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Multipole mixing ratios of gamma rays emitted in the decay of polarized ^{160}Tb

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Abstract. It has been demonstrated that successful nuclear polarization of terbium isotopes can be achieved by making use of the high internal field experienced when the activity is dissolved in a gadolinium lattice cooled to 10 mK. The isotope ^{160}Tb was chosen as an example, since some features of the decay have already been established by $\gamma\gamma$ directional correlation methods. The directional distribution of the emitted radiation was used to determine the E2:M1 and M2:E1 multipole mixing ratios of thirteen γ ray transitions in ^{160}Dy. There is a general agreement with earlier results. Furthermore, the following values of previously undetermined M2:E1 multipole mixing ratios, $\delta(E_\gamma)$, were found:

$\delta(310) = -0.020(14)$, $\delta(337) = 0.039(32)$, $\delta(1003) = -0.004(17)$
$\delta(1103) = -0.156(25)$, $\delta(1115) = 0.000(12)$, $\delta(1200) = -0.017(8)$
$\delta(1312) = -0.017(8)$.

1. Introduction

The directional distribution of radiation emitted by nuclei polarized at a low temperature has proved a valuable technique for investigating the nuclear properties of a radioactive decay. By comparing different decays from the same level the mixing ratio of a γ ray transition and the spins of the levels which it links can often be obtained without knowing the degree of nuclear polarization (Fox *et al* 1972). The validity of this technique is largely unaffected by problems associated with source preparation, thermal contact, or misalignment of the hyperfine magnetic field. However, the polarization of rare-earth nuclei presents particular problems. The most important of these is the insolubility of the rare earths in iron, which is the host lattice most commonly used to produce large axial internal fields at solute nuclei. Previous attempts to overcome this have been made by implanting the rare-earth ions into iron using an isotope separator. In a recent experiment (Krane *et al* 1972), ytterbium was dissolved in gold to produce a hyperfine field of 1·8 MG at the nucleus. Unfortunately, the axial hyperfine magnetic field applicable to this case (Spanjaard *et al* 1973) does not occur for most other members of the series which are soluble in gold. Only for erbium is it likely that a simple axial magnetic field (2·9 MG), will be sufficient to describe the magnetic hyperfine interaction.

The present experiment was carried out to demonstrate that polarization could be achieved by using the high internal field that a rare-earth nucleus experiences when the atom is dissolved in a gadolinium host lattice. The isotope ^{160}Tb was chosen as its decay has already been extensively studied by $\gamma\gamma$ directional correlation methods and some multipole mixing ratios are well established. The polarization experiment also provides an opportunity to measure some transitions from the negative parity states in ^{160}Dy which are not readily amenable to $\gamma\gamma$ correlation measurement.

The measurement with polarized nuclei is complementary to the $\gamma\gamma$ directional correlation experiments since now the transition of interest is the second (analysing) rather than the first (orienting) member of the cascade. This is particularly useful since the γ ray orienting and analysing coefficients $B_2(\gamma)$ and $A_2(\gamma)$ have a different δ dependence namely:

$$B_2(\gamma) = [F_2(L\ L\ I'I) - 2\delta F_2(L\ L+1\ I'I) + \delta^2 F_2(L+1\ L+1\ I'I)](1+\delta^2)^{-1} \tag{1}$$

and

$$A_2(\gamma) = [F_2(L\ L\ I'I) + 2\delta F_2(L\ L+1\ I'I) + \delta^2 F_2(L+1\ L+1\ I'I)](1+\delta^2)^{-1} \tag{2}$$

where in each case I' is the state which is oriented. The definition of the multipole mixing ratio of the $L+1:L$ amplitudes used in this paper and denoted by δ is that introduced by Krane and Steffen (1970). The F coefficients have been frequently tabulated (eg Biedenharn and Rose 1953).

