible to deduce I_0 in the fitting procedure. In practice the presence of more than one lattice site with non-zero hyperfine interaction, or even the relative effects of the second and fourth rank terms, often have a greater influence on the shape of the temperature dependence and so the more subtle shape differences due to I_0 may be lost.

3.5.2 The angular distribution and de-orientation coefficients

Once the hyperfine interaction and hence the orientation parameters are known, then the U_λ and A_λ coefficients can be calculated from (3.10) and (3.11) (and also from (3.9) if only second rank terms are present). They are also obtained, if sometimes less reliably where both second and fourth rank terms occur, as by-products of the above fitting procedure. In fact if only axial/equatorial ratios are available then this latter method is the only way in which the second and fourth rank terms can be separated if both are present.

Since neither the U_λ nor the A_λ coefficients are temperature dependent they cannot be distinguished experimentally. In the case where the source is “soluble” in the host matrix (1.88) the same is also true of f and so experimentally it is only possible to determine the product $fU_\lambda A_\lambda$. The $U_\lambda A_\lambda$ can only be calculated separately if the factor f can be determined from a gamma ray with known $U_\lambda A_\lambda$ coefficients. If the source is “insoluble” then the more general directional distribution formula (1.87) must be invoked. Here the f_i , by virtue of their association with the $B_\lambda(B_{hy}^i)$ coefficients, scale not only the magnitude of the anisotropy but also its temperature dependence. Thus the parameters f_i are determined directly in the fitting process and the $U_\lambda A_\lambda$ can be obtained immediately.

Further analysis depends on the knowledge of certain details in the decay scheme of the daughter:

1. If the U_λ coefficients can be calculated theoretically for a certain level according to (1.67) and (1.69), then the A_λ coefficients can be deduced for all gamma rays arising from this level. From these it is possible to calculate any one of the properties δ , I_i and I_f using (1.63), provided that the other two are known. Occasionally, if the A_λ coefficients are consistent with a transition of pure multipolarity, then it is possible to deduce either one of the initial or final level spins knowing only the other.

2. If the U_λ coefficients of a level are not calculable, then in order to deduce them experimentally one relies on the knowledge of the A_λ coefficients for one of the daughter transitions. The U_λ coefficients thus derived can, if sufficient information is known *a priori*, be used to determine any one of Δ_β , δ_γ , I_i , I_f and the relative intensities of the feeding transitions (see subsection 1.2.7). Further analysis then proceeds using the method outlined in 1.

In this way it is possible to extract a great deal of useful spectroscopic information about the daughter nucleus, provided that certain details of the decay scheme are known *a priori*. Without being privy to such details, however, the analysis of LTNO data frequently proceeds little further than the extraction of the hyperfine interaction.

References

- [1] . H.H. Bolotin *et al.*, Nucl. Instr. and Meth. **83** (1970) 1
- [2] . M. Wiernik, Nucl. Instr. and Meth. **96** (1971) 325
- [3] . D.W.O. Rogers, Nucl. Instr. and Meth. **127** (1975) 253
- [4] . F. James and M. Roos, Comp. Phys. Comm. **10** (1975) 343

Chapter 4

The magnetic moments of the light bromine isotopes

4.1 Introduction

Among the nuclei off the line of stability those in the $Z \sim N \sim 40$ region are of particular interest, exhibiting large quadrupole deformations, triaxiality and shape coexistence [1–5]. The nuclear ground state spins of the odd A bromine nuclides $^{75-87}\text{Br}$ with $Z=35$ are all $3/2^-$. Nilsson orbital calculations show that the last odd proton occupies the $[301]3/2$ orbital for deformations $\epsilon < 0.22$ and the $[312]3/2$ orbital at larger prolate deformations. The magnetic dipole moments of these two configurations differ by a factor of three and thus clearly identify the single particle configuration and hence the approximate deformation of the ground state.

With this aim static LTNO measurements have been performed on the light bromine isotopes $^{72g,72m,73,74m,75,76,77}\text{Br}$. For clarity, the analysis of these experiments will be separated into three chapters. This chapter will describe the experimental details and the measured magnetic dipole moments. Chapter 5 will be concerned with $U_\lambda A_\lambda$ coefficients and the spectroscopy of the daughter Se isotopes, while the theoretical interpretation of the magnetic moments will be given in chapter 7.

4.2 Experimental details and data analysis

Sources of ^{76}Br (16 hrs) and ^{77}Br (57 hrs) implanted at room temperature into polycrystalline iron foils were prepared at the ISOLDE facility, CERN, using the reaction of 600 MeV protons on a target of niobium powder. These were subsequently studied in Oxford at temperatures down to 2 mK using the ^3He - ^4He dilution refrigerator in conjunction with the PrNi_5 demagnetization stage. Data was recorded by two $\text{Ge}(\text{Li})$ detectors positioned axially and equatorially relative to the axis of orientation, defined by a 1.0 T polarizing field. The source to detector distance was 12 cm.

The isotopes $^{72}\text{Br}^{g,m}$ (1.3 m, 10.1 s), ^{73}Br (3.4 m), $^{74}\text{Br}^m$ (46 m) and ^{75}Br (97 m) were produced on-line using the reaction of 150 MeV ^{28}Si on a ^{54}Fe target which formed part of the FEBIAD ion source of DOLIS. After acceleration to 60 keV, the selected bromine ions were implanted into a polycrystalline iron foil soldered to the cold finger of the dilution refrigerator at temperatures down to 8 mK. Production rates were low for the lighter isotopes, but well resolved gamma spectra were recorded in four Ge(Li) detectors, two in both the axial and equatorial positions. The detector to source distance was 8 cm. The iron foil was polarized by a 0.7 T magnetic field in a direction parallel to its surface and perpendicular to the implanted beam direction.

With regard to data analysis, two different models describing the lattice site occupation of the bromine nuclei in the iron matrix will be assumed. The first model, henceforth known as model 1, is that applicable to “soluble” implants (1.88) where a fraction f of nuclei experience the full substitutional hyperfine field B_{sub} , while the remaining $1 - f$ occupy lattice sites with zero hyperfine field. As an alternative, hereafter referred to as model 2, a simplified form of the model applicable to “insoluble” implants (1.87) is taken. Here, as before, a fraction f of nuclei experience the full substitutional hyperfine field B_{sub} , but now it is assumed that the remaining $1 - f$ occupy a second lattice site subject to a single non-zero hyperfine field, B_{low} .

Herzog *et al.* [6], using NMR/ON techniques in ^{82}Br , have measured the substitutional magnetic hyperfine field of BrFe to be $B_{sub} = +81.38(6)\text{T}$. Using this field it was found that the magnetic moment extracted from a model 1 fit to the integral LTNO data, as obtained from a low temperature implanted sample, differed by 10% from the known moment. A satisfactory fit could, however, be made using model 2 with $B_{low} = \pm 26(2)\text{T}$. A subsequent work [7] on both room and low temperature implanted samples over a wide range of doses showed that it was generally necessary to invoke model 2 in order to reproduce the magnetic moment. The variation in the value of B_{low} extracted from the various runs was no greater than would be expected from the statistical error of the value quoted above. No further complexity of site distribution can be meaningfully tested by the integral LTNO method. Extensive channelling studies by Alexander *et al.* [8] in the BrFe system at room temperature and relatively high implantation dose support this multiple site picture. They concluded that there was indeed a second, well defined, non-substitutional site and that this location was independent of the local bromine concentration over a wide range. Building upon this work, Callaghan *et al.* [9] performed an integral LTNO experiment on $^{82}\text{BrFe}$ in order to evaluate the two hyperfine fields. On the reasoned assumption that the second field contributed little to the experimental anisotropy and could therefore be neglected, they determined the substitutional hyperfine field to be $\pm 84(12)$

T with an associated population of 36(5)%. Thus even using the crude model 1 they were able to predict the substitutional hyperfine field to within 15%.

On the basis of these results it can be expected that the model uncertainties will only influence the extracted moments at the level of 10 – 15%. Fitted moments obtained from both of the above models will be quoted in this chapter. The values ultimately adopted, however, will be those of model 2 after assigning errors which reflect the model site distribution uncertainties as well as the statistical errors.

4.3 The isotopes $^{76,77}\text{Br}$

Both of these nuclei were studied prior to the publication of references [6,7] and so it was not anticipated that the shortcomings of model 1 would be significant. Since the magnetic moment of $^{76}\text{Br } 1^-$ had already been measured ($\pm 0.5482(1)\mu_N$ [10]), the experimental effort on the decay of this isotope was concentrated on the extraction of U_2A_2 coefficients and hence spectroscopic information on ^{76}Se . To this end the PrNi_5 demagnetization stage was used to cool the nuclei to 2 mK, as measured by a $^{60}\text{CoNi}$ thermometer, which resulted in the saturation of the gamma ray anisotropies. Unfortunately, these anisotropies were generally rather small, even close to saturation. Of the strongest lines, the 559 and 657 keV transitions showed anisotropies of only +3.8(3)% and -0.6(4)% at 3 mK (see table 5.2). Only the 1130, 1854 and 2951 keV transitions gave anisotropies with sufficiently favourable proportional errors to produce reasonable temperature dependences. These are illustrated in figure 4.1.

The decay scheme of ^{76}Br is shown in figure 5.2 [11]. Due to a combination of uncertain spin assignments and complicated level feedings by a proliferation of weak gamma rays, no U_2A_2 coefficients can be calculated *a priori*. Therefore within the framework of model 1 it is necessary to perform a fit to the experimental axial/equatorial anisotropies with both μB_{sub} and fU_2A_2 as variable parameters. The fitted hyperfine interaction parameters are listed in the second column of table 4.1. From this data the average hyperfine interaction is calculated to be $\mu B_{sub} = 35.0(25)\mu_N\text{T}$. This is to be compared with the expected value (corrected for applied field) of $\mu B_{sub} = 45.16(3)\mu_N\text{T}$.

This 25(6)% difference highlights once again the failure of model 1 to adequately describe the BrFe system as first noted by Herzog *et al.* Following their lead therefore, the more realistic model 2 is invoked. Here, with μ required to reproduce the known value, U_2A_2 , f and B_{low} remain as variable parameters. In order to reduce the number of unknowns, B_{low} was taken

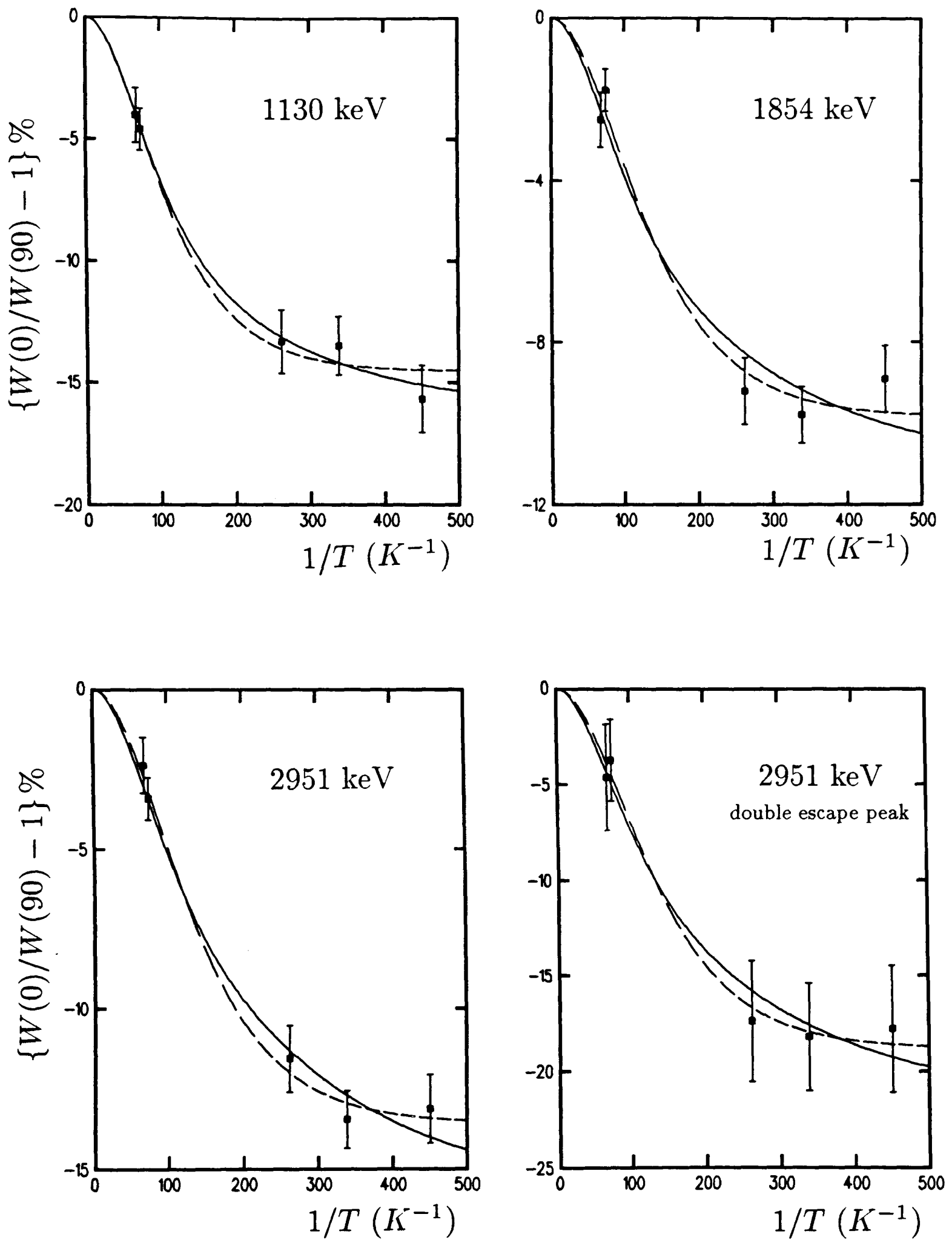


Figure 4.1. The temperature dependences of the 1130, 1854 and 2951 keV transitions in the decay of ^{76}Br . The 2591 double escape peak is also shown. Both the model 1 (dashed) and model 2 (solid) fitted curves are shown for comparison.

Table 4.1. The fitted model parameters for ^{76}Br . The single and double escape peaks of the 2951 keV transition are denoted s.e. and d.e. respectively.

E_γ (keV)	Model 1 fitted μB ($\mu_N\text{T}$)	χ^2	Model 2 fitted f (%)	χ^2
1130	39.9(5.1)	0.4	69.2(17.3)	0.2
1854	33.4(4.5)	1.1	56.8(16.9)	1.5
2951	33.3(4.3)	0.3	51.2(15.5)	0.7
2951 s.e.	34.7(10.9)	0.4	61.6(38.3)	0.6
2951 d.e.	33.2(9.3)	0.1	53.5(32.9)	0.3

to be 26(2)T as discussed in section 4.2. In this way the fitted site distribution parameters f given in the third column of table 4.1 are obtained. The weighted average of this data yields $f = 58(9)\%$.

In order to understand how model 2 is able to correctly reproduce the substitutional hyperfine interaction it is instructive to consider the individual contributions from the various lattice sites. These are illustrated in figure 4.2. The anisotropy data from the 1130, 1854 and 2951 keV transitions (including escape peaks) has been inverted to yield $\sum_i f_i B_2 U_2 A_2$ and then normalized to the 14 mK temperature points, which for this purpose have been combined, and finally summed. The resulting data therefore represents the relative orientation of the ^{76}Br nuclei as a function of temperature. The orientation predicted by model 1, shown dashed, arises solely from the hyperfine interaction of nuclei in substitutional sites. It is clear that the true substitutional hyperfine interaction, $\mu B_{sub} = 45.2\mu_N\text{T}$, increasingly underestimates the experimental orientation as the temperature reduces. This gap between the observed orientation and that due to the fully substitutional lattice site may be bridged by introducing a second lattice site which experiences a non-zero hyperfine interaction. The individual contributions of the substitutional and low field sites, weighted by their relative populations, to the total model 2 orientation are illustrated by the solid lines. The discrepancy between the magnetic moments extracted using the two models may be attributed to the proportion of the total orientation arising from this second non-substitutional hyperfine interaction. In the present case this proportion is as high as 35% at the lowest temperature datum, which results in the large disparity between the two extracted moments. The magnetic moments of the other nuclei studied in this work have all been extracted from data taken at temperatures above 6 mK, where the relative contribution from the second field site is greatly reduced. The model differences and hence the ambiguity in the extracted moment will therefore be correspondingly smaller.

