

## Nuclear Orientation of <sup>152</sup>Eu in Gold\*

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### Abstract

Nuclear orientation of <sup>152</sup>Eu in gold in the temperature range 9 mK–1 K has been used to determine the mixing ratios of a number of gamma transitions and the multipolarities of several beta transitions of the <sup>152</sup>Eu decay scheme. The results agree closely with those from  $\gamma$ - $\gamma$  and  $\gamma$ - $\beta$  angular correlation measurements where these have been done. Our results on the hyperfine interaction of <sup>152</sup>Eu in gold contradict those previously published.

### 1. Introduction

The nucleus <sup>152</sup>Eu, which decays to both <sup>152</sup>Gd and <sup>152</sup>Sm (see Fig. 1), has been the subject of a number of investigations (see e.g. Barrette *et al.* 1970, 1971; Riedinger *et al.* 1970; Kalfas *et al.* 1973). Nuclear orientation (NO) of <sup>152</sup>Eu allows an independent determination of the angular distributions of radiations that are emitted in the decay cascade. Barclay and Perczuk (1975) have published an earlier NO study, but they obtained poor agreement with the literature. A discrepancy of this kind can occur in an NO experiment when the lifetime of an intermediate state is long enough to allow spin-lattice relaxation to cause reorientation of the nucleus. Due to the short lifetimes of the intermediate states in the decay to both <sup>152</sup>Sm and <sup>152</sup>Gd (except for the 122 keV level of <sup>152</sup>Sm), no reorientation is expected. In an attempt to resolve this discrepancy, a second NO investigation with larger applied fields (2.2 T as compared with 0.75 T) is presented here. For this second experiment a good agreement between our results and those in the literature was obtained. In addition, the NO technique allowed the magnetic environment of the <sup>152</sup>Eu nucleus to be investigated.

### 2. Outline of Theory

The normalized angular distribution of the  $\gamma$ -ray intensity along the axis of an axially symmetric system is given by

$$W(\theta, T) = \sum_K B_K(T) U_K A_K G_K Q_K P_K(\theta=0). \quad (1)$$

Matthias *et al.* (1971) gave a detailed description of each parameter in (1):  $B_K(T)$  are statistical parameters that contain all of the information concerning the magnetic

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environment of the nucleus;  $U_K$  are 'deorientation coefficients' that describe the effect of transitions previous to the one being observed;  $A_K$  are angular distribution coefficients of the observed radiation. The coefficients  $U_K$  and  $A_K$  depend only on the spins of the initial and final states and on the multipolarity of the radiation that is emitted during the transition between them. A number of tabulations of these quantities exist (see e.g. Krane *et al.* 1973). The  $Q_K$  are solid angle correction factors which, for the experimental arrangement used here, have been calculated by Stewart (1976),  $G_K$  are the reorientation coefficients representing the effects of relaxation in the intermediate states, and  $P_K$  are Legendre polynomials.

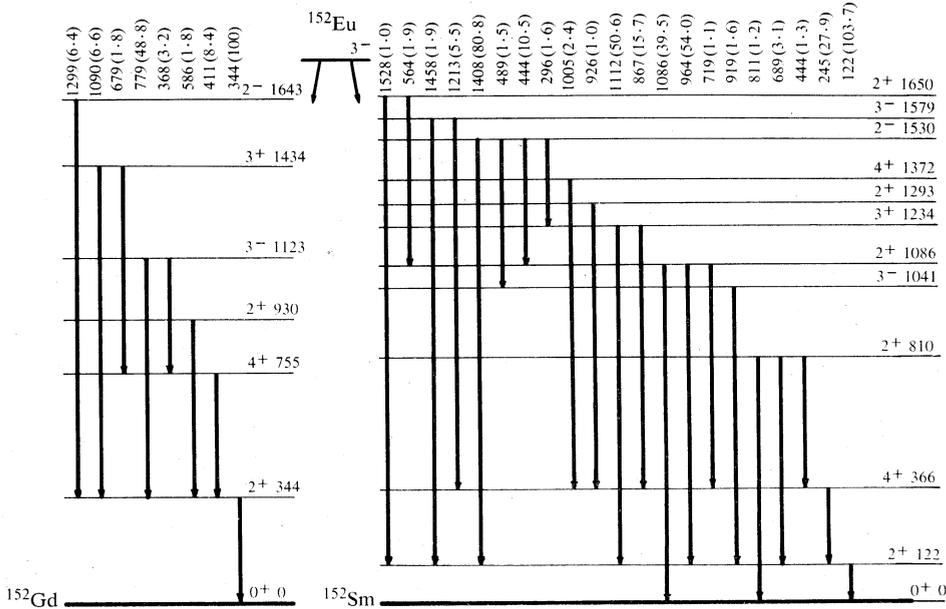


Fig. 1. Decay of  $^{152}\text{Eu}$  to  $^{152}\text{Gd}$  and  $^{152}\text{Sm}$ . Energies of the levels and of the  $\gamma$  transitions are in keV. The relative intensity of each  $\gamma$  transition is indicated in parentheses. Spin and parity of each level are as indicated.