The measured magnetic and electric hyperfine interactions for rare earths in gadolinium are very similar to those obtained for the free rare-earth ions (Kobayashi *et al* 1967). This indicates that the exchange field in the gadolinium lattice is sufficiently strong to produce an electronic ground state with $\langle J_z \rangle = J$ for the rare-earth impurity. A review of the hyperfine interactions for rare earths in gadolinium has recently been published (Bleaney 1972). A simple hamiltonian

$$\mathscr{H} = g_I \beta_I \boldsymbol{I} \cdot \boldsymbol{B}_z + P[I_z^2 - \tfrac{1}{3}I(I+1)] \tag{3}$$

which is diagonal along the axis of the external magnetic field, should be sufficient to describe the hyperfine interaction. The first term in this equation represents the interaction of the nuclear dipole moment $g_I \beta_I \boldsymbol{I}$ with the effective magnetic field $\boldsymbol{B}_z$ at the nucleus. The measurements of Kobayashi *et al* indicate that this field should be 3·03(1) MG. The second term represents an axial quadrupole interaction where P contains the nuclear quadrupole moment.

The directional distribution of radiation from an ensemble of nuclei, polarized under the influence of the interaction of equation (3), is then given by (Blin-Stoyle and Grace 1957)

$$W(\theta) = \sum_{\lambda\ \mathrm{even}} B_\lambda(I) U_\lambda A_\lambda(\gamma) Q_\lambda P_\lambda(\cos\theta).$$

B_λ is a statistical tensor whose value is determined by the nuclear level population at a given temperature; U_λ describes the effect of the unobserved transitions, both β and γ decay, which precede the γ ray transition; $A_\lambda(\gamma)$ is the directional distribution coefficient which depends on the level spins and multipolarities involved in the γ ray transition (see equation (2)). The final coefficient, Q_λ, corrects for the finite solid angle subtended at the source by the γ ray detector.

2. Experimental procedure

Radioactive ^{160}Tb in the form of Tb Cl_3 in 0·1 N hydrochloric acid was obtained from the Radiochemical Centre, Amersham. Approximately 5 μCi of this was dried onto the surface of a piece of gadolinium of mass 0·1 g, which was then melted under argon in a drawn silica tube. The concentration of terbium in gadolinium was less than 0·1 %. The resulting needle of gadolinium was cleaned with emery paper to remove surface activity and then tinned in an indium bath which was agitated ultrasonically to maintain a clean gadolinium surface. This sample was soldered without flux to the copper thermal link of a chrome alum–glycerol salt pill.

A sample of ^{54}Mn in iron was soldered to the thermal link beside the ^{160}Tb sample. This was prepared by melting iron, coated with carrier free ^{54}Mn activity, in an atmosphere of hydrogen. This sample was cleaned, rolled, annealed and finally tinned with indium. The 835 keV γ ray of ^{54}Mn has been well studied by nuclear orientation methods so that its anisotropy provides a convenient and accurate measure of the sample temperature.

Both radioactive samples were cooled to 10 mK by contact made to the salt pill via 10 000 strands of 48 gauge copper wire. The pill was suspended beneath a ^{3}He:^{4}He dilution refrigerator operating at 30 mK, to which it was thermally linked by a lead heat switch. After demagnetizing the salt pill, a small polarizing field of 6 kG was used to align the magnetic domains of the gadolinium and iron. The anisotropy of the γ radiation from ^{160}Tb was observed to increase with applied field until about 4 kG after which it remained constant to within the experimental error. This suggests that the magnetic domains of the gadolinium should be quite well polarized in a field of 6 kG.

Figure 1 depicts the relevant parts of the decay scheme of ^{160}Tb. For convenience the transitions have been grouped into those involving the ground state band, the gamma vibrational band and two negative-parity bands.