Having determined the site distribution parameter $f = 58(9)\%$ of $^{76}\text{BrFe}$ it is assumed that the same value may be applied to the case of ^{77}Br . This is justified since both were implanted

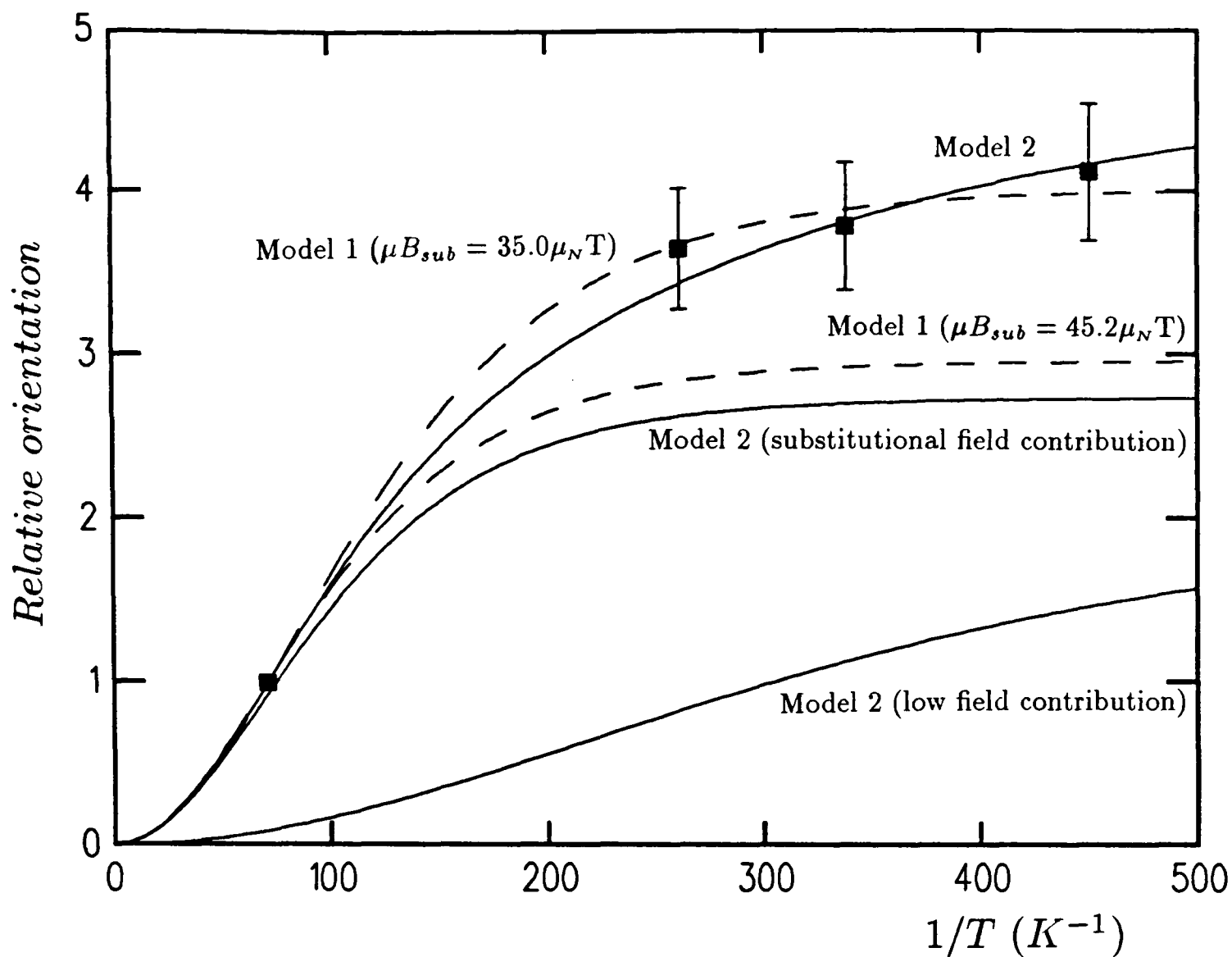


Figure 4.2. The contributions to the total orientation from the model lattice sites. The data is taken from transitions in the decay of ^{76}Br . The model 1 and model 2 fitted curves are distinguished by dashed and solid lines respectively.

under the same conditions and with similar dosage. Further, it is likely that the experimental error in f is sufficient large to take account of any minor variations between the samples.

In contrast the magnetic moment of ^{77}Br $3/2^-$ had not previously been measured. The experiment was therefore performed in two parts. The first took a full temperature dependence down to a temperature of 6 mK as deduced from the anisotropy of a $^{60}\text{CoCo}(\text{hcp})$ thermometer. The second was conceptually identical to the previous ^{76}Br experiment, employing the PrNi_5 demagnetization stage to extend and saturate the temperature dependence data.

The decay scheme of ^{77}Br is given in figure 5.1 [12]. Again no U_2A_2 coefficients are calculable *a priori* even though most of the level spins are known. The variable model 1 parameters are therefore μ and fU_2A_2 , while for model 2 there remains μ and U_2A_2 , since f and B_{low} are considered as known. It should be noted that the site distribution parameter f is *not* the same in the two models. Figure 4.3 shows the temperature dependences of the 304, 439, 575 and 585 keV transitions obtained using the dilution refrigerator alone. The results of both model fits to

this data are shown in table 4.2 for a total of ten transitions. From these results an average model 1 moment of $\mu = 0.82(3)\mu_N$ is obtained. For model 2 a moment of $\mu = 0.92(5)\mu_N$ is extracted. Notice that as a result of the higher temperatures there is only an 11(7)% disagreement between the two models. Comparing the χ^2 values of the two sets of model fits, which are to all intents and purposes identical, it can be seen that even with data of such relatively good quality the integral LTNO method is unable to distinguish between the two models.

Table 4.2. The fitted moments of ^{77}Br .

E_γ (keV)	Model 1		Model 2	
	fitted moment (μ_N)	χ^2	fitted moment (μ_N)	χ^2
239	0.76(30)	5.4	0.90(71)	5.4
250	0.87(19)	1.7	0.99(37)	1.7
304	0.79(10)	0.8	0.90(11)	0.7
385	0.87(11)	1.1	1.00(23)	1.1
439	0.83(12)	0.8	0.95(16)	0.8
485	1.04(26)	0.5	1.22(60)	0.5
521	0.93(19)	3.3	1.14(41)	3.2
575	0.76(5)	0.5	0.85(9)	0.6
585	0.90(8)	0.4	1.04(14)	0.4
755	0.73(21)	1.0	0.81(30)	1.1

Finally, the data taken with the demagnetization stage will be considered. This is illustrated in figure 4.4. Although both parts of the experiment used the same sample, at a given temperature the anisotropies of the demagnetization data are consistently greater than those obtained using the dilution refrigerator alone. This is due to the unsoldering and slight cleaning of the surface of the host iron foil prior to resoldering onto the demagnetization stage in preparation for the latter part of the experiment. This resulted in a small loss of surface activity. Model 1 fits to these anisotropies resulted in moment of $0.81(4)\mu_N$. The close agreement of the two model 1 values indicate that this model is equally valid both before and after the sample was cleaned. In turn this suggests that only surface activity, with no significant hyperfine interaction, was removed. Since the lattice site occupation has clearly changed it is no longer justifiable to take the model 2 site distribution parameter from ^{76}Br . Thus μ , U_2A_2 and now also f must be considered as variable parameters in model 2. However, just as in the case of ^{76}Br , the demagnetization temperature dependence is rather sparse and certainly insufficient for meaningful three parameter fits to be performed. Therefore the model 2 value of μ already derived is assumed and a two parameter fit is performed to obtain f and U_2A_2 . In this way

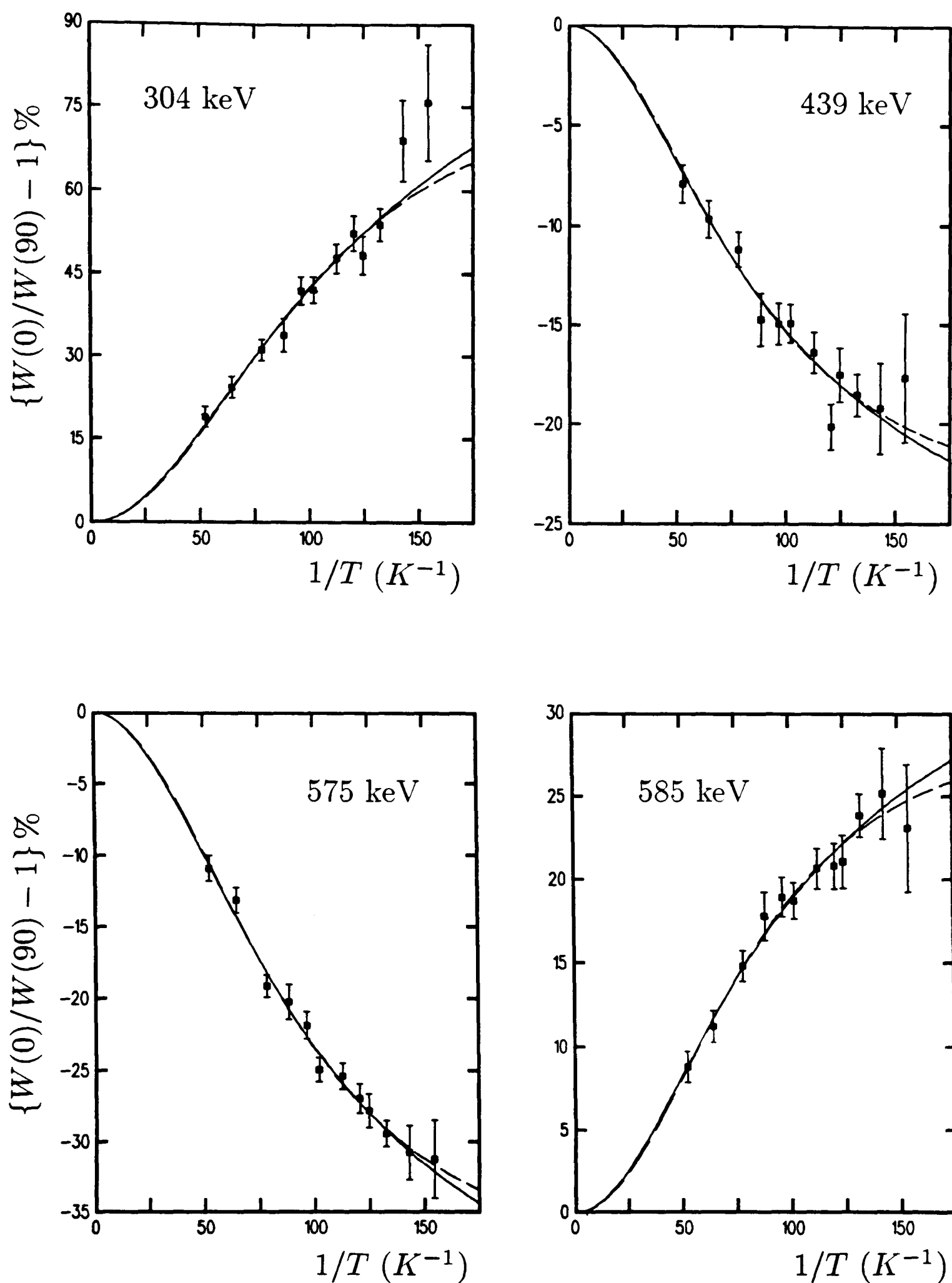


Figure 4.3. The temperature dependences down to 6 mK of the 304, 439, 575 and 585 keV transitions in the decay of ^{77}Br . Both the model 1 (dashed) and model 2 (solid) fitted curves are shown for comparison.

the new site distribution parameter $f = 78(9)\%$ is obtained. Again the χ^2 values of the two models are virtually identical. Since this data has no relevance to the magnetic moment, further discussion will be postponed until chapter 5.

4.4 The isotopes $^{72-75}\text{Br}$

These isotopes were on-line implanted into a cold iron foil at the DOLIS-COLD facility where temperatures as low as 8 mK were achieved, as deduced from the anisotropy of a $^{57}\text{CoFe}$ thermometer. Unlike the previous case, there is no control isotope with a known moment with which to determine f . Therefore within the framework of model 2 there are at least four unknown parameters, μ , f , $U_\lambda A_\lambda$ and B_{low} . In order to be able to extract reliable values for the magnetic moment from the fitting procedure it is desirable to reduce this number to two. Since the $U_\lambda A_\lambda$ coefficients are not generally known *a priori* they must be considered as variable parameters. Therefore not only B_{low} , but also a range of values for the site distribution parameter f is assumed.

The variation in f as a function of implantation conditions of $^{82}\text{BrFe}$ has also been studied by Herzog *et al.* [7]. It is apparent from this work that the value of f is considerably dependent upon implantation dose and also the method of production. However, the values for different on-line implantations were all found to lie within $75\% \geq f \geq 58\%$. Although some dependence upon the method of preparation of the iron foil might also be expected, this range will be considered to be valid for the present work. Since all iron foils used in the present experiments were prepared in an identical manner this will not lead to any problems with internal self consistency. Thus the likely variation in f will be spanned if a maximum value of 75%, model 2a, and a minimum value of 58%, model 2b, is taken. It should be noted that the choice of 58% as the minimum value for the site distribution parameter in on-line samples coincides the value deduced for the off-line prepared samples $^{76,77}\text{Br}$. That the site distribution parameter is probably lower for off-line rather than for on-line prepared samples is apparent from the data of Herzog *et al.* The likely reason for this behaviour in such “insoluble” implants is discussed in subsection 1.3.3.

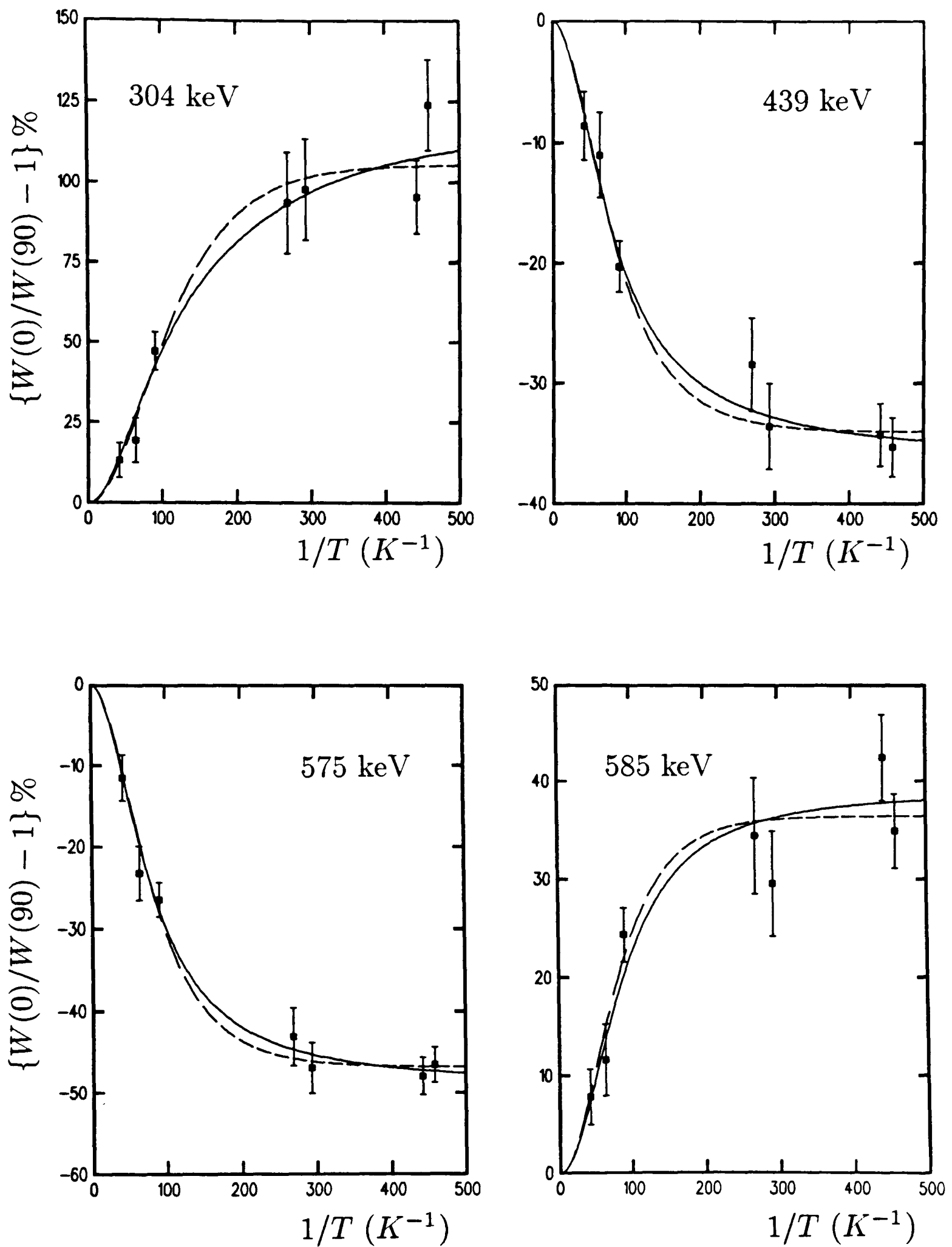


Figure 4.4. The temperature dependences down to 2 mK of the 304, 439, 575 and 585 keV transitions in the decay of ^{77}Br . Both the model 1 (dashed) and model 2 (solid) fitted curves are shown for comparison.