For the observation of  $\gamma$  radiation from an axially symmetric system, the summation in (1) is limited to the even integers  $K = 0, 2, 4$ . If terms for  $K > 2$  are ignored, the intensity of one  $\gamma$  ray in the cascade plotted against that of a second (reference)  $\gamma$  ray will yield a linear graph, with a slope  $\alpha$  given by

$$\alpha = (U_2 A_2 G_2 Q_2)_1 / (U_2 A_2 G_2 Q_2)_R. \quad (2)$$

The advantage of this treatment is that  $B_2(T)$  is eliminated, so that data taken at all temperatures and values of the magnetic field may be readily included in the analysis.

For some  $\gamma$  rays, multipole terms for  $K > 2$  are important. This is evidenced by a deviation from linearity when the  $\gamma$  intensity is plotted against that of the reference  $\gamma$  ray. The data for these  $\gamma$  rays may be corrected, if the ratio  $\alpha$  is interpreted as

$$\alpha = \frac{(U_2 A_2 G_2 Q_2)_1}{(U_2 A_2 G_2 Q_2)_R} + \frac{B_4(T) (U_4 A_4 G_4 Q_4)_1}{B_2(T) (U_2 A_2 G_2 Q_2)_R}. \quad (3)$$

The temperature dependence of  $\alpha$  is isolated in the ratio  $B_4(T)/B_2(T)$ , which is zero at high temperatures. The method of grouping (Keeping 1962) that we have used to evaluate  $\alpha$  places most weight on the points of highest and lowest temperature, so that the  $B$  coefficients in (3) may be evaluated at the lowest temperature used in this experiment (9 mK). Where appropriate, the results presented in Table 1 have been corrected in this manner.

**Table 1. Measured ratios  $\alpha$  and decay scheme parameters derived from the present experiment**

$E_\gamma$ (keV)	$\alpha$	Derived parameters	Results of other authors <sup>A</sup>
<sup>152</sup> Gd			
1299	$0.88 \pm 0.18$	$\delta(1299) = -0.00 \pm 0.08$	$\delta = -0.05 \pm 0.10^C$
779	$-0.72 \pm 0.05$	$\delta(779) = -0.02 \pm 0.02$	$\delta = 0.01 \pm 0.02^C$
368	$0.57 \pm 0.52$	$\delta(368) = 0.1 \pm 0.2$	$\delta = 0.04 \pm 0.08^C$
411	$0.86 \pm 0.20$	$U_2(755) = 0.75 \pm 0.19$	
344	$0.80 \pm 0.05$	$U_2(344) = 0.52 \pm 0.04$ $0.64 \leq Y \leq 1.56^B$	$0.50 \leq Y \leq 0.90^D$
<sup>152</sup> Sm			
1458	$-0.39 \pm 0.18$	$0.03 \leq \delta(1458) \leq 0.14$	$\delta = -0.05 \pm 0.12^C$
1213	$-0.43 \pm 0.28$	$-0.06 \leq \delta(1213) \leq 0.16$	$\delta = 0.03 \pm 0.06^C$
444	$0.88 \pm 0.16$	$0.21 \leq U_2(810) \leq 0.83$ $-0.05 \leq \delta(444) \leq 0.11$	$\delta = 0.03 \pm 0.16^E$
1112	$-0.29 \pm 0.05$	$-0.11 \leq 1/\delta(1112) \leq -0.08^B$	$1/\delta = -0.07 \pm 0.02^C$
867	$-0.32 \pm 0.12$	$-0.17 \leq 1/\delta(867) \leq -0.10^B$	$\delta = -0.14 \pm 0.04^C$
1086	$1.20 \pm 0.06$	$0.73 \leq U_2(1086) \leq 0.77^B$ $1/\delta(1090) \geq 0.04^B$	$\delta(1090) = -0.22 \pm 0.06^C$ or $1/\delta(1090) = -0.01 \pm 0.14^C$
964	$-0.46 \pm 0.06$	$-0.13 \leq 1/\delta(964) \leq -0.06^B$	$1/\delta = -0.12 \pm 0.02$
245	$0.70 \pm 0.07$	$U_2(366) = 0.61 \pm 0.07$	
122	$0.27 \pm 0.06$	$0.20 \leq U_2(122) \leq 0.25$ $G_2(122) = 0.8 \pm 0.3$	$G_2(122) = 0.88 \pm 0.01^C$

<sup>A</sup> Uncertainties in these results have been doubled to approximately convert to 95% confidence limits.