The γ ray count rate was taken at angles of 0 and $\pi/2$ with respect to the applied field direction, using a 30 cm^3 Ge(Li) detector which had a resolution of 2·5 keV at 1·3 MeV. The voltage pulses were amplified and fed into a 4096 channel analyser. The full energy spectrum was read onto magnetic tape every 2000 s. The observed spectrum is illustrated in figure 2. Computer analysis involved summing the counts in the channels across the peak and subtracting the background radiation from under it. The latter was obtained by averaging the count rate at a position a few keV on the high energy side of the peak. A correction for source decay was also made. The anisotropies, $1-W(0)$ and $W(\pi/2)-1$, were obtained by comparing the cold counts with the warm (1 K) counts for which the radiation is isotropic.

These anisotropies are used to determine the value of the A_λ coefficients by applying equation (4). In the present experiment with a source–detector separation of 9 cm, the solid angle correction factors, to better than 0·5 % accuracy, have the values $Q_2 = 0{\cdot}98$ and $Q_4 = 0{\cdot}93$. At this accuracy they are effectively independent of energy and have therefore been used for all γ ray transitions.

3. Results

The anisotropies at 9·8(2) mK for eighteen γ ray transitions in ^{160}Dy observed in the decay of ^{160}Tb are given in table 1. These have been analysed using the result of Krane and Steffen (1971) for the 299 keV γ ray as a basis for comparing all other transitions. It is justifiable to assume that the allowed β transitions to the negative-parity levels are pure Gamow–Teller when calculating the contribution of these β decays to the U_λ coefficients. This statement is not altered by the small anisotropy measured by Cipolla *et al* (1966) for the β–γ correlation in ^{160}Tb in which the 1265 keV level is intermediate.

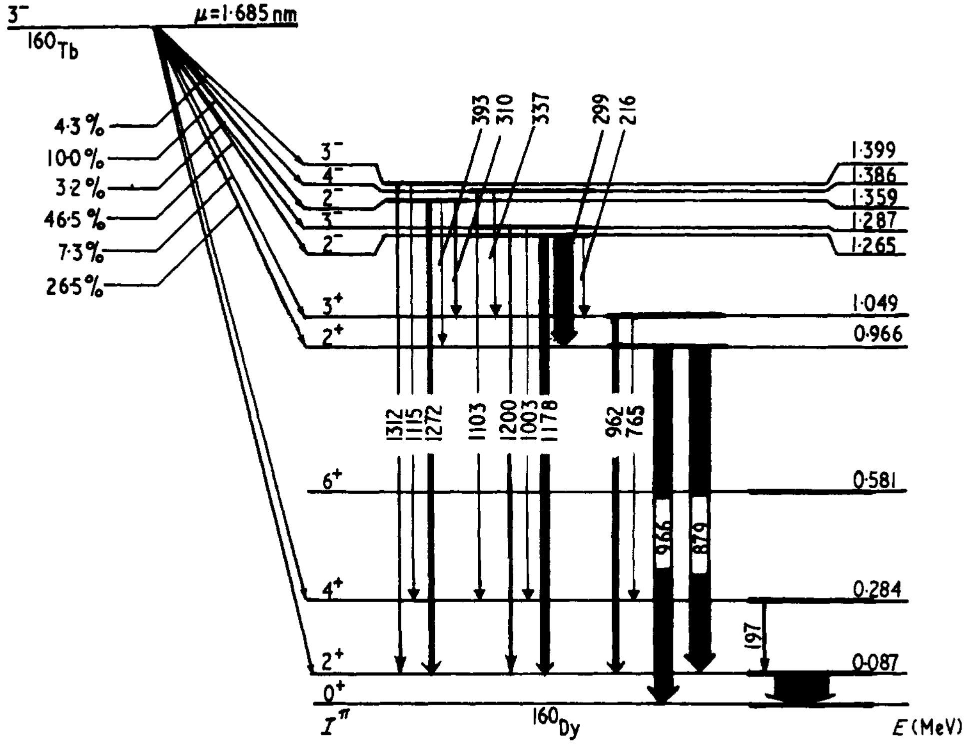

Figure 1. The decay scheme of ^{160}Tb. Only transitions measured in the present experiment have been included. The thickness of the line indicates the approximate γ ray transition intensity. The intensity of the dominant β transitions derived from the γ ray intensities of Ludington *et al* (1968) are indicated.