4.4.1 The isotope ^{75}Br

The presence in the DOLIS output of an inactive contaminant beam at this mass required that the ^{75}Br source was built up by implanting for 2.5 h at approximately 50 mK. This was then followed by a period of cooling down to temperatures of 9 mK with the isotope separator closed off. The cycle was repeated several times.

With the exception of the 377 keV transition, which gave data of good statistical accuracy, the resulting anisotropies were rather small. The temperature dependence of this peak is illustrated in figure 4.5. In addition and uniquely among all the isotopes studied, the U_2A_2 coefficient can be calculated for this transition with sufficient accuracy to enable some constraint to be placed on the fitting procedure. The decay scheme is illustrated in figure 5.3 [13]. Using the 664 keV level de-orientation coefficient deduced in section 5.4, in conjunction with the previously known mixing ratio for ^{the} 377 keV transition ($\delta(377) = -0.73(45)$ [13]) leads to the inequality $+0.62 \leq U_2A_2 \leq +0.81$. The model 2 fitted U_2A_2 coefficient is compatible with this result to within one standard deviation for all site distribution parameters $f \geq 44\%$, which greatly exceeds the likely range of variation given above. Therefore the limiting values 75% (model 2a) and 58% (model 2b) are retained and with f thus fixed the inequality for U_2A_2 is used to limit the error on the fitted moment. The results of the fitting procedure are shown in table 4.3 for models 1, 2a and 2b, both with and without the constraint on U_2A_2 . Of course in model 1 this constraint has no effect on the fitted moment and merely serves to provide the inequality $f \geq 53\%$.

Note that despite the large difference between the site distribution parameters of model 2a and 2b there is only a very slight difference in the fitted moments.

Table 4.3. The fitted moments of ^{75}Br . [†]The values tabulated in the second row are constrained by the known U_2A_2 coefficient of the 377 keV transition.

E_γ (keV)	Model 1 fitted moment (μ_N)	Model 2a ($f = 75\%$) fitted moment (μ_N)	Model 2b ($f = 58\%$) fitted moment (μ_N)
377 377 [†]	0.73(9) 0.73(9)	0.75(19) 0.75(15)	0.78(24) 0.78(17)

4.4.2 The isomer $^{74}\text{Br}^m$

The heavy ion reaction of 150 MeV ^{28}Si on ^{54}Fe preferentially produced the high spin isomer. The proportion of the ground state present, as deduced from the intensity ratio of the 615 to 634 keV transitions, was less than 5% and therefore essentially negligible. Large anisotropies with small statistical errors were observed for the 634, 728 and 1250 keV transitions, shown in figure 4.5, and also for the 615, 839, 1201 and 1269 keV transitions.

The decay scheme of $^{74}\text{Br}^m$ is shown in figure 5.4 [14]. The isomeric spin has been deduced by atomic beam resonance methods to be $I = 4$ [15]. As will be discussed in sections 5.5 and 7.5.2, the tentative negative parity assignment to this state, made on the basis of proposed first unique forbidden β^+ /EC decays to levels in ^{74}Se , appears to be wrong. Partly as a result of this, but mainly due to the complex decay scheme, no $U_\lambda A_\lambda$ coefficients are known or can be calculated *a priori*. Further, since the parent has spin $I > 3/2$, these coefficients involve two terms corresponding to $\lambda = 2$ and 4.

Table 4.4. The fitted moments of $^{74}\text{Br}^m$ excluding $\lambda = 4$ terms.

E_γ (keV)	Model 2a ($f = 75\%$)	
	fitted $U_2 A_2$	fitted moment (μ_N)
615	-0.31(3)	1.80(26)
634	-0.21(1)	1.50(8)
728	-0.34(1)	1.70(7)
839	-0.41(5)	1.35(18)
1201	-0.33(5)	1.88(29)
1250	-0.50(3)	1.95(14)
1269	-0.26(4)	1.61(27)

As a first step towards dealing with this extra parameter the data was fitted to model 2a ignoring fourth rank terms. The results are listed in table 4.4 for the seven transitions given above. The weighted mean of these moments is $1.64(5)\mu_N$ for which at 10 mK the ratio of the high field orientation parameters is $B_4(B_{sub})/B_2(B_{sub}) \sim 30\%$. Although this ratio is likely to become smaller when allowance is made for the relative effects of the two $U_\lambda A_\lambda$ terms, the fourth rank terms possibly constitute a sizeable effect. Generally the quality of the temperature dependences is not sufficient to allow the parameter $U_4 A_4$ to be varied meaningfully. However, the temperature dependences of the strong 634 and 728 keV transitions might allow for this extra degree of freedom. Since statistically these contribute most to the final moment there is little to be lost in considering these exclusively. The three fitted parameters for the two transitions

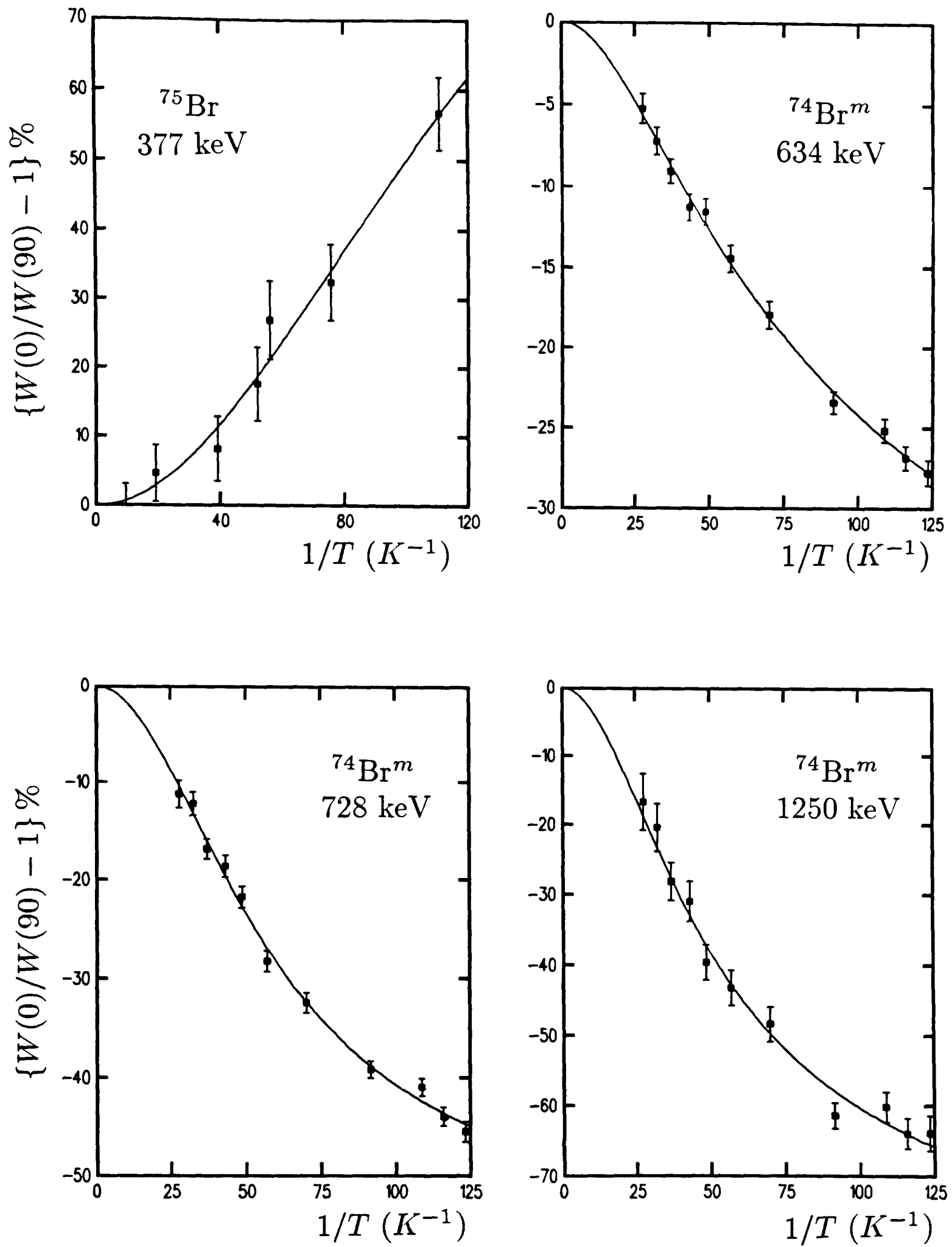


Figure 4.5. The temperature dependences of the 377 keV transition in the decay of ^{75}Br and the 634, 728 and 1250 keV transitions in the decay of $^{74}\text{Br}^m$. The model 1 and model 2 fitted curves coincide.

are shown in table 4.5 from which the weighted averages of the fitted moments are found to be 1.63(6), 1.59(8) and 1.77(11) μ_N for models 1, 2a and 2b respectively. Once again the relatively small difference in the model 2a and 2b moments demonstrates their limited sensitivity to the assumed site distribution parameter.

Table 4.5. The fitted moments of $^{74}\text{Br}^m$ including $\lambda = 4$ terms.

E_γ (keV)	Model 1			Model 2a ($f = 75\%$)			Model 2b ($f = 58\%$)		
	U_2A_2	U_4A_4	μ	U_2A_2	U_4A_4	μ	U_2A_2	U_4A_4	μ
634	-0.10(1)	-0.25(6)	1.95(15)	-0.13(1)	-0.25(7)	1.96(22)	-0.18(1)	-0.18(6)	1.91(17)
728	-0.30(1)	+0.02(8)	1.56(7)	-0.39(1)	+0.21(9)	1.54(8)	-0.42(1)	+0.21(11)	1.68(14)

4.4.3 The isotope ^{73}Br

Anisotropies were observed for 16 of the strongest transitions in the decay of ^{73}Br . These are tabulated in section 5.6. The anisotropy of the $^{57}\text{Co}/\text{Fe}$ thermometer corresponds to an average temperature of 12 mK. All the ^{73}Br effects are consistent with zero orientation. Since the iron foil in which ^{73}Br was studied was the same as that used to successfully orient $^{72,74m}\text{Br}$ in the same on-line run, there can be no possibility of a non-nuclear origin for this lack of anisotropy.

The decay scheme of ^{73}Br is shown in figure 4.6 [14]. Experimentally the ground state spin of ^{73}Br had been assigned $(1/2, 3/2, 5/2)^-$ as a consequence of its allowed β^+/EC decay to the 26 keV $3/2^-$ level in ^{73}Se . On the basis of systematics in the neighbouring odd A isotopes this was assumed to be $3/2^-$. However, the lack of anisotropy is at variance with this hypothesis.

If the ground state were $3/2^-$ and had a moment similar to that of $^{75,77}\text{Br}$ then the relaxation time would be of the order of 5 s (see subsection 4.4.4). When compared to the 3.4 m half life it is clear that incomplete relaxation cannot be responsible for reducing the orientation to the virtually zero levels observed.

It must therefore be concluded that the lack of observed anisotropy is due to the presence of one or more un-oriented spin 1/2 levels. The transitions in ^{73}Se originate from many different levels which are independently fed in the decay. A recent spectroscopic study of the decay of ^{73}Br [16] has assigned spins to some of the levels assuming that the parent spin is $3/2^-$. While most of these are tentative it is clear that not all of the levels which give rise to the observed

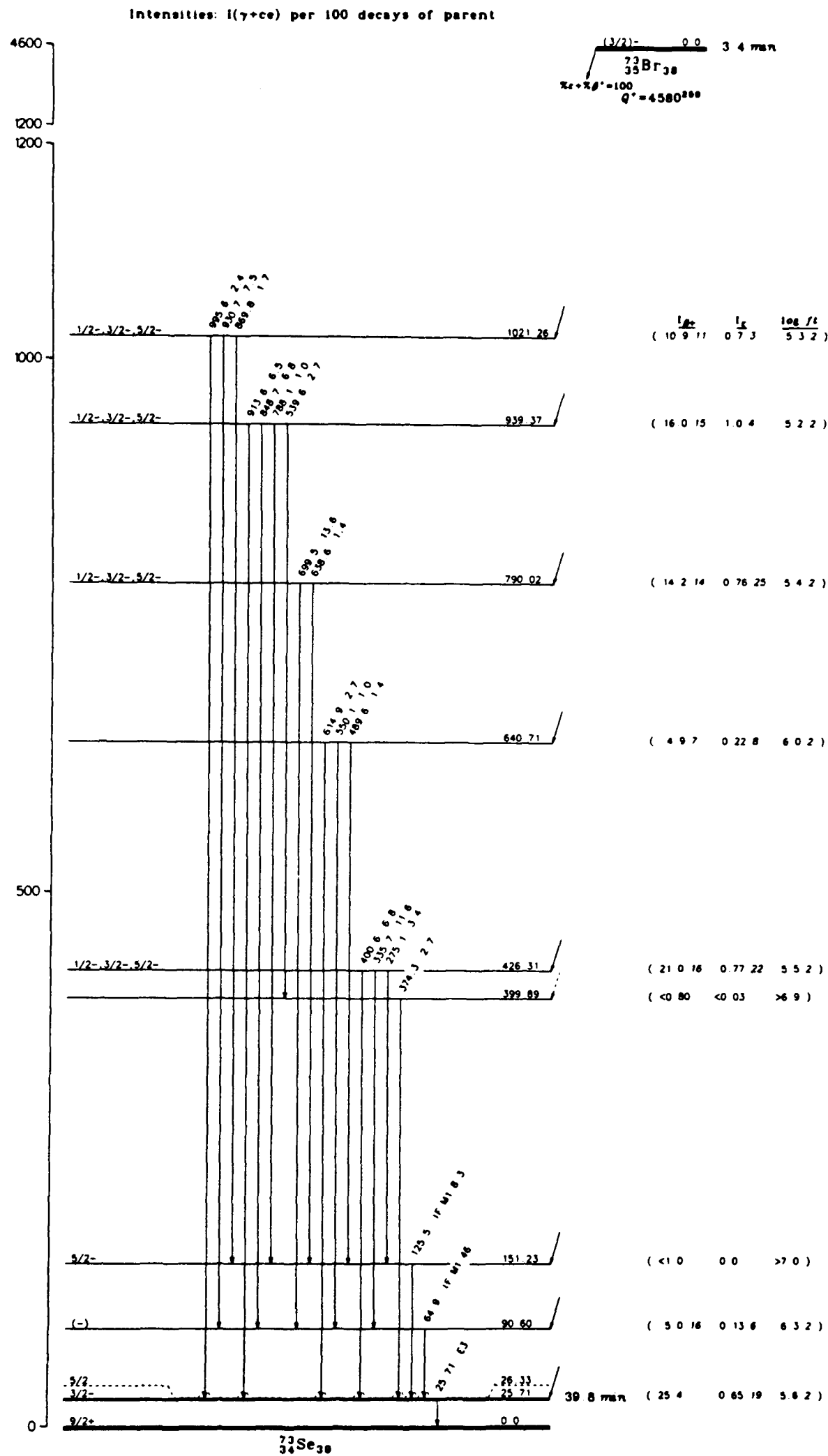


Figure 4.6. The decay scheme of ^{73}Br [14].

gamma transitions are likely to be of spin $1/2$. It is therefore most probable that at least a few of the relevant levels in ^{73}Se should be capable of orientation. In principle the lack of orientation might also be due to unfortunate values of the gamma multipole mixing ratios which lead to near zero angular distribution coefficients. This possibility is also inherently unlikely to be simultaneously true for several transitions. Two levels in ^{73}Se (other than the ground state) are in fact known to have spins other than $1/2$ [14]. Unfortunately neither figure prominently in this work. The 151 keV $5/2^-$ level gives rise only to the 126 keV transition which lies between the 122 and 136 keV peaks of the thermometer, $^{57}\text{CoFe}$. Although the triplet is resolved, the background is discontinuous making its subtraction difficult and ambiguous. However, a crude manual background subtraction showed no significant anisotropy for this transition. The second level at 26 keV ($3/2^-$, 40 m) is isomeric and therefore orients in its own right.