<sup>B</sup> Values corrected for the effects of  $B_4$ .

<sup>C</sup> Value taken from Barrette *et al.* (1970).

<sup>D</sup> Value taken from Alexander and Steffen (1962).

<sup>E</sup> Value taken from Kalfas *et al.* (1973).

### 3. Experimental Procedure

The sample used in this work was the same europium in gold alloy (EuAu) that was used by Barclay and Perczuk (1975), and its preparation has been described by them. An activity measurement showed the europium content to be 2 ppm when the sample was prepared in 1972. At the time of this experiment about 30% of the europium had decayed to either samarium or gadolinium.

Experiments were performed with an adiabatic demagnetization cryostat, in applied fields of up to 2.2 T. The sample temperature was monitored by the use of a <sup>54</sup>MnNi NO thermometer, which showed the lowest temperature reached to be 9 mK. An ND812 mini-computer, run in a pulse height mode, was used to collect the data, and the  $\gamma$  intensities were determined from the resulting spectra by means

of a least-squares fitting procedure. The fitting functions used in this analysis have been described by Barclay *et al.* (1978).

The deduced  $\gamma$  intensities were plotted against a reference  $\gamma$  ray and straight lines were fitted to the data by using the method of grouping (Keeping 1962). This allows for the random nature of both variables. The 1408 keV  $\gamma$  ray was chosen as the reference since it is of high relative intensity and is of almost pure E1 multipolarity. The appropriate  $U_2$  and  $A_2$  coefficients for this  $\gamma$  ray are well documented.

#### 4. Decay Scheme Results

The experimental ratios  $\alpha$  are given in Table 1 together with the derived quantities  $U_2$  and the mixing ratio  $\delta$ . For this latter quantity we have followed the convention of Krane and Steffen (1970). The 1408 keV  $^{152}\text{Sm}$   $\gamma$  ray was chosen as a reference against which the other data were plotted. Nine published values of the angular correlation coefficient  $A_{22}$  for the 1408–122 keV cascade (Barrette *et al.* 1970; Kalfas *et al.* 1973; Helppi and Hattula 1970; Debrunner and Kündig 1960; Ofer 1957) were averaged to give  $A_{22} = 0.2162 \pm 0.0025$ . This yields  $A_2(1408) = -0.473 \pm 0.008$ . The uncertainties given in the present paper are expressed as 95% confidence limits except when stated otherwise. In most cases, standard deviations can be estimated by dividing the extent of the limits by two. Where no published data were available, the full range of possible values allowed by the selection rules was included in the uncertainty of the result.

To calculate  $U_2$  for any level in the decay scheme, the relative intensities of the decay paths leading to that level and the multiplicities of the transitions making up the paths are needed. For some paths, it was necessary to make a number of assumptions in arriving at the deduced values of the parameters  $U_2$ ,  $A_2$  and  $\delta$ . A detailed enumeration of these assumptions is included in Blamey (1980); the relevant material has also been deposited.\*

We are able to set bounds on the deorientation coefficient for the  $\beta$  decay to the 344 keV level of  $^{152}\text{Gd}$  of

$$U_2(344, \beta) = 0.50^{+0.15}_{-0.11}. \quad (4)$$

This result has been corrected for fourth order effects. Several authors (Alexander and Steffen 1962; Appalacharyulu *et al.* 1969) have performed  $\beta$ - $\gamma$  angular correlation and polarization measurements of this cascade. Their results have been expressed in terms of the Kotani (1959) parameters  $Y, x, u, z$ . These are the relative contributions of the matrix elements of a first forbidden  $\beta$  transition;  $Y$  represents the sum of the vector-type components of multipolarity one, so that the total intensity is proportional to  $1 + Y^2$ , where  $Y^2$  is the intensity ratio of multipolarity one to multipolarity two components. Equation (4) is thus equivalent to

$$Y = 0.89^{+0.67}_{-0.25}, \quad (5)$$

which is in agreement with the results of Alexander and Steffen (1962) and Appalacharyulu *et al.* (1969), who obtained  $0.50 \leq Y \leq 0.90$  and  $0.60 \leq Y \leq 0.85$  respectively.

\* Copies of the supplementary material are available on application to the Editor-in-Chief, Editorial and Publications Service, CSIRO, 314 Albert Street, East Melbourne, Vic. 3002.

A reorientation is observed in the 122 keV level of <sup>152</sup>Sm and we find

$$G_2 = 0.8 \pm 0.3. \tag{6}$$

This result is not inconsistent with the value of  $G_2(122) = 0.88 \pm 0.01$  (see Table 1) obtained in the angular correlation measurements of the 366–122 keV cascade (Barrette *et al.* 1970). However, our data were not precise enough to allow meaningful comparisons with other determinations.