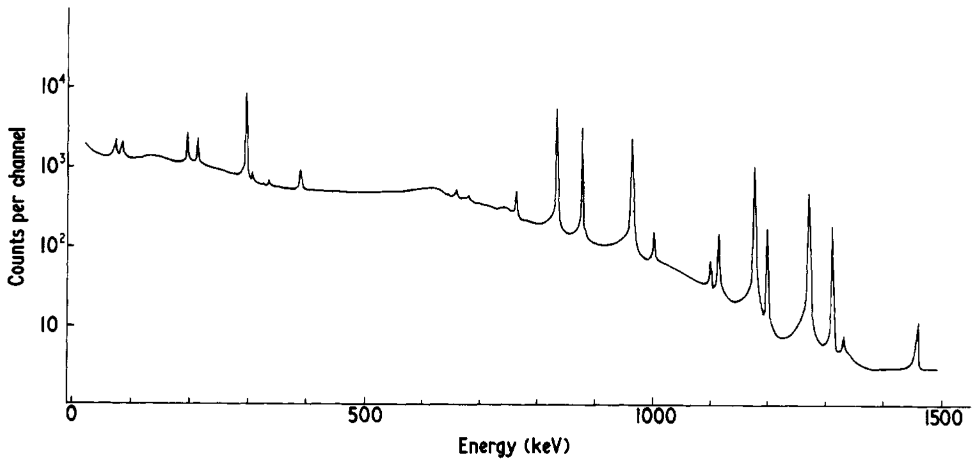

Figure 2. The γ ray spectrum observed for ^{160}Tb with a 30 cm^3 Ge(Li) detector.

Table 1. The anisotropies at 0 and $\pi/2$ to the applied field direction are tabulated for all measured γ ray transitions.

Energy of excited level (keV)	Energy of transition (keV)	Spin sequence I_i–I_f	$[1-W(0)]\%$	$[W(\pi/2)-1]\%$
1399	1115	3^-–4^+	−6·0(7)	−1·7(20)
	1312	3^-–2^+	−15·7(4)	−5·1(11)
1386	337	4^-–3^+	−12·0(30)	
	1103	4^-–4^+	16·1(10)	8·9(34)
1358	310	2^-–3^+	−4·3(8)	−0·8(21)
	392	2^-–2^+	16·7(6)	6·5(19)
	1272	2^-–2^+	19·0(3)	10·0(7)
1287	1003	3^-–4^+	−5·7(10)	−3·0(27)
	1200	3^-–2^+	−15·7(5)	−7·8(12)
1265	215	2^-–3^+	−5·3(3)	−1·3(8)
	299	2^-–2^+	19·4(3)	9·5(3)
	1178	2^-–2^+	17·4(3)	8·4(5)
1049	765	3^+–4^+	−3·5(5)	−2·7(14)
	962	3^+–2^+	−2·5(20)	
966	880	2^+–2^+	−7·5(3)	−2·6(3)
	966	2^+–0^+	18·0(10)	
284	197	4^+–2^+	14·0(3)	6·6(7)
87	87	2^+–0^+	4·7(8)	6·2(20)

This is because the U_λ coefficients are not affected by cross terms between allowed and second forbidden β decay matrix elements. The measured anisotropy for the 299 keV transition together with the value $\delta(299) = 0{\cdot}005(10)$ measured by Krane and Steffen lead to $B_2(I) = 0{\cdot}56(2)$. It will be seen that a similar value for $B_2(I)$ may be derived using only the present measurements.