One concludes therefore that it is in fact the parent nucleus, ^{73}Br , which has a ground state spin $1/2^-$. Although such an assignment differs from the spin $3/2^-$ ground states of all the heavier odd isotopes up to ^{87}Br , it is fully consistent with all known features of the decay.

4.4.4 The ground state and isomer $^{72}\text{Br}^{g,m}$

Both the ground and isomeric states of ^{72}Br were produced, if rather weakly, in the heavy ion reaction and gave small, but nevertheless non-zero, anisotropies. Part of the reason for the modest anisotropies lies in the fact that the half-lives of the ground and isomeric states, 1.3 m and 10.1 s respectively, are not significantly larger than the spin-lattice relaxation times.

Herzog *et al.* [6], by assuming a single exponential recovery of the anisotropy after a period of resonance, obtained a spin-lattice relaxation time constant [17] of 19.2(14) s for $^{82}\text{BrFe}$ at a temperature of 7.3(1) mK. This enables the value $(\mu/I)^2 C_k = 0.032(2)$ to be determined from figure 1.5 (which is appropriate for the oriented spin of ^{82}Br). This is an isotope independent quantity which allows a self consistent calculation to be performed on the data of ^{72}Br taking into account the effects of imperfect relaxation on the observed anisotropies.

Firstly the β^+/EC decay of $^{72}\text{Br}^g$, illustrated in figure 5.5 [18], will be considered. The spin parity of this state has been assigned $(3)^+$ [19]. A brief review of the underlying arguments leading to this assignment will be given in section 5.7. Unfortunately this could not be confirmed by the particle-rotor calculations of subsection 7.5.3.

As a result of the weak anisotropies only the strongest line in the decay, at 862 keV, yielded a reasonable temperature dependence. This is illustrated in figure 4.7. As in the case of $^{74}\text{Br}^m$ it is in principle necessary to consider terms of fourth rank. As a first approximation it is assumed

that only the second rank orientation parameters associated with the full substitutional hyperfine field need be considered. In this limit, where the low field orientation is ignored, model 2 becomes formally identical to model 1. Thus from a model 1 fit to the data a moment of $0.56(30)\mu_N$ is obtained.

Further, the U_2A_2 coefficient of the 775 keV $(3)^+ \rightarrow 4^+ \rightarrow 2^+$ transition is calculated to be -0.41 . Therefore from the average anisotropy of this transition, $-6.9(45)\%$ at 15 mK (see table 5.6), one obtains $fB_2(B_{sub}) = 0.14(9)$. This is corrected for the $\sim 15\%$ attenuation of orientation at this temperature due to relaxation effects. Had data of better statistical accuracy been available it would have been possible to extract the site distribution parameter from this result. As it is, a value $f = 67(9)\%$ is assumed (see section 4.5) which leads to a moment of $0.54(28)\mu_N$. This is in satisfactory agreement with the value obtained from the fit to the temperature dependence of the 862 keV transition. However, with due regard for the size of the errors, this gives no independent verification of the assumptions that $f = 67(9)\%$ nor that $I^\pi(^{72}\text{Br}^g) = 3^+$. Averaging the two results leads to a moment of $0.55(21)\mu_N$.

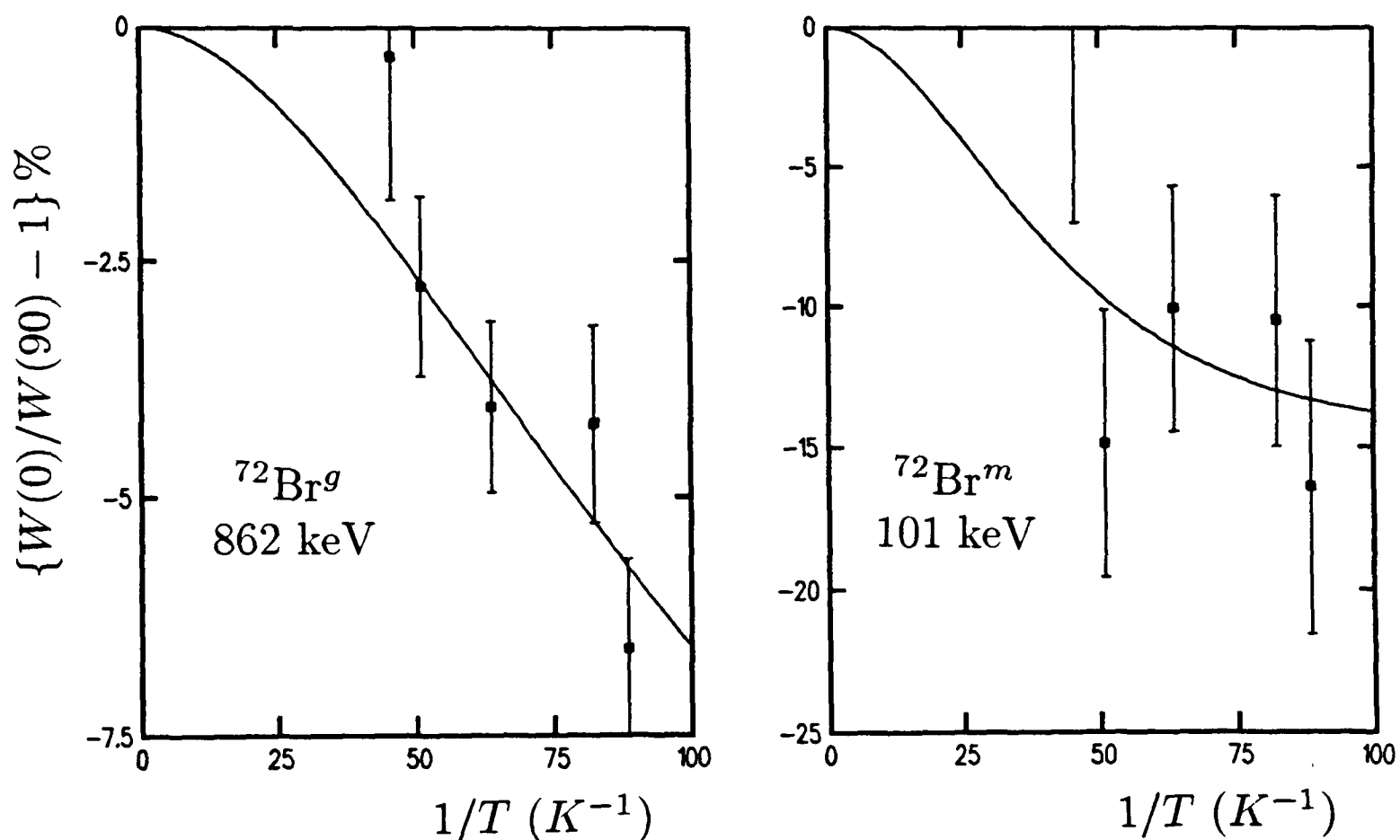


Figure 4.7. The temperature dependences of the 862 keV and 101 keV transitions in the decays of $^{72}\text{Br}^g$ and $^{72}\text{Br}^m$ respectively. The fitted model 1 curves are shown.

With this moment the ratio of the high field orientation parameters at the average temperature of 15 mK is calculated to be $B_4(B_{sub})/B_2(B_{sub}) \sim 2\%$. Thus the neglect of the fourth order orientation parameters is justified *a posteriori*. The model dependence of this result is

determined by the ratio $B_2(B_{low})/B_2(B_{sub}) \sim 15\%$. Since this is well within the statistical error the final moment of $^{72}\text{Br}^g$ is taken to be $0.55(21)\mu_N$.

Consider now the 101 keV transition from the 10.1 s isomeric state to the ground state. The $(1)^-$ spin parity assignment ^[18,19] was made on the basis of an assumed M2 multipolarity for this transition. This has been confirmed in the present work by means of an electron conversion measurement (see section 5.8).

Returning to the LTNO data, due to difficulties in subtracting the photopeak from the large Compton background at 101 keV the form of the temperature dependence was relatively poor. A model 1 fit to this data, which is illustrated in figure 4.6, results in a fitted moment of $1.3(^{+10.0}_{-0.6})\mu_N$. As before the model dependence is well within the experimental error.

Finally, it should be pointed out that as a result of the partial orientation of the isomeric state the ground state is not produced with randomly populated sublevels, contrary to the assumptions of the relaxation model based on (1.99). From the intensity ratio of the 101 keV to 862 keV gamma rays it can be deduced that only 20(1)% of the total feed to the ground state comes from the isomer. (This has been corrected for detection efficiency by comparison with the ratio of the 65 to 849 keV gamma rays in the decay of ^{73}Br which were measured in the same experiment). Since at 15 mK the isomer only ever achieves $\sim 60\%$ of its equilibrium orientation before decay, it can be concluded that only a small fraction of the ensemble ground states are modestly pre-oriented in this way. In relation to the large experimental errors this constitutes a negligible effect.

4.5 Conclusion

LTNO measurements on the light bromine nuclei have yielded the nuclear magnetic dipole moments of $^{72g,72m,74m,75,76,77}\text{Br}$. Apart from ^{76}Br none of these have been previously measured. In addition the ground state spin of ^{73}Br has been established as $1/2^-$.

The experimental data has been analysed within the framework of the two models described in sections 4.2 and 4.4. The results of this analysis are summarised in table 4.6. It has been shown, both here for the case of ^{76}Br and by Herzog *et al.* for the case of ^{82}Br , that model 1 fails to predict the correct moment. Therefore it is necessary to adopt the more flexible model 2. This requires an extra parameter, B_{low} , which has been deduced by Herzog *et al.* and also causes f and $U_\lambda A_\lambda$ to become independent. Since the $U_\lambda A_\lambda$ coefficients are generally not known

Table 4.6. A summary of the fitted moments of ^{72–77}Br.

Nuclide	Fitted Moment (μ _N)			Adopted Moment (μ _N)
	Model 1	Model 2a (f=75%)	Model 2b (f=58%)	
⁷⁷ Br	0.82(2)	—	0.92(5)	±0.92(5)
⁷⁶ Br	0.43(3)	—	0.55	±0.55 [10]
⁷⁵ Br	0.73(9)	0.75(15)	0.78(17)	±0.76(18)
⁷⁴ Br ^m	1.63(6)	1.59(8)	1.77(11)	±1.68(18)
⁷² Br ^g	0.55(21)	0.55(21)	0.55(21)	±0.55(21)
⁷² Br ^m	> 0.7	> 0.7	> 0.7	μ > 0.7

a priori for the isotopes studied, a limited range of values for *f* has been assumed in order to reduce the number of free parameters to be extracted from the experimental data.

For ⁷⁷Br, *f* is taken to be the same as that deduced from the data on the control isotope ⁷⁶Br since the two were produced under similar conditions. For that part of the data on ⁷⁷Br collected using the demagnetization stage the iron foil was treated in a different manner leading to a somewhat different site distribution. The parameter *f* was therefore unknown *a priori* and so the data is unuseable in terms of extracting a magnetic moment, although it is still useful in the calculation of *U*₂*A*₂ coefficients (see subsection 5.2). For ^{72–75}Br no such control exists and it is necessary to adopt a range of likely values for *f*, taken to be 75% ≥ *f* ≥ 58% (models 2a and 2b respectively). It has been shown that within this range the model 2 extracted moments have only a limited sensitivity to the value of *f*.

In addition to this it is in principle necessary to also consider the effects of a variable low field *B*_{low} since this need not correspond to a well defined field but, instead, to some average over many fields. However, the work of Herzog [7] has given some evidence that *B*_{low} varies by no more than 2 T between different warm or cold implantations. The value of *f* on the other hand varied by 20% between warm and cold implantations and so it is clear that this and not *B*_{low} should be treated as the variable parameter.

In view of the proven inability of model 1 to correctly describe the experimental data the adopted moments are taken to be those derived from model 2. In the case of ^{74m,75}Br, where both models 2a and 2b have been applied, the mean of the two results is taken with errors chosen so as to encompass the range of values emerging from the two models. Effectively this corresponds to the assumption that *f* = 67(9)%. The resulting increase in the errors is relatively modest despite realistically reflecting the site distribution uncertainties. These adopted moments are also given in table 4.6. Clearly precise NMR/ON values would be of great value and this would be the natural direction for future work. Nonetheless these results allow clear nuclear structure deductions to be made in chapter 7.

References

- [1] . R. Bengtsson *et al.*, Phys. Scripta **29** (1984) 402
- [2] . W. Nazarewicz *et al.*, Nucl. Phys. **A435** (1985) 397
- [3] . L. Luhmann *et al.*, Phys. Rev. **C31** (1985) 828
- [4] . K.P. Lieb and J.J. Kolata, Phys. Rev. **C15** (1977) 939
- [5] . R.B. Piercey *et al.*, Phys. Rev. **C19** (1979) 1344
- [6] . P. Herzog *et al.*, Z. Phys. **B64** (1986) 353
- [7] . P. Herzog *et al.*, Nucl. Instr. and Meth. **B26** (1987) 471
- [8] . R.B. Alexander *et al.*, Phys. Rev. **B9** (1974) 3022
- [9] . P.T. Callaghan *et al.*, Hyp. Int. **3** (1977) 267
- [10] . C.M. Lederer and V.S. Shirley, Table Of Isotopes, (Wiley, 1978)
- [11] . Nuclear Data Sheets **42** (1984)
- [12] . Nuclear Data Sheets **29** (1980)
- [13] . Nuclear Data Sheets **32** (1981)
- [14] . Nuclear Data Sheets **51** (1987)
- [15] . C. Ekstrom and L. Robertsson, Phys. Scripta **22** (1980) 344
- [16] . K. Heiguchi *et al.*, Nucl. Phys. **A474** (1987) 484
- [17] . P.K. James *et al.*, Phys. Rev. **B13** (1976) 59
- [18] . Nuclear Data Sheets **56** (1989)
- [19] . G. Garcia-Bermudez *et al.*, Phys. Rev. **C25** (1982) 1396

Chapter 5

The spectroscopy of the light selenium isotopes

5.1 Introduction

This chapter will deal with that spectroscopic information on the daughter selenium isotopes which can be deduced from the LTNO data on the decay of the light bromine parents.

Firstly those quantities will be defined which are relevant to this analysis. The $U_\lambda A_\lambda$ coefficients are related to the axial to equatorial anisotropy ratio, E , via the expression (3.9)

$$E = \frac{W(0)}{W(\frac{\pi}{2})} - 1 = \frac{\sum_j \{N/p\}_j(0)}{\sum_k \{N/p\}_k(0)} \times \frac{\sum_k \{N/p\}_k(\frac{\pi}{2})}{\sum_j \{N/p\}_j(\frac{\pi}{2})} - 1$$

$$= \frac{\sum_{\lambda i} f_i B_\lambda(B_{hyp}^i) U_\lambda A_\lambda Q_\lambda P_\lambda(1)}{\sum_{\lambda i} f_i B_\lambda(B_{hyp}^i) U_\lambda A_\lambda Q_\lambda P_\lambda(0)} - 1. \quad (5.1)$$

In order to improve statistics, those spectra corresponding to the lower temperatures attained for each isotope are included in the summation over the index j . The quoted anisotropies therefore correspond to some average of the low temperature data points presented in chapter 4. The factor $\sum_{\lambda i} f_i B_\lambda(B_{hyp}^i)$ is calculated within the framework of model 2 using the adopted moments of table 4.6 in conjunction with the two hyperfine fields, B_{sub} and B_{low} and their associated populations f and $1 - f$, as summarised in section 4.5. The solid angle correction factors Q_λ are calculated individually for each detector and gamma ray energy.