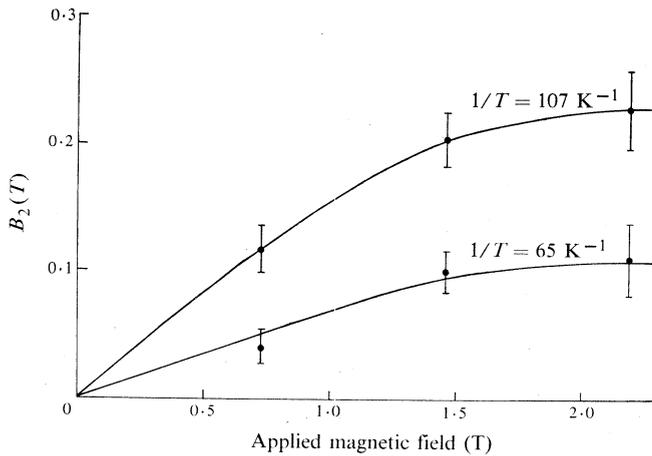


Fig. 2. Orientation parameter  $B_2(T)$  as a function of applied magnetic field.

### 5. Magnetization Curve

By using the intensity of the 1408 keV  $\gamma$  ray,  $B_2(T)$  was found from equation (1). Fig. 2 shows  $B_2(T)$  for two different temperatures as a function of applied magnetic field. It was expected that the magnetization curve should follow the spin  $\frac{7}{2}$  Brillouin curve appropriate for a dilute  $\text{Eu}^{2+}\text{Au}$  paramagnetic alloy. The Brillouin function for spin  $\frac{7}{2}$  reaches 90% of its maximum value when the ratio of applied field to temperature is  $1 \text{ T K}^{-1}$ . The data of Fig. 2 for  $1/T = 107 \text{ K}^{-1}$  have not saturated at  $100 \text{ T K}^{-1}$ .

Gainon *et al.* (1967) have observed ferromagnetic order below 6.5 K for a 0.5 mol%  $\text{EuAu}$  alloy. An autoradiograph of our sample revealed the existence of small regions of higher radioactivity. We estimate the europium concentrations in these regions to be about  $10^{-2}$  mol%, well below that required for ferromagnetic order. In addition a new sample was produced by using a fast quench technique; this showed no evidence of clustering but gave a magnetization curve similar to that of Fig. 2. A more likely explanation is that the europium nuclei are in crystal defect, rather than substitutional sites. They would then experience an anisotropic interaction with the crystal field, and the magnetic interaction would have to overcome this force before saturation was achieved. We can therefore gain no information from our experiment concerning the hyperfine interaction of dilute europium in gold. However, we were able to obtain a good fit to our data by considering a combined magnetic and quadrupolar interaction of the form

$$\mathcal{H} = g_n \beta_n I_z (B_z + B_{\text{EXT}}) + \{3eQV_{zz}/4I(2I-1)\} \{I_z^2 - \frac{1}{3}I(I+1)\},$$

where  $g_n$  is the nuclear  $g$  factor,  $\beta_n$  the nuclear magneton,  $I_z$  the component of nuclear spin  $I$  in the direction of the magnetic field  $B_z$ ,  $Q$  the nuclear quadrupole moment and  $V_{zz}$  the electric field gradient at the nucleus. Using the values obtained by Heinecke *et al.* (1970) of  $g_n = 1.936 \pm 0.002$  and  $Q = 3.14 \pm 0.4 \times 10^{-28} \text{ m}^2$ , we find that

$$B_z = \pm 9.6 \text{ T}, \quad V_{zz} = 1.2 \times 10^{21} \text{ V m}^{-2}.$$

## 6. Conclusions

We have obtained some useful results on the decay scheme parameters for the decay of  $^{152}\text{Eu}$  to  $^{152}\text{Sm}$  and  $^{152}\text{Gd}$ . These results are in good agreement with results achieved by other techniques. Because the measurements of Barclay and Perczuk (1975) were taken with an applied field of only 0.75 T, it is likely that incomplete magnetic saturation of the sample was the cause of the inconsistency in their results for both the decay scheme and hyperfine interaction. Due to the distribution of activity in the sample, our results for the hyperfine interaction given earlier should not be taken as applicable to a *dilute*  $^{152}\text{EuAu}$  alloy. The hyperfine magnetic field at a europium nucleus in an  $\text{EuAu}$  alloy is made up of contributions from core polarization, conduction electron polarization and neighbour effects; the latter contribution is negligible for europium concentrations below 1 mol%. An NO experiment could, in principle, give an estimate of the conduction electron contribution, if the europium concentration and the nature of any other impurities were known. Barclay and Perczuk (1975) attempted such an estimate, but their results should be approached with caution because of the inadequacies of the sample.

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