3.1. *Transitions from negative-parity levels*

All transitions from the negative-parity levels are expected to be predominantly E1 so that the fourth order term in equation (4) may be neglected. Even for the 1103 keV transition with $\delta = -0{\cdot}16$, which is the largest value measured, U_4A_4 is only 0·01 so that the correction to A_2 due to the fourth order term is much smaller than the standard deviation on the measurement. Consequently, the anisotropy of the 215 keV and 1178 keV transitions may be compared directly with that of the 299 keV transition from the same level. Comparison of other transitions with the 299 keV γ ray relies on a knowledge of the U_2 coefficients. These arise directly from the assumption of pure Gamow–Teller allowed β decays. The results of the analysis of all observed transitions from the negative parity levels are contained in table 2.

3.2. *Transitions from positive-parity levels*

The transitions from the positive-parity levels are expected to be predominantly E2, so that the fourth order term in equation (4) is not necessarily negligible. It can however

Table 2. The calculated values of A_2 and δ are shown for transitions from the negative-parity levels. Values of δ measured by Krane and Steffen (1971) are also included. For the evaluation, the β decay was taken as pure Gamow–Teller.

Initial level (keV)	U_2	Transition energy (keV)	A_2	δ† Present results	δ† Krane and Steffen
1399	0·75	1115	0·145(17)	0·000(12)	
		1312	0·379(17)	−0·017(8)	
1386	0·9047	337	0·24(6)	0·039(32)	
		1103	−0·32(2)	−0·156(25)	
1358	0·8281	310	0·094(17)	−0·020(14)	
		392	−0·365(18)	−0·043(15)	0·02(4)
		1272	−0·415(15)	−0·003(12)	−0·03(3)
1287	0·75	1003	0·138(25)	−0·004(17)	
		1200	0·379(17)	−0·017(8)	
1265	0·8281	215	0·116(8)	−0·003(6)	−0·18(10)
		299			0·005(10)
		1178	−0·380(14)	−0·031(12)	0·02(2)

† Previously measured values of δ have been tabulated by Krane and Steffen (1971).

be calculated if B_4 is known. The analysis (Fox 1974) of the temperature dependence of the anisotropy of the 299 keV transition using equations (3) and (4) indicates a larger quadrupole interaction than suggested by the results of Kobayashi *et al* (1967). This leads to a value $B_4 = -0{\cdot}56(15)$ at the lowest temperature reached in the experiment. There may be other causes for our results which would have the effect of considerably reducing the magnitude of B_4. Work is at present in progress to clarify this situation. The following analysis will assume the value for B_4 given above, the effect of reducing the fourth order term will be considered at the end of this section.

The asymmetry of the γ decay from the positive-parity levels may be used either to determine γ ray mixing ratios when certain assumptions are made about the preceding β decay or alternatively it is possible to take previously determined mixing ratios and evaluate the contributions to the angular momentum carried by the β decay. For the present analysis it will be assumed that β decay transitions to these positive-parity levels carry one unit of angular momentum. With this assumption and the transition intensities and branching ratios quoted by Ludington *et al* (1968), it is possible to derive values of U_2 and U_4 for each positive-parity level. The value of A_4 is not very sensitive to small admixtures of M1 radiation with E2, and when the values of the initial and final spin states are known A_4 may be evaluated; the small M1 admixtures which have been measured in the present work alter the calculated values of A_4 by less than 2%. Fortunately it is found that in most cases the calculated values for U_4A_4 are small, which diminishes the effect of the fourth order term on the derived A_2 values.

The results of the analysis of the observed transitions from positive-parity levels are contained in table 3. In two cases the transition must be pure E2, so that an independent value of $B_2(I)$ may be derived. This agrees with the value, $B_2(I) = 0{\cdot}56(2)$, which was used in the analysis. In view of the assumption made in deriving the U_λ coefficients, the mixing ratio results involving positive-parity states should only be regarded as confirming previous work.

Table 3. The parameters used in the analysis of transitions from the positive-parity levels are tabulated together with the derived values of $B_2(I)$ and δ for pure and mixed transitions respectively. The values of U_λ are calculated assuming the β decays to positive-parity levels carry one unit of angular momentum only.