For those parent nuclei whose oriented spin is less than 2, the fourth and higher rank terms disappear so that (5.1) may be inverted to yield the $U_2 A_2$ terms directly

$$U_2 A_2 = \frac{1}{Q_2 \sum_i f_i B_2(B_{hyp}^i)} \times \left\{ \frac{2E}{E+3} \right\}. \quad (5.2)$$

Further, where two or more gamma transitions arise from the same level, then by using (5.2) and the fact that the de-orientation coefficients are identical

$$A(\gamma_m : \gamma_n) = \frac{A_2(\gamma_m)}{A_2(\gamma_n)}$$

$$= \frac{E_m(E_n + 3)}{E_n(E_m + 3)}. \quad (5.3)$$

This ratio is independent of any site distribution or hyperfine field assumptions and its error arises solely from uncertainties in the measured anisotropy.

In those cases where the oriented spin is greater than or equal to 2, namely $^{74}\text{Br}^m$ and $^{72}\text{Br}^g$, fourth rank terms must also be considered. In the former case the U_2A_2 and U_4A_4 coefficients are extracted separately from a least squares fit to the temperature dependent data. For the latter isotope it has been shown that at the temperatures achieved experimentally the fourth rank terms are negligible so that both (5.2) and (5.3) may be applied.

Finally, the errors on all quantities are obtained from the standard propagation expression

$$\Delta F^2 = \sum_p \left\{ \frac{\partial F}{\partial x_p} \Delta x_p \right\}^2. \quad (5.4)$$

5.2 The isotope ^{77}Se

As was noted in section 4.3, the decay of ^{77}Br was studied using both the dilution unit alone and latterly in conjunction with the PrNi_5 demagnetization stage. The averaged anisotropies, taken from those spectra corresponding to the lowest temperatures, for both parts of the experiment are listed in table 5.1. For the former experiment these anisotropies correspond to a temperature of 9.8 mK and for the latter 2.5 mK. Taking the site distribution parameters to be $f = 58(9)\%$ and $78(9)\%$ for the two parts of the experiment results in the values $\sum fB_2 = 0.48(5)$ and $0.96(2)$ respectively. The U_2A_2 coefficients extracted from the two essentially independent sets of data are in extremely good agreement, giving a clear indication of the basic soundness of the assumed site distribution model and the reliability of the resulting $\sum fB_2$ values. These coefficients are also given, together with their mean values, in table 5.1.

The decay scheme of ^{77}Br , shown in figure 5.1, is taken from Nuclear Data Sheets [1]. This includes a number of E2/M1 mixing ratios † which are mostly from the work of Braga *et al.* [2]. A few independent mixing ratios are also given by Mohsen *et al.* [3], Robinson *et al.* [4] and Zell *et al.* [5]. Mixing ratios derived from the present results which are not consistent with previous electron conversion measurements by Tokunaga *et al.* [6] are enclosed thus [].

1005 keV level : $3/2^-$

The mixing ratio of the 755 keV transition has been measured by both Braga *et al.* and more recently by Mohsen *et al.* to be $\delta(755) = +0.31^{+8}_{-9}$ and $+0.30(1)$ respectively. These

† Unless otherwise stated all electromagnetic mixing ratios δ refer to E2/M1 multipoles.

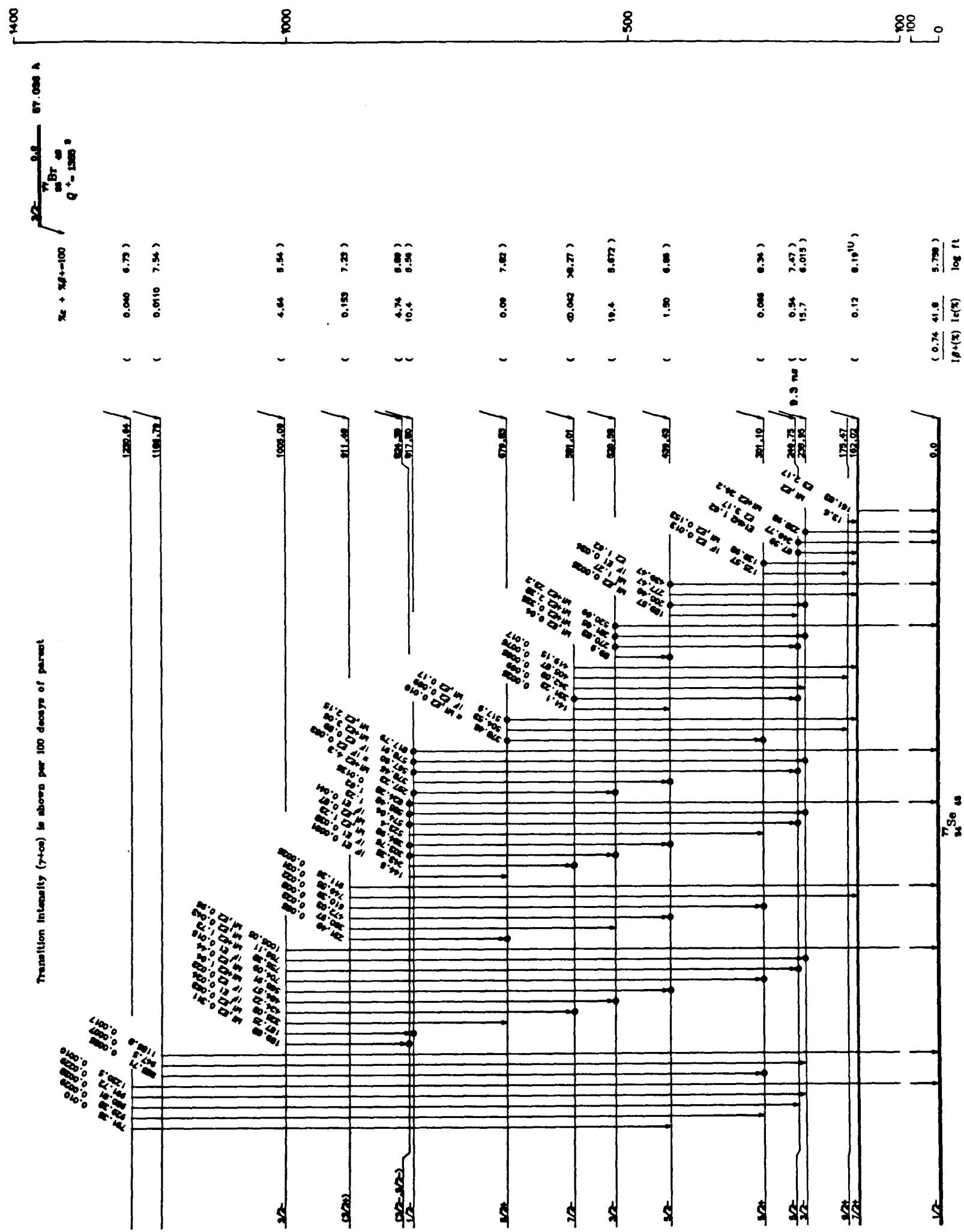


Figure 5.1. The decay scheme of ^{77}Br [1].

Table 5.1. The axial/equatorial anisotropies and U_2A_2 coefficients observed in the decay of ^{77}Br at 9.8 mK and 2.5 mK. * indicates that the error is multiplied by 10 and † indicates a doublet.

Energy(keV)	E (Unit) (%)	U_2A_2	E (Demag) (%)	U_2A_2	mean U_2A_2
88	− 1.2(1.5)	−0.02(2)	0.8(5.6)	0.01(4)	−0.01(2)
181	− 8.6(5.1)	−0.13(8)	− 40.8(16.7)	−0.34(16)	−0.17(7)
200	6.9(1.2)	0.10(2)	11.8(4.8)	0.08(3)	0.09(2)
239	1.7(0.2)	0.02(0)	4.5(0.5)	0.03(0)	0.03(*2)
250	− 5.4(0.5)	−0.08(1)	− 8.5(1.6)	−0.06(1)	−0.07(1)
282	− 1.5(0.6)	−0.02(1)	− 0.6(2.0)	−0.00(1)	−0.02(1)
297	0.3(0.4)	0.00(1)	1.9(1.3)	0.01(1)	0.01(*4)
304	42.9(1.4)	0.55(6)	106.7(8.5)	0.57(4)	0.57(3)
379†	−11.9(8.9)	−0.18(14)	− 22.9(35.7)	−0.18(31)	−0.18(13)
385	−21.3(1.0)	−0.34(4)	− 44.3(3.1)	−0.38(3)	−0.36(2)
439	−15.4(0.6)	−0.24(3)	− 33.7(1.8)	−0.28(2)	−0.27(1)
485	− 8.1(0.8)	−0.12(2)	− 15.7(3.0)	−0.12(2)	−0.12(1)
521	3.1(0.2)	0.05(1)	7.1(0.5)	0.05(0)	0.05(*3)
568†	0.4(0.6)	0.01(1)	− 2.5(2.6)	−0.02(2)	0.00(1)
575	−24.0(0.5)	−0.38(4)	− 46.5(1.5)	−0.40(2)	−0.40(2)
579	0.2(0.4)	0.00(1)	− 0.6(1.3)	−0.00(1)	0.00(*5)
585	19.0(0.7)	0.26(3)	36.4(2.9)	0.24(2)	0.24(2)
755	6.3(0.6)	0.09(1)	15.3(3.2)	0.11(2)	0.09(1)
766	−11.0(9.1)	−0.17(15)	25.4(56.0)	0.17(35)	−0.12(13)
818	− 0.4(0.5)	−0.01(1)	− 5.3(2.2)	−0.04(2)	−0.01(*6)
911	0.3(6.9)	0.00(10)	− 11.9(13.6)	−0.09(11)	−0.04(7)
1005	− 4.6(0.9)	−0.07(2)	− 15.2(4.6)	−0.12(4)	−0.08(1)

predict an angular distribution coefficient of $A_2(755) = +0.45(1)$ and thence from the measured U_2A_2 value one obtains $U_2(1005) = +0.20(2)$. This is exactly consistent with a pure G-T ($3/2^- \rightarrow 3/2^- : U_2 = +0.20$) β^+/EC transition to this level. Assuming this to be the case, then

$A_2(1005) = -0.40(5) \qquad 3/2^- \rightarrow 1/2^- \quad \delta(1005) = +0.50^{+4}_{-3} \quad \text{or} \quad -18^{+6}_{-17}$ $A_2(766) = -0.60(65) \qquad 3/2^- \rightarrow 3/2^- \quad \delta(766) \geq -0.29$ $A_2(755) = +0.45(5) \qquad 3/2^- \rightarrow 5/2^- \quad \delta(755) = +0.30^{+5}_{-4}$ $A_2(485) = -0.60(5) \qquad 3/2^- \rightarrow 3/2^- \quad \delta(485) = +0.14^{+4}_{-4} \quad \text{or} \quad \left[+2.4^{+3}_{-2} \right]$ $A_2(181) = -0.85(35) \quad \text{if} \quad 3/2^- \rightarrow 3/2^- \quad \delta(181) = +0.07 \rightarrow +3.0$ $\qquad \qquad \qquad \text{if} \quad 3/2^- \rightarrow 5/2^- \quad A_2 \geq -0.38$

The results for the 181 keV transition will be discussed further under the 824 keV level heading.

911 keV level : $3/2^+$

The 911 keV pure E1 transition yields the result $U_2(911) = -0.08(14)$. This is to be compared with the theoretical de-orientation coefficients for pure $j_\beta = 0, 1, 2$ transitions of

$U_2 = +1.00, +0.20$ and -0.60 respectively. The experimental value clearly indicates that the lepton field is partly of quadrupole character. In the assumed absence of $j_\beta = 0$ the calculated lepton mixing ratio is $\Delta^2(j_\beta = 2/j_\beta = 1) = 0.54(41)$, while in the assumed absence of $j_\beta = 1$ a value of $\Delta^2(j_\beta = 2/j_\beta = 0) = 2.1(8)$ is obtained. Thus in the presence of all possible first forbidden components $0.13 \leq \Delta^2(j_\beta = 2/j_\beta = 0, 1) \leq 2.9$.

824 keV level : $(3/2^-, 5/2^-)$

The spin and parity assignments of this level were established on the basis of angular correlation measurements [2] and transition strengths respectively [1].

The possibility of positive parity can be ruled out by the present measurements since this would imply that the 304 and 585 keV transitions are pure E1 ($3/2^+, 5/2^+ \rightarrow 3/2^-$) and therefore must have identical $U_2 A_2$ values. From the results of table 5.1 this is clearly not the case.

Turning now to the question of the level spin, both possibilities have been considered in the calculation of the mixing ratio of the 181 keV transition from the 1005 keV level. The large negative angular distribution coefficient of this transition, $A_2 = -0.85(35)$, is inconsistent with a $5/2^-$ spin assignment for the 824 keV level, since then the theoretical minimum would be $A_2 = -0.38$. On this basis the 824 keV level spin is temporarily taken to be $3/2^-$. With this assumption, from the $U_2 A_2$ coefficient of the 304 keV transition ($3/2^- \rightarrow 3/2^- : A_2 \leq +0.60$) the de-orientation parameter for this level is calculated to be $U_2(824) = +0.95(5)$. The level feeding is 94% direct β^+/EC and 6% via the 181 keV transition. From the above values of $U_2(1005)$ and $\delta(181)$ a partial de-orientation coefficient of $-0.08 \leq U_2 \leq +0.03$ can be calculated for the gamma feed. Since the F and G-T components of the allowed β^+/EC feed have $U_2 = +1.00$ and $+0.20$ respectively, a purely F lepton field results in a calculated total de-orientation parameter of $+0.93 \leq U_2(824) \leq +0.94$, in excellent agreement with the experimental data. Ignoring therefore any possible small G-T admixtures the following results are obtained

$$A_2(575) = -0.43(2) \quad \text{if} \quad 3/2^- \rightarrow 5/2^- \quad A_2 \geq -0.38$$

$$A_2(304) = +0.61(3) \quad \text{if} \quad 3/2^- \rightarrow 3/2^- \quad \delta(304) = [-1.6 \rightarrow -1.0]$$

These results present a number of inconsistencies. Firstly, since ^{77}Br is not a mirror nucleus the F matrix elements of the β^+/EC decay are likely to be hindered by the $\Delta T = 0$ isospin selection rule. Since a similar decay to the 1005 keV level has already been shown to be essentially pure G-T, a pure F decay to the present level is unlikely. Secondly, the present mixing ratio for the 304 keV transition is not predicted to be of mainly M1 character as required by the electron conversion measurements of Tokunaga *et al.* Finally, the theoretical angular distribution

coefficient for the 575 keV transition has as its lower limit the value $A_2 = -0.38$. However, the experimental value of $A_2 = -0.43(2)$ lies outside this range. In fact the discrepancy ($\sim 2.5\sigma$) is greater than that of the 181 keV transition ($\sim 1.3\sigma$) upon which the presently assumed level assignment is based.

For these reasons it would appear that a 824 keV level spin of $3/2^-$ is not compatible with the present data and that the measured anisotropy of the 181 keV transition is one of the 20% of observations which are expected exceed their true values by more than 1.3σ . The spin of the 824 keV level is therefore taken to be $5/2^-$. Such an assignment is supported by the fact that this level is only weakly populated in the (n, γ) reaction [7] and the tentatively placed transition [6] to the 162 keV $7/2^+$ level.