Initial level energy (keV)	Transition energy (keV)	U_2	U_4	$B_4U_4A_4Q_4$†	$B_2U_2A_2Q_2$‡	$B_2(I)$	A_2	Present results	Krane and Steffen (1971)
1049	765	0·727(20)	0·178(5)	−0·014(4)	0·049(6)		0·122(16)	$-7{\cdot}7^{+0{\cdot}6}_{-0{\cdot}7}$	-7^{+5}_{-20}
966	966	0·61(3)	0·06(2)	0·03(1)	−0·210(14)	0·59(5)			
	880	0·61(3)	0·06(2)	0·010(4)	0·065(5)		0·193(19)	-18^{+4}_{-8}	$-11{\cdot}0^{+1{\cdot}5}_{-1{\cdot}9}$
284	197	0·612(54)	0·15(4)	0·024(9)	−0·164(10)	0·61(6)			

† These values are calculated using $B_4 = -0{\cdot}56(15)$ and taking pure E2 radiation in the evaluation of A_4. The text should be consulted for discussion of these points.
‡ These values of the second order term are derived from the measured anisotropy, $1 - W(0)$, and the calculated value of $B_4U_4A_4Q_4$.

It is useful to discuss some particular points associated with table 3. These are considered in order of decreasing level energy as in the table.

3.2.1. The 765 keV transition. From the measured anisotropies at 0 and $\pi/2$ one can derive $B_4U_4A_4Q_4 = -0{\cdot}011(16)$. This is in good agreement with the calculated value given in table 3. The measured δ value agrees with the value of Krane and Steffen which gives confidence that the β decay carries one unit of angular momentum. It is difficult however to draw detailed conclusions, since a change of either zero or two units of angular momentum is possible.

3.2.2. The 962 keV transition. This transition has not been analysed in view of the inaccuracies arising due to its overlap with the 966 keV transition, and to the large fourth order term which occurs in this case.

3.2.3. The 966 keV transition. A plot was made of the anisotropy, corrected for background, of each channel across the 962 and 966 keV peaks. The resulting curve is shown in figure 3. The anisotropies for each γ ray were obtained by averaging the values for

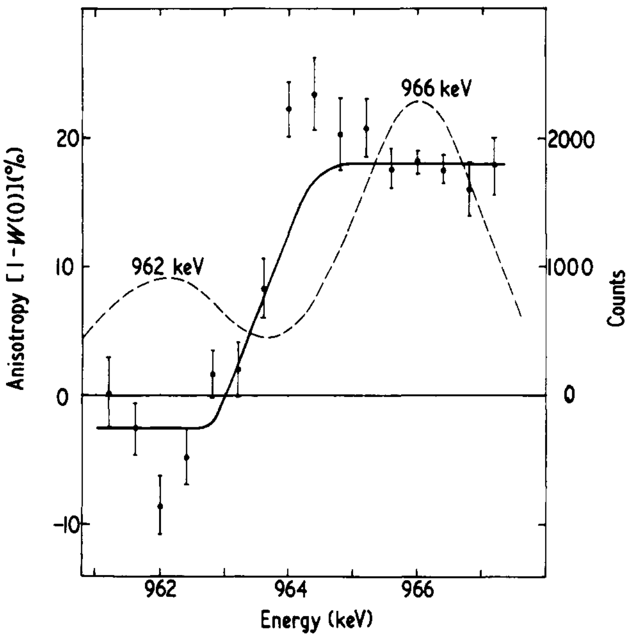

Figure 3. The anisotropy for each channel across the 962 and 966 keV peaks is plotted against the energy in keV. The full line indicates the basis of the anisotropy calculation; the broken line indicates the γ ray spectrum in this region.

individual channels over the parts of the peak regions which were considered to be undisturbed by the neighbouring line. Since the 966 keV transition is pure E2, it is possible to analyse the result to give an independent value of $B_2(I) = 0{\cdot}59(5)$ as listed in table 3. Keeping the same assumption for the β decay, this result for $B_2(I)$ can be used to re-analyse the 299 keV transition, and yields a value for the mixing ratio,

$\delta(299) = -0{\cdot}011(29)$. The other measured mixing ratios would also be modified, although generally by less than one standard deviation. Alternatively, with the value $B_2(I) = 0{\cdot}56(2)$, the measurement on the 966 keV transition can be used to predict that, for the 3^- to 2^+ β transition to the 966 keV level, 18 % or less the radiation carries two units of angular momentum.