With this level assignment the β^+/EC feed is now pure G-T which, with the inclusion of 181 keV gamma feed, leads to a level de-orientation coefficient of $U_2(824) = +0.71$. The resulting mixing ratios are given below

$$\begin{aligned} A_2(585) &= +0.34(3) \quad \text{if } 5/2^- \rightarrow 3/2^- \quad \delta(585) = +0.03_{-2}^{+1} \quad \text{or} \quad -3.7_{-3}^{+3} \\ A_2(575) &= -0.56(3) \quad \text{if } 5/2^- \rightarrow 5/2^- \quad \delta(575) = +0.11_{-4}^{+4} \quad \text{or} \quad +1.3_{-1}^{+1} \\ A_2(385) &= -0.51(3) \quad \text{if } 5/2^- \rightarrow 5/2^- \quad \delta(385) = +0.05_{-3}^{+3} \quad \text{or} \quad +1.5_{-1}^{+1} \\ A_2(304) &= +0.80(4) \quad \text{if } 5/2^- \rightarrow 3/2^- \quad \delta(304) = -0.23_{-3}^{+3} \quad \text{or} \quad [-1.8_{-1}^{+1}] \end{aligned}$$

All of the experimental angular distribution coefficients are now within their corresponding theoretical limits and the former of the two predicted mixing ratios for the 304 keV transition is consistent with the work of Tokunaga *et al.*

521 keV level : $3/2^-$

Using the known feeding intensities to this level it is possible to deduce that $+0.12 \leq U_2(521) \leq +0.77$. Thus

$$\begin{aligned} +0.06 \leq A_2(521) \leq +0.42 \quad & 3/2^- \rightarrow 1/2^- \quad \delta(521) = +0.05 \rightarrow +0.24 \quad \text{or} \quad [-3.4 \rightarrow -1.9] \\ -0.25 \leq A_2(282) \leq -0.01 \quad & 3/2^- \rightarrow 3/2^- \quad \delta(282) = -0.25 \rightarrow -0.10 \quad \text{or} \quad \geq [+6.3] \end{aligned}$$

439 keV level : $5/2^-$

From the angular distribution coefficient of the 439 keV E2 transition a level de-orientation factor of $U_2(439) = +0.51(2)$ can be calculated. Thus from the ratio $A_2(200 : 439) = -0.39(6)$

$$A_2(200) = +0.21(3) \quad 5/2^- \rightarrow 3/2^- \quad \delta(200) = +0.09_{-2}^{+2} \quad \text{or} \quad [-4.8_{-4}^{+4}]$$

239 keV level : $3/2^-$

Drawing on the present results for higher lying levels from which the feeding gamma rays originate enables a theoretical de-orientation coefficient in the range $+0.09 \leq U_2(239) \leq +0.72$ to be calculated. Thus

$$+0.04 \leq A_2(239) \leq +0.33 \quad 3/2^- \rightarrow 1/2^- \quad \delta(239) = +0.09 \rightarrow +0.25 \quad \text{or} \quad [-3.5 \rightarrow -2.2]$$

Discussion

With the exception of the 755 keV transition the mixing ratios obtained in this work disagree entirely with those of Braga *et al.* which have been adopted by Nuclear Data Sheets. Independent mixing ratios are available only for the 200, 239 ^[4,5] and 755 ^[3] keV transitions. The various experimental values obtained for 200 and 239 keV mixing ratios are tabulated below. The experimental methods are indicated.

Energy (keV)	Present $\gamma(\theta, H, T)$	NDS ^[1]	Braga ^[2] $\gamma\gamma(\theta)$	Robinson ^[4] Coul. ex. $\gamma(\theta)$	Zell ^[5] $^{74}\text{Ge}(\alpha, n\gamma) \gamma(\theta)$
200	+0.09(2)	-0.094(3)	+0.00(9)	+0.05(3)	+0.094(3)
239	+0.09 $\rightarrow$ +0.25	-0.184(30)	-0.13(4)	+0.18(3)	+0.188(30)

Braga *et al.* claims agreement with Robinson *et al.* on the value of $\delta(239)$. However the former work uses the convention of Rose and Brink ^[8] while the latter uses that of Biedenharn and Rose ^[9]. Since the 239 keV gamma ray is the *de-orienting* transition in the Coulomb excitation experiment of Robinson *et al.* and the mixing is such that $L' = L + 1$, these conventions differ in sign. Therefore the apparent agreement in the quoted values is in fact false. The present work uses the convention of Krane and Steffen ^[10] which in this case is identical to that of Biedenharn and Rose. Therefore the values of Braga *et al.* and Nuclear Data Sheets are in disagreement with both the present results and those of Robinson *et al.* and Zell *et al.* (The values tabulated above are all expressed in the convention of Krane and Steffen).

To illustrate these discrepancies further, the possibility that the present U_2A_2 coefficients are erroneous as a result of assuming incorrect values for $\sum fB_2$ will now be investigated. Thus, for example, the 239 keV mixing ratio of Braga *et al.* predicts an angular distribution coefficient of $A_2 = +0.70(5)$ as compared with the present value of $+0.04 \leq A_2 \leq +0.33$. In order to achieve agreement the effective orientation parameters $\sum fB_2$ would have to be *reduced* by a factor of at least 2. On the other hand, the 304 keV mixing ratio of Braga *et al.*, $\delta(304) = -0.09^{+4}_{-5}$, predicts

that $A_2 = +0.54(9)$. With the present experimental value of $A_2 = +0.80(4)$ consistency would require an *increase* of the effective orientation parameters by a factor of 1.5. Since the effective orientation parameter for the demagnetization part of the experiment is virtually at the limiting value of unity, no such increase is possible. By applying these arguments to other transitions it is found that no common value for the required correction factor exists. Thus the discrepancies in the mixing ratios are clearly *not* a result of the factors $\sum fB_2$ being incorrect.

Conversely, a comparison of the A_{22} angular correlation coefficients predicted by the present results and the experimental values of Braga *et al.* are given below. For the sake of argument both the $3/2^-$ and $5/2^-$ spin assignments for the 824 keV level are considered, even though the former has been excluded by the present results.

Energy	Spin Sequence	predicted A_{22}	observed A_{22} [2]
755 – 250	$3/2 \rightarrow 5/2 \rightarrow 1/2$	$-0.46 \rightarrow -0.45$	$-0.460(27)$
575 – 250	if $3/2 \rightarrow 5/2 \rightarrow 1/2$ if $5/2 \rightarrow 5/2 \rightarrow 1/2$	$\sim +0.41$ $+0.14 \rightarrow +0.19$ or $-0.25 \rightarrow -0.23$	$+0.038(27)$
385 – 440	if $3/2 \rightarrow 5/2 \rightarrow 1/2$ if $5/2 \rightarrow 5/2 \rightarrow 1/2$	$+0.34 \rightarrow +0.46$ $+0.18 \rightarrow +0.22$ or $-0.25 \rightarrow -0.24$	$+0.086(14)$
385 – 200	if $3/2 \rightarrow 5/2 \rightarrow 3/2$ if $5/2 \rightarrow 5/2 \rightarrow 3/2$	$-0.21 \rightarrow -0.12$ $-0.10 \rightarrow -0.06$ or $+0.08 \rightarrow +0.11$	$-0.100(26)$
200 – 239	$5/2 \rightarrow 3/2 \rightarrow 1/2$	$-0.01 \rightarrow +0.00$	$+0.033(13)$
282 – 239	$3/2 \rightarrow 3/2 \rightarrow 1/2$	$-0.24 \rightarrow -0.02$	$-0.153(3)$
304 – 521	if $3/2 \rightarrow 3/2 \rightarrow 1/2$ if $5/2 \rightarrow 3/2 \rightarrow 1/2$	$-0.41 \rightarrow -0.05$ $+0.02 \rightarrow +0.17$	$+0.162(19)$
485 – 521	$3/2 \rightarrow 3/2 \rightarrow 1/2$	$-0.10 \rightarrow -0.01$	$+0.011(10)$

It is interesting to note that there is only disagreement between prediction and experiment where the experimental A_{22} value is small. It is possible therefore that there are small, undetected errors in the work of Braga *et al.* which by propagating through their analysis have lead to an incorrect interpretation of the angular correlation data. The universal agreement on the 755 keV mixing ratio can then be attributed to the fact that the experimental A_{22} value is both large *and* the 755 keV gamma ray is taken in coincidence with the 250 keV pure E2 transition. Therefore the only unknown to be extracted from the data is the 755 keV mixing ratio. This is in

contrast to the other cascades which exhibit large A_{22} coefficients. There both transitions in the cascade are mixed so that two unknown parameters are involved which requires the knowledge of a second experimental quantity, A_{44} . Since the experimental A_{44} coefficients are invariably small with large errors these two mixing ratios are not reliably extracted.

Finally, it should be noted that the predicted A_{22} coefficient for the $304 \rightarrow 521$ keV cascade, calculated using the present results, disagrees in sign with the experimental value of Braga *et al.* if the 824 keV level is taken to have spin $3/2^-$. Since the experimental A_{22} value is comparatively large it may be presumed that the fitting procedure employed by Braga *et al.* is relatively reliable. This may therefore be taken as further evidence for the present $5/2^-$ spin assignment of the 824 keV level.

To conclude this section on the spectroscopy of ^{77}Se , the mixing ratios derived from the present work are listed below. Only those values consistent with electron conversion measurements ^[1,6] are included. For comparison the values of Braga *et al.* are also tabulated.

Present	: Braga ^[2]
$\delta(1005) = +0.50^{+4}_{-3}$ or -18^{+6}_{-17}	
$\delta(766) \geq -0.29$	
$\delta(755) = +0.30^{+5}_{-4}$	: $+0.31^{+8}_{-9}$
$\delta(585) = +0.03^{+1}_{-2}$ or -3.7^{+3}_{-3}	
$\delta(575) = +0.11^{+4}_{-4}$ or $+1.3^{+1}_{-1}$	: $+0.33^{+9}_{-8}$
$\delta(521) = +0.05 \rightarrow +0.24$	: -0.17^{+7}_{-6}
$\delta(485) = +0.14^{+4}_{-4}$	: $+0.27^{+3}_{-4}$
$\delta(385) = +0.05^{+3}_{-3}$ or $+1.5^{+1}_{-1}$	: $+0.23^{+5}_{-7}$
$\delta(304) = -0.23^{+3}_{-3}$	: -0.09^{+4}_{-5}
$\delta(282) = -0.25 \rightarrow -0.10$	: $+0.12^{+4}_{-3}$
$\delta(239) = +0.09 \rightarrow +0.25$	: -0.13^{+4}_{-4}
$\delta(200) = +0.09^{+2}_{-2}$	: $+0.00^{+9}_{-9}$

5.3 The isotope ^{76}Se

Anisotropies were measured for 42 gamma transitions in the decay of ^{76}Br at an average temperature of 2.9 mK. The site distribution parameter f was measured to be 58(9)% (see section 4.3) leading to the value $\sum fB_2 = 0.59(3)$. The anisotropies and U_2A_2 coefficients thus deduced are listed in table 5.2. It is unfortunate that despite the full orientation achieved the anisotropies observed from this $I = 1$ nucleus were all rather small, limiting the precision of the extracted decay parameters. The decay scheme, illustrated in figure 5.2, is taken from Nuclear Data Sheets [11].

3971 keV level : $(1^+, 2^+)$

The presence of a transition direct to the 0^+ ground state and the observation of orientation limits the spin of this level to $1^\pm, 2^+$. The 1301 keV transition between this level and the 2670 keV (2^-) level has an observed U_2A_2 coefficient of $-0.69(23)$. The theoretical coefficients for the possible spin sequences are listed below

$$1^- \rightarrow 1^+ \rightarrow 2^- : U_2A_2 = -0.04 \rightarrow +0.07$$

$$1^- \rightarrow 1^- \rightarrow 2^- : U_2A_2 = -0.35 \rightarrow +0.71$$

$$1^- \rightarrow 2^+ \rightarrow 2^- : U_2A_2 = -0.25 \rightarrow +0.25$$

None of these are consistent with the observed coefficient to within 1.5σ . This brings into doubt the (2^-) spin assignment of the 2670 keV level. As will be discussed later, there is reason to believe that the spin of the 2670 keV level can also be assigned to $1^+, 2^+$ or 3^+ . The theoretical coefficients for these spin sequences are

$$1^- \rightarrow 1^+ \rightarrow 1^+ : U_2A_2 = -1.41 \rightarrow +0.71$$

$$1^- \rightarrow 1^- \rightarrow 1^+ : U_2A_2 = -0.35 \rightarrow +0.18$$

$$1^- \rightarrow 2^+ \rightarrow 1^+ : U_2A_2 = -0.56 \rightarrow +0.63$$

$$1^- \rightarrow 1^+ \rightarrow 2^+ : U_2A_2 = -0.35 \rightarrow +0.71$$

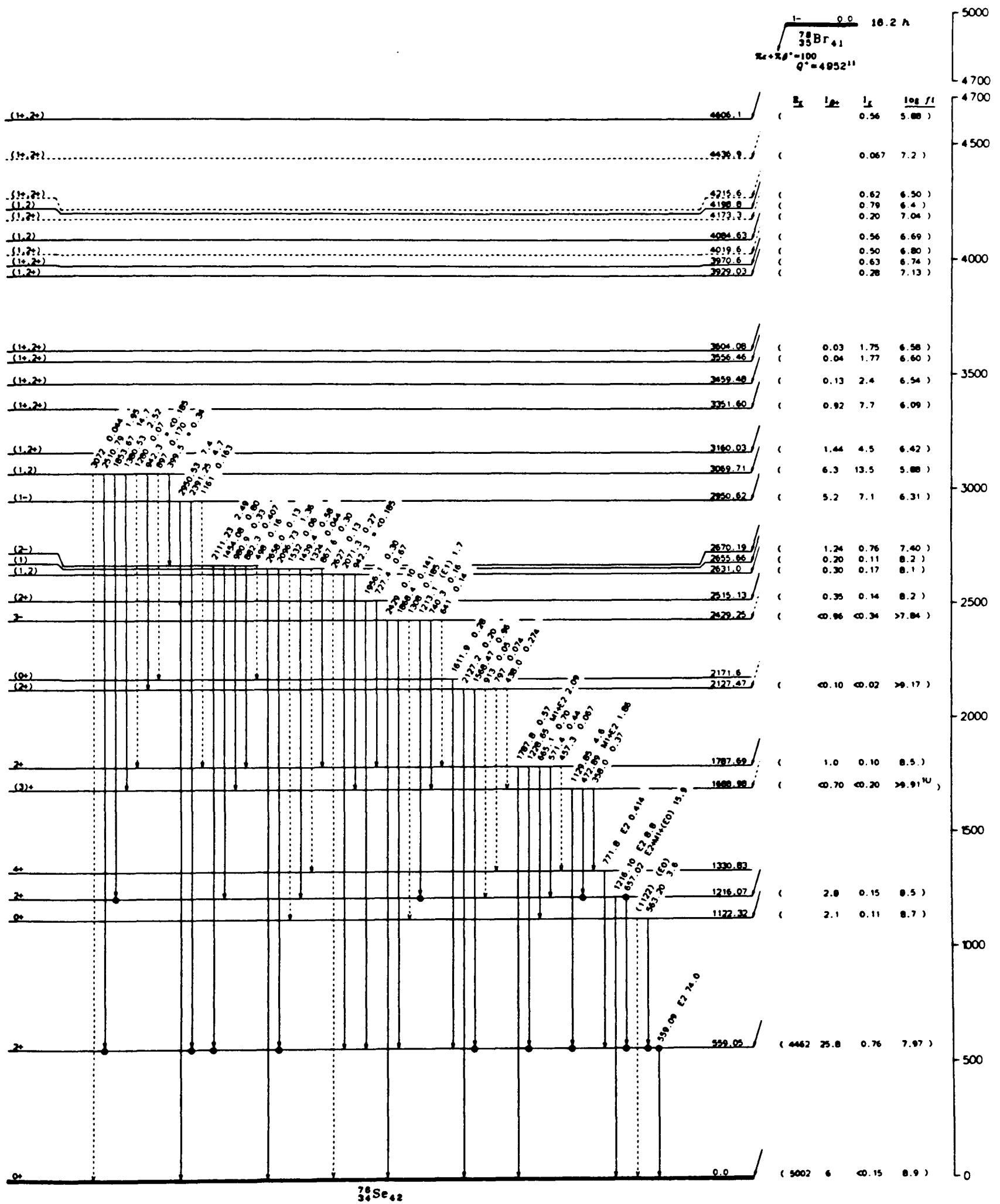
$$1^- \rightarrow 1^- \rightarrow 2^+ : U_2A_2 = -0.04 \rightarrow +0.07$$

$$1^- \rightarrow 2^+ \rightarrow 2^+ : U_2A_2 = -0.48 \rightarrow +0.48$$

$$1^- \rightarrow 1^+ \rightarrow 3^+ : U_2A_2 = -0.10 \rightarrow +0.05$$

$$1^- \rightarrow 1^- \rightarrow 3^+ : U_2A_2 = -0.35 \rightarrow +0.70$$

$$1^- \rightarrow 2^+ \rightarrow 3^+ : U_2A_2 = -0.53 \rightarrow +0.53$$

Figure 5.2. The decay scheme of ^{76}Br (Part 1 of 2) [11].