3.2.4. The 880 keV transition. The measured anisotropies at 0 and $\pi/2$ independently predict that $B_4U_4A_4Q_4 = 0{\cdot}012(4)$ in excellent agreement with the calculated value given in table 3.

3.2.5. The 197 keV transition. This pure E2 transition again enables the value of B_2 to be evaluated (cf table 3).

Our results give a lower value for the anisotropy than predicted by the measurements of Kobayashi *et al* (1967). If our results are explained by poor thermal contact or incomplete solution of terbium in gadolinium the value of $B_4(I)$ will be smaller than that assumed above.

The measurements may be re-analysed with a reduced fourth order term to give some indication of its effect on the final results. Taking a value of $B_4(I) = 0{\cdot}0 \pm 0{\cdot}15$, the E2:M1 mixing ratios become $\delta(765) = -6{\cdot}5 \pm 0{\cdot}5$ and $\delta(880) = 13^{+2}_{-4}$, which compare satisfactorily with those in table 3. Furthermore, $B_2(I)$ may be derived from the anisotropies of 966 and 197 keV transitions, the results are 0·50(5) and 0·52(5) respectively. These are still in satisfactory agreement with $B_2 = 0{\cdot}56(2)$ which was derived previously. Analysis of the β decay to the 966 keV level with $B_2 = 0{\cdot}51(5)$, suggests that it does not contain any component carrying two units of angular momentum. Thus the previous results can be left unaltered.

4. Discussion

Our analysis of the pure E2 transitions show that the experimental results are self-consistent and suggests that the β transitions to the positive-parity levels at 1049 and 966 keV carry one unit of angular momentum.

All γ ray transitions from negative-parity levels to the ground state band exhibit extremely small admixtures of M2 radiation. This supports the analysis of Günther *et al* (1968) who were able to explain the observed negative-parity levels by assuming two bands each consisting of mixtures of $K = 0$, $K = 1$ and $K = 2$ states. Even the 1265 keV level, which Günther *et al* describe as 85 % $K = 2$, has a 1178 keV, $2^- \rightarrow 2^+$ transition to the ground state band containing less than 0·2 % M2 radiation. The 1103 keV transition from the 1384 keV level does however show a moderately large admixture of M2 (2 %), indicating hindrance in the E1 transition rate.

Our results contradict the large value of δ observed for the 215 keV transition in previous experiments (cf table 2). Furthermore, the signs of δ are found to be negative for both the 1178 and 1272 keV transitions contrary to the conclusion of Krane and Steffen (1971), whose results do not disagree significantly with the present more accurate data. The analysis of the 1178 keV transition involves no assumption since a direct comparison with the 299 keV transition is possible. A positive mixing ratio for this γ ray would require that its anisotropy equalled that of the 299 keV transition. This is repudiated by all individual measurements.

5. Conclusion

This experiment demonstrates the feasibility and potential of producing large hyperfine fields at rare-earth nuclei when these are dissolved in ferromagnetic gadolinium. Typically, fields are expected to have magnitudes in the region of several megagauss. These are suitable for application to nuclear orientation or perturbed directional correlation measurements.

The present measurements confirm the existing spin and parity assignments for the levels of ^{160}Dy populated in the decay of ^{160}Tb. The M2:E1 mixing ratios for transitions between negative-parity levels and the ground state band are found to be extremely small. This provides further evidence that these levels must be considered as mixtures of different *K* states.

Acknowledgments

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