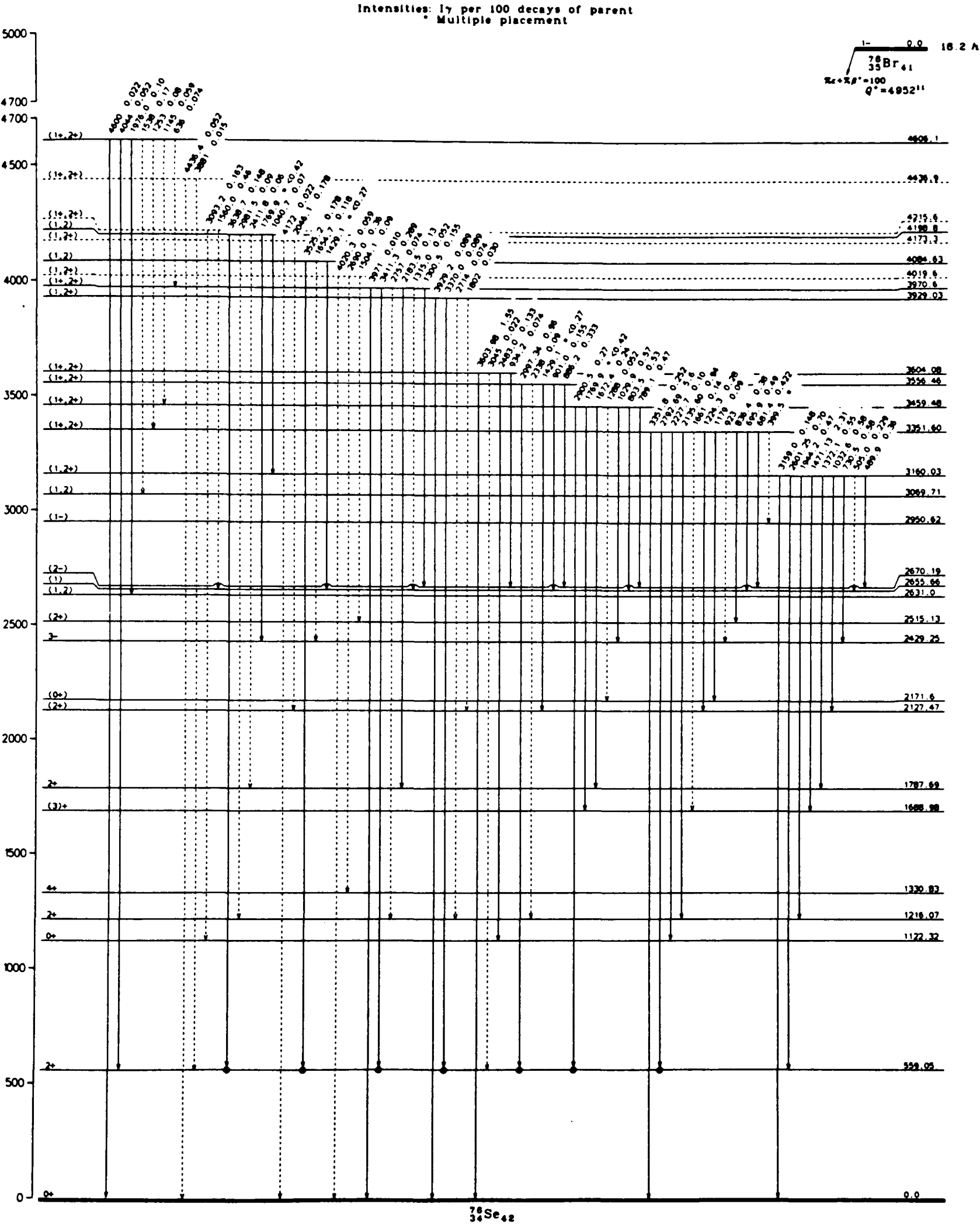


Figure 5.2. The decay scheme of ^{76}Br (Part 2 of 2) [11].

Table 5.2. The axial/equatorial anisotropies and U_2A_2 coefficients observed in the decay of ^{76}Br at 2.9 mK.

Energy (keV)	E (%)	U_2A_2
473	−9.2(1.8)	−0.11(2)
490	18.0(10.3)	0.20(10)
559	3.8(0.3)	0.04(1)
599	−1.5(4.6)	−0.01(5)
604	3.7(8.4)	0.04(9)
657	−0.6(0.4)	−0.01(5)
665	−0.8(3.7)	−0.01(4)
681	−8.8(4.8)	−0.10(6)
696	2.7(4.8)	0.03(5)
728	−1.1(4.7)	−0.01(5)
731	6.7(4.2)	0.07(4)
772	−1.0(6.1)	−0.01(7)
789	−6.2(5.6)	−0.07(6)
804	9.3(5.8)	0.10(6)
882	−5.1(5.8)	−0.06(7)
886	−17.5(7.7)	−0.22(10)
1030	1.6(6.7)	0.01(7)
1033	−13.4(6.1)	−0.16(8)
1130	−14.2(0.9)	−0.17(1)
1213	2.0(3.4)	0.02(3)
1216	−4.1(1.0)	−0.04(1)
1229	2.5(1.8)	0.02(2)
1301	−48.8(13.8)	−0.69(23)
1372	−13.7(6.4)	−0.17(8)
1381	4.8(1.3)	0.05(1)
1439	−5.7(4.2)	−0.06(5)
1454	−5.6(3.2)	−0.06(3)
1471	4.3(1.8)	0.05(2)
1560	−12.4(5.3)	−0.15(6)
1568	−4.2(4.2)	−0.05(5)
1672	−22.1(12.5)	−0.28(17)
1788	−0.9(5.6)	−0.01(6)
1854	−9.5(0.5)	−0.11(1)
1944	−12.1(6.9)	−0.15(8)
2097	2.1(2.8)	0.02(3)
2111	−3.1(1.6)	−0.03(1)
2136	−4.7(3.2)	−0.05(3)
2511	−10.6(1.8)	−0.13(2)
2601	−8.1(4.7)	−0.09(5)
2793	−2.0(0.9)	−0.02(1)
2901	−9.1(4.2)	−0.11(5)
2951	−12.9(0.7)	−0.16(1)

Thus the 1301 keV transition spin changes are limited to $1^+ \rightarrow 1^+$ and $2^+ \rightarrow 1^+, 2^+, 3^+$. The 3971 keV level spin is therefore 1^+ or 2^+ . This confirms the previous assignment, based on the $\log ft$ value and angular momentum transfer in the (^3He , d) reaction [12].

Finally, a $2^+ \rightarrow 3^+$ spin assignment for the 1301 keV transition would imply a level de-orientation coefficient of $U_2(3971) \leq -0.78(26)$. This corresponds to an almost pure $j_\beta = 2$ decay ($U_2 = -0.59$) to the 3971 keV level. However, the corrected beta transition probability [13] ($\log f^{1U}t = 7.31$) is far too small to correspond to a unique first forbidden decay and so this spin sequence must be discarded. The spin of the 2670 keV level is not therefore 3^+ .

3459 keV level : ($1^+, 2^+$)

The β^+/EC transition to this level is too fast to be unique first forbidden and so, since anisotropies were also observed, the spin assignment is limited to $1^\pm$ and $2^\pm$. Taking these spins in turn the following results are obtained

	experiment	theory
if 1^+ $U_2A_2(804, 1^- \rightarrow 1^+ \rightarrow 1^-) = +0.10(6)$	$-0.35 \rightarrow +0.18$	
if 1^- $U_2A_2(1672, 1^- \rightarrow 1^- \rightarrow 2^+) = -0.28(17)$	$-0.04 \rightarrow +0.07$	
if 1^- $U_2A_2(2901, 1^- \rightarrow 1^- \rightarrow 2^+) = -0.11(5)$	$-0.04 \rightarrow +0.07$	
if 2^+ $U_2A_2(804, 1^- \rightarrow 2^+ \rightarrow 1^-) = +0.10(6)$	$-0.25 \rightarrow +0.25$	
if 2^- $U_2A_2(1672, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.28(17)$	-0.25	
if 2^- $U_2A_2(2901, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.11(5)$	-0.25	

Therefore the observed U_2A_2 coefficients rule out the possibility of spins 1^- and 2^- , confirming the previous ($1^+, 2^+$) assignment [11,12].

3160 keV level : ($1, 2^+$)

Using an identical argument to that presented for the 3459 keV level the spin assignment is limited to $1^\pm$ and $2^\pm$. Assuming these values in turn

	experiment	theory
if 1^+ $A(490, 1^+ \rightarrow (2^-) : 1471, 1^+ \rightarrow 3^+) = +4.0(2.7)$		-0.7
if 1^- $A(731, 1^- \rightarrow 3^- : 1033, 1^- \rightarrow 2^+) = -0.47(36)$		-1.4
$A(731, 1^- \rightarrow 3^- : 1372, 1^- \rightarrow 2^+) = -0.46(36)$		-1.4
$A(731, 1^- \rightarrow 3^- : 1944, 1^- \rightarrow 2^+) = -0.52(44)$		-1.4
$A(731, 1^- \rightarrow 3^- : 2601, 1^- \rightarrow 2^+) = -0.78(67)$		-1.4
if 2^+ $A(490, 2^+ \rightarrow (2^-) : 731, 2^+ \rightarrow 3^-) = +2.6(2.1)$		-3.5
if 2^- $U_2A_2(1033, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.16(8)$		-0.25
$U_2A_2(1372, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.17(8)$		-0.25
$U_2A_2(1471, 1^- \rightarrow 2^- \rightarrow 3^+) = +0.05(2)$		$+0.07$
$U_2A_2(1944, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.15(8)$		-0.25
$U_2A_2(2601, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.09(5)$		-0.25

Apparently none of these spin assignments are favoured by the data. However it has already been argued that the 2670 keV level, to which the 490 keV transition decays, is not (2^-) . Therefore the above comparisons serve only to rule out the possibility of spins 1^- and 2^- . Taking the 2670 keV level to have spin 1^+ or 2^+ would allow both spin 1^+ and 2^+ for the present level. However, in view of the presence of gamma transitions to levels of spin 0^+ , 2^+ and $3^\pm$, a spin 1^+ assignment is unlikely. Hence the spin of the 3160 keV level is taken to be 2^+ .

With this assumption, the U_2A_2 coefficient of the 731 keV $2^+ \rightarrow 3^-$ transition yields $U_2(3160) = +0.59(33)$, whence a lepton field mixing ratio of $\Delta^2(j_\beta = 2/j_\beta = 1) \leq 0.38$ is derived. Further, from the experimental ratio $A(\gamma : 731)$ the following mixing ratios can be calculated

$$\begin{array}{ll}
 A_2(2601) = -0.15(12) & 2^+ \rightarrow 2^+ \quad \delta(2601) = -0.21_{-10}^{+10} \quad \text{or} \quad +4.6_{-1.4}^{+3.5} \\
 A_2(1944) = -0.23(20) & 2^+ \rightarrow 2^+ \quad \delta(1944) = -0.12_{-16}^{+10} \quad \text{or} \quad +3.3_{-1.3}^{+3.2} \\
 A_2(1471) = +0.08(6) & 2^+ \rightarrow 3^+ \quad \delta(1471) = -0.02_{-5}^{+4} \quad \text{or} \quad -5.2_{-1.7}^{+1.1} \\
 A_2(1372) = -0.26(21) & 2^+ \rightarrow 2^+ \quad \delta(1372) = -0.10_{-17}^{+18} \quad \text{or} \quad +3.0_{-1.2}^{+3.5}
 \end{array}$$

3070 keV level : (1, 2)

Again the spin assignment can be limited to $1^\pm$, $2^\pm$. Hence

	experiment	theory
if 1^+ $U_2A_2(1381, 1^- \rightarrow 1^+ \rightarrow 3^+) = +0.05(1)$	$-0.10 \rightarrow +0.05$	
if 1^- $U_2A_2(1854, 1^- \rightarrow 1^- \rightarrow 2^+) = -0.11(1)$	$-0.04 \rightarrow +0.07$	
if 1^- $U_2A_2(2511, 1^- \rightarrow 1^- \rightarrow 2^+) = -0.13(2)$	$-0.04 \rightarrow +0.07$	
if 2^- $U_2A_2(1854, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.11(1)$	-0.25	
if 2^- $U_2A_2(2511, 1^- \rightarrow 2^- \rightarrow 2^+) = -0.13(2)$	-0.25	

Consistency for a 2^+ assignment may be also by achieved by a suitable choice of mixing ratio. Thus the experimental anisotropies indicate that the 3070 keV level is of positive parity with possible spins 1^+ or 2^+ .

2951 keV level : (1^-)

From the U_2A_2 coefficient of the 2951 keV $(1^-) \rightarrow 0^+$ transition it is possible to calculate a de-orientation parameter $U_2(2951) = -0.23(1)$. The G-T to F mixing in the allowed β^+ /EC decay is therefore $\Delta^2(j_\beta = 1/j_\beta = 0) = 4.6(2)$.

2670 keV level : (2^-)

The observed beta reduced transition probability to this level is $\log ft = 7.40$. Assuming a unique first forbidden decay then this can be corrected to $\log f^{1U}t = 8.38$, which is just slow enough to be plausible. Therefore the β^+ /EC feed to this level limits the possible spins to $0^\pm, 1^\pm, 2^\pm$ or 3^+ . The presence of gamma transitions to levels of spin 2^+ and 3^+ and the observation of non-zero anisotropies further limit the assignments to 1^+ , $2^\pm$ or 3^+ . From previous arguments relating to the 3971 and 3160 keV levels the spins 2^- and 3^+ can also be excluded. In fact the possibility of spin 2^- can be also be ruled out directly from the anisotropies of the 2111, 1454 and 882 keV transitions from this level. All the feeding gamma transitions arise from levels assigned $(1^+, 2^+)$, some of which are verified in the present work. Therefore it is possible to calculate their contribution to the total de-orientation coefficient of the 2670 keV level. Thus for transitions $1^- \rightarrow 1^+ \rightarrow 2^-$; $-0.3 \leq U_2 \leq +0.60$, while for transitions $1^- \rightarrow 2^+ \rightarrow 2^-$; $-0.30 \leq U_2 \leq +0.30$. Since the beta feed would be pure G-T and using the measure de-orientation coefficient for the 3160 keV level, then the total level de-orientation coefficient is calculated to lie in the range $+0.18 \leq U_2(2670) \leq +0.58$. The expected

U_2A_2 coefficient for the 2111, 1454 and 882 keV E1 transitions to levels of spin 2^+ is then predicted to be $-0.24 \leq U_2A_2 \leq -0.08$. When compared to the respective experimental values of $-0.03(1)$, $-0.06(3)$ and $-0.06(7)$ it is clear that the former value at least is inconsistent with the assumption of spin 2^- . Therefore the spin of this level must be either 1^+ or 2^+ . Such an assignment disagrees with the conclusions of Kaur *et al.* [14] who proposed spin 2^- on the basis of $\gamma\gamma(\theta)$ measurements in the decay of ^{76}As . However the possibility of positive parity was not considered since the beta decay to this level was assumed to be allowed.

2656 keV level : 1^-

From the U_2A_2 coefficients of the 1439 and 2097 keV E1 transitions the level de-orientation coefficient is calculated to be $U_2(2656) = -0.01(36)$.

2429 keV level : 3^-

From the anisotropy of the 1213 keV E1 transition the coefficient $U_2(2429) = +0.58(87)$ is obtained.

1788 keV level : 2^+

The de-orientation coefficient for this level is $U_2(1788) = +0.02(6)$, as calculated from the 665 and 1788 keV $2^+ \rightarrow 0^+$ transitions.

1689 keV level : 3^+

The angular distribution coefficient for the 1130 keV transition has been measured by Subber *et al.* in the β^- decay of ^{76}As [15] to be $A_2 = -0.73(15)$. From this it can be deduced that $U_2(1689) = 0.23(5)$. Further, from the ratio $A(473 : 1130) = +0.64(14)$

$$A_2(473) = -0.47(14) \quad 3^+ \rightarrow 2^+ \quad \delta(473) = +0.47_{-10}^{+12} \quad \text{or} \quad +5.0_{-17}^{+38}$$

This mixing ratio agrees with, but is more accurate than, the corresponding result of Subber *et al.* [15] and is consistent with the earlier work of Matsuzaki *et al.* [16].

1331 keV level : 4^+

The de-orientation coefficient is calculated to be $U_2(1330) = +0.02(16)$ from the 772 keV E2 transition.

1216 keV level : 2^+

From the known angular distribution coefficient of the 1216 keV E2 transition the level de-orientation parameter $U_2(1216) = +0.07(2)$ is obtained. Also $A(657 : 1216) = +0.14(10)$, thus

$$A_2(657) = -0.09(6) \quad 2^+ \rightarrow 2^+ \quad \delta(657) = -0.26_{-5}^{+5} \quad \text{or} \quad +5.8_{-15}^{+23}$$

The latter of the two mixing ratios is consistent with the previously known values of $+4.2(2)$ [15] and $+5.2(2)$ [17].

559 keV level : 2^+

The total feeding to this level is 36% by direct β^+ /EC and 64% by preceeding gamma transitions. The contribution of the gamma feeding to the total level de-orientation coefficient may be estimated from the the present results. Taking the above de-orientation coefficients and mixing ratios as appropriate (and also $\delta(1229)$ [14,15], $\delta(1130)$ [11] and $\delta(657)$ [15,17] from other works) allows a partial intensity weighted de-orientation coefficient of $-0.05 \leq \sum_{\gamma} I_{\gamma} U_2(\gamma) \leq +0.07$ to be calculated. This corresponds to 43% of the total feeding. The contribution of the remaining 21% of the gamma feed, for which no experimental values exist, may be estimated by taking the maximum and minimum theoretical values for the spin sequence $1^- \rightarrow 1, 2 \rightarrow 2^+$, which are $+0.12$ and -0.12 respectively. Again these values are intensity weighted. In this way the total intensity weighted gamma de-orientation coefficient is estimated to be $-0.17 \leq \sum_{\gamma} I_{\gamma} U_2(\gamma) \leq +0.19$.

Together with the de-orientation coefficient corresponding to the lepton decay this may be related to the total measured coefficient thus

$$U_2(559) = I_{\beta} U_2(\beta) + \sum_{\gamma} I_{\gamma} U_2(\gamma). \quad (5.5)$$

The observed level de-orientation coefficient, as deduced from the 559 keV E2 transition, is $U_2(559) = -0.075(7)$. Thus $-0.76 \leq U_2(\beta) \leq +0.28$. With limiting theoretical values of $+0.59$ and -0.59 for $j_{\beta} = 1, 2$ respectively this result leads to a lepton mixing ratio of $\Delta^2(j_{\beta} = 2/j_{\beta} = 1) \geq 0.36$.

In practice the limits assumed for the gamma de-orientation coefficients will be rather too generous. The mid-point value of the partial intensity weighted de-orientation coefficient for the gamma feeds with known parameters is $+0.01$ and there is little reason to expect much difference for the remaining unknown feeds. Thus the actual total gamma de-orientation coefficient is likely to be fairly close to zero. In this more realistic approximation the lepton mixing ratio is calculated to be $\Delta^2 \sim 1.3$ implying a very significant, but not necessarily pre-eminent, contribution from the quadrupole field.

The corrected first unique forbidden transition probability [13] is calculated to be $\log f^{1U} t = 9.5$. This is entirely in order with the values of other such transitions in this region which are

typically ~ 10 and so the present result is certainly not without foundation. On the other hand, the present lepton mixing ratio of the first forbidden decay to the higher lying 3160 keV level suggests little, if any, contribution from the quadrupole field. An increasing presence of $j_\beta = 2$ components as the decay energy becomes greater might in fact explain the gradual increase in the observed $\log ft$ values towards the lower part of the level scheme.

5.4 The isotope ^{75}Se

The anisotropies of gamma transitions in the decay of ^{75}Br were observed at an average temperature of 12.7 mK. Using the measured moment of $0.76(18)\mu_N$ and the site distribution parameter $f = 67(9)\%$, an effective orientation parameter $\sum fB_2 = 0.32(9)$ is obtained. The experimental anisotropies and U_2A_2 coefficients are listed in table 5.3 .

The decay scheme, shown in figure 5.3 , is taken from Nuclear Data Sheets ^[18]. In addition, several spin and parity assignments have been given more recently by Tokunaga *et al.* ^[19].

Table 5.3. The axial/equatorial anisotropies and U_2A_2 coefficients observed in the decay of ^{75}Br at 12.7 mK.

Energy (keV)	E (%)	U_2A_2
141	-0.5(8.7)	-0.01(19)
236	-5.7(9.8)	-0.12(22)
287	0.9(0.4)	0.02(1)
377	41.1(4.3)	0.79(23)
428	-12.1(2.2)	-0.27(9)
432	7.1(3.0)	0.15(7)
573	-5.5(4.8)	-0.12(11)
609	-8.4(3.7)	-0.18(10)
702	-29.6(15.0)	-0.71(45)
952	14.5(6.4)	0.30(15)

1561 keV level : (5/2)

The $\log ft$ value for the β^+/EC decay to this level indicates an allowed or non-unique first forbidden transition from the $3/2^-$ ground state of ^{75}Br . The observation of orientation in the 702 keV transition rules out spins $1/2^\pm$ absolutely and the presence of gamma transitions to levels of spin $3/2^-$, $5/2^+$ and $7/2^+$ further limit the possible spins to $3/2^+$ or $5/2^\pm$. Further,

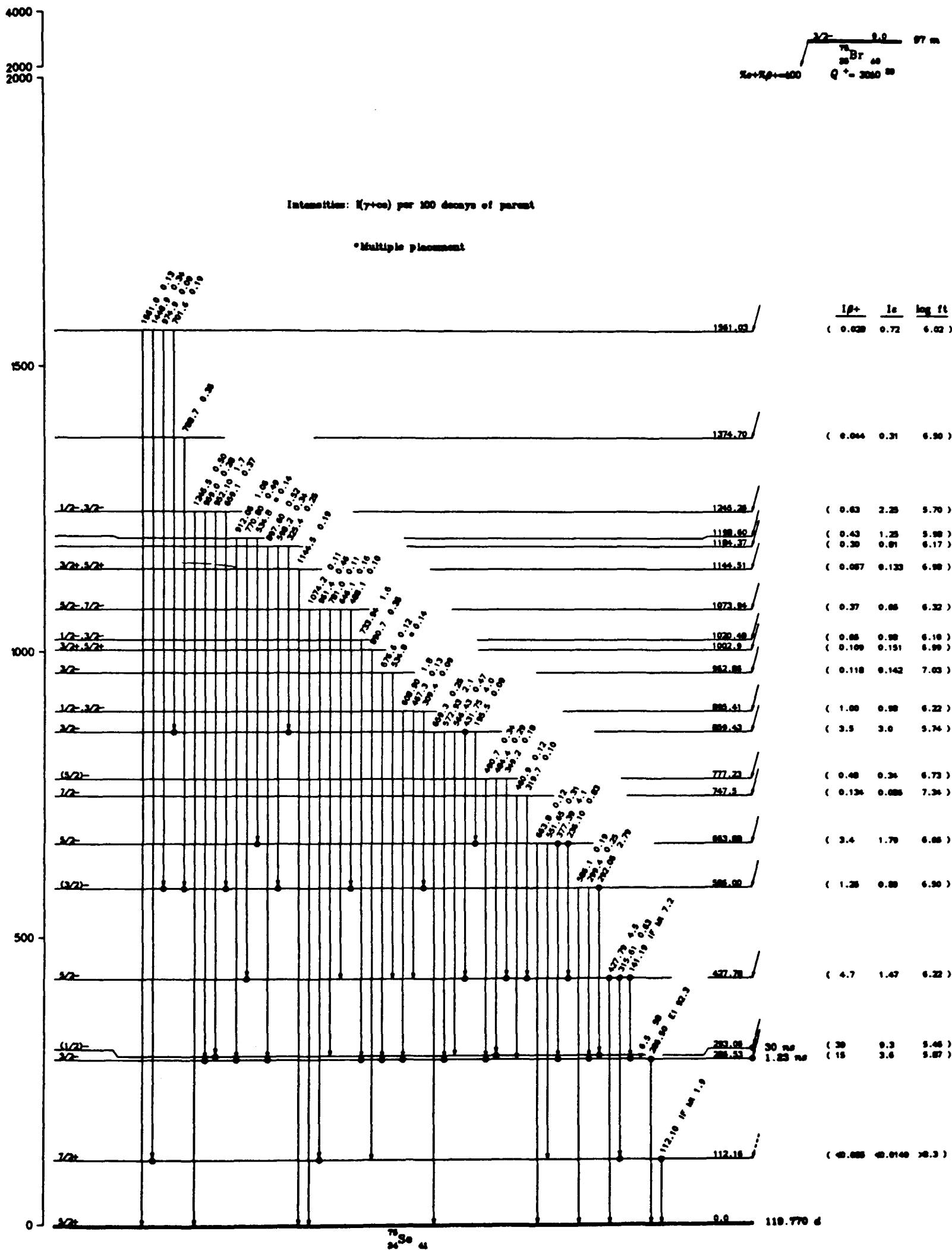


Figure 5.3. The decay scheme of ^{75}Br [18].

an assignment of $5/2^+$ can be excluded as this would imply that the U_2A_2 coefficient of the 702 keV transition is in the range $-0.04 \rightarrow +0.28$, contrary to the observed value of $-0.71(45)$. Since Tokunaga *et al.* have tentatively proposed a spin assignment of $(5/2)$, the 1561 keV level spin is taken to be $5/2^-$.

With this assignment the β^+/EC decay is pure G-T, so that

$$A_2(702) = -0.95(60) \quad 5/2^- \rightarrow 3/2^- \quad \delta(702) \geq +0.40$$

1245 keV level : $1/2^-$, $3/2^-$

The fact that anisotropies were observed for the 952 keV transition, which decays to the 293 keV $1/2^-$ level, can be used to limit the spin assignment for this level to $3/2^\pm$ or $5/2^-$. The latter possibility can be excluded since it would predict that $U_2A_2 = -0.40$, in contrast to the observed positive value. The present data is consistent with both $3/2^-$ and $3/2^+$ spin assignments. Previous reaction transfer data ^[18,20] have suggested a spin of either $1/2^-$ or $3/2^-$. Therefore the spin of this level is taken to be $3/2^-$.

The level de-orientation coefficient then lies in the range $+0.2 \leq U_2(1245) \leq +1.0$ and hence

$$A_2(952) \geq +0.15 \quad 3/2^- \rightarrow 1/2^- \quad -2.9 \leq \delta(952) \leq +0.18$$

895 keV level : $1/2^-$, $3/2^-$

Again previous transfer data ^[18,20] have suggested level spin assignments of $1/2^-$ or $3/2^-$. Since the 609 keV transition exhibits anisotropy the former possibility may be excluded. The 895 keV level spin is therefore $3/2^-$.

The level de-orientation coefficient thus lies in the same range as for the 1245 keV level, whence

$$A_2(609) \leq -0.08 \quad 3/2^- \rightarrow 3/2^- \quad \delta(609) \geq -0.20$$

859 keV level : $3/2^-$

Taking the previously known mixing ratio for the 432 keV transition of $\delta(432) = +0.34(24)$ ^[18,20,21] yields the theoretical angular distribution coefficient $A_2 = +0.49(23)$. Thus the level de-orientation coefficient is calculated to be $U_2(859) = +0.31(20)$. Only 6% of the total feeding to this level is by preceeding gamma transitions which can therefore be neglected. Thus the lepton mixing ratio $\Delta^2(j_\beta = 1/j_\beta = 0) \geq 1.6$ is deduced.

Further, from the experimental ratio $A(573 : 432) = -0.81(79)$

$$A_2(573) = -0.40(43) \quad 3/2^- \rightarrow 3/2^- \quad \delta(573) \geq -0.24$$

664 keV level : $5/2^-$

This level is fed almost entirely (97%) by pure G-T beta decay with a theoretical de-orientation parameter $U_2 = +0.75$. The known mixing ratio of the 377 keV transition ^[20] has already been used to limit the error on the fitted moment of ⁷⁵Br (see subsection 4.4.1). In addition

$$A_2(236) = -0.16(29) \quad 5/2^- \rightarrow 5/2^- \quad \delta(236) = -0.24^{+26}_{-26} \quad \text{or} \quad [1.6 \rightarrow 17]$$

Electron conversion measurements by Tokunaga *et al.* ^[19] give $|\delta(236)| = 0.31^{+15}_{-31}$, in agreement with the former of the present values.

5.5 The isotope ⁷⁴Se

The averaged anisotropies of gamma rays resulting from the β^+/EC decay of ⁷⁴Br^m are listed in table 5.4 . Since the oriented spin is $I = 4$ ^[22] the directional distribution of these gamma rays in general contain terms of both second and fourth rank. Taking the fitted moment of $1.68(18)\mu_N$ and site distribution parameter $f = 67(9)\%$ leads to effective orientation parameters $\sum fB_2 = 0.92(9)$ and $\sum fB_4 = 0.26(5)$ at the average experimental temperature of 9.8 mK.

From the relative magnitudes of the two effective orientation parameters it is clear that fourth rank terms may contribute significantly to the anisotropies appearing in table 5.4. The two $U_\lambda A_\lambda$ coefficients must therefore be determined from the temperature dependence of the anisotropies. With the moment and site distribution parameter fixed to the above values, $U_2 A_2$ and $U_4 A_4$ are thus extracted separately as the only variable parameters. (It has been stated in subsection 4.4.2 that such a fitting procedure, including $U_2 A_2$ and $U_4 A_4$ as variable parameters, is only reliable for the 634 and 728 keV transitions. There, however, the moment was also a variable parameter). The resulting fourth rank terms, generally having the smaller contribution to the anisotropy, are of rather poor precision and necessarily the error in the corresponding second rank terms is increased. In this respect, where it can be shown theoretically that the

U_4A_4 coefficient of a given gamma transition either vanishes or is comparatively small, then it is better to consider the second rank terms exclusively by applying (5.2) to the averaged anisotropy point. There is then no additional error in U_2A_2 introduced by the uncertainty in the fitted value of U_4A_4 . Values thus derived are denoted $\overline{U_2A_2}$ in order to highlight the fact that in the general case they represent some mean value of the U_2A_2 and U_4A_4 coefficients. The $\overline{U_2A_2}$ coefficients together with the fitted values U_2A_2 and U_4A_4 are also given in table 5.4. Only the former quantities are tabulated for the statistically weaker transitions.

Table 5.4. The axial/equatorial anisotropies and U_2A_2 coefficients observed in the decay of $^{74}\text{Br}^m$ at 9.8 mK. † indicates a doublet.

Energy (keV)	E (%)	$\overline{U_2A_2}$	U_2A_2	U_4A_4
615	−38.7(1.8)	−0.33(3)	−0.39(6)	0.27(38)
634†	−23.5(0.4)	−0.19(2)	−0.19(4)	−0.10(14)
679	−26.5(12.3)	−0.22(11)	—	—
728	−39.4(0.6)	−0.34(3)	−0.38(5)	0.11(25)
745	13.8(7.9)	0.10(5)	—	—
839	−41.8(2.0)	−0.36(4)	−0.33(7)	−0.25(32)
868	−41.1(10.7)	−0.36(11)	—	—
980	−45.6(16.5)	−0.40(17)	—	—
985†	−33.1(3.3)	−0.28(4)	−0.22(6)	−0.53(31)
1201†	−41.3(2.4)	−0.36(4)	−0.42(9)	0.45(53)
1204	−33.4(8.4)	−0.28(8)	−0.30(17)	−0.28(99)
1250	−58.8(1.3)	−0.55(5)	−0.68(10)	0.55(46)
1269	−30.3(1.8)	−0.25(2)	−0.26(6)	−0.08(33)
1367	−35.6(5.2)	−0.30(5)	−0.29(11)	−0.38(83)

Although the isomeric spin has been firmly assigned, the parity has not. Negative parity was tentatively assigned on the basis of $\log f^{1U}t$ values to the 2231 keV 6^+ level and to several (2^+) states in ^{74}Se [23]. However by their very nature such first unique forbidden beta transitions have very low intensities. In view of the highly complex decay scheme, illustrated in figure 5.4 [24], the small gamma intensity imbalances from which they are deduced could well be accounted for by weak unobserved gamma transitions. As a result the negative parity assignment is by no means definite.

In fact a comparison of the band structures in $^{74,76}\text{Br}$ [25] has lead to the proposal of positive parity. Strong corroborative evidence for this assignment is provided by the present magnetic moment of $^{74}\text{Br}^m$ which only finds an explanation in the particle-rotor model for a positive parity configuration (see subsection 7.5.2).

