

## Nuclear Orientation of $\text{La}^{140}$ , $\text{Eu}^{154}$ , $\text{Gd}^{159}$ , $\text{Lu}^{177}$ , and $\text{Lu}^{177m}$ by Sternheimer Antishielding\*

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Five rare-earth nuclei were oriented in neodymium ethyl sulfate and/or cerium magnesium nitrate lattices at very low temperatures, by utilizing the amplification of the crystal-field gradient (by a factor of  $\sim 10^2$ ) provided by Sternheimer antishielding. The following quadrupole coupling constants  $P$  were obtained in the ethylsulfate lattice (units of  $10^{-6} \text{ cm}^{-1}$ ):  $\text{La}^{140}$ ,  $-1.42(12)$ ;  $\text{Eu}^{154}$ ,  $-41.7(28)$ ;  $\text{Gd}^{159}$ ,  $-144(5)$ ;  $\text{Lu}^{177}$ ,  $-67(3)$ ;  $\text{Lu}^{177m}$ ,  $-13.0(12)$ ; and in the double nitrate lattice:  $\text{La}^{140}$ ,  $+1.10(16)$ ;  $\text{Eu}^{154}$ ,  $+1.5(1)$ . The quadrupole moment for  $\text{La}^{140}$  was found to be  $+0.127(13) \text{ b}$ . The ratio of  $Q(\text{Lu}^{177m})/Q(\text{Lu}^{177})$  is  $+2.33(25)$ . The 815-keV  $\gamma$  ray in  $\text{Ce}^{140}$  has  $\delta(E2/M1) = -0.11(5)$ , if taken last in cascade. The signs and magnitudes of  $(g_K - g_R)$  for the  $\frac{7}{2}^- [514]$  band and the  $\frac{3}{2}^- [624]$  band in  $\text{Hf}^{177}$  were determined. Of special interest is the retention of nuclear orientation through the  $\sim 1$ -sec isomeric states in  $\text{Hf}^{177}$ .

### I. INTRODUCTION

THE nuclei of paramagnetic ions in ionic crystals usually experience large hyperfine magnetic fields produced by the unpaired electrons of the ions. The resulting energy splitting of the nuclear-magnetic substates can often be used to achieve nuclear orientation at very low temperatures, the degree of orientation being governed by the temperature and the details of the electron-nucleus interactions.<sup>1</sup> This technique has been especially useful in orienting nuclei of rare-earth ions during the last decade. Indeed a combination of magnetic and quadrupole hfs arising from the  $4f$ -electron configuration should be adequate to orient eleven of the fifteen rare earths ( $\text{La}^{3+}$  through  $\text{Lu}^{3+}$ ); the method has actually been used for ten of these eleven ions. The other four rare earths,  $\text{La}^{3+}$ ,  $\text{Eu}^{3+}$ ,  $\text{Gd}^{3+}$ , and  $\text{Lu}^{3+}$ , are not readily susceptible to this method because they show, in their usual trivalent states, either small hfs arising from  $4f$  electrons [ $\text{Eu}^{3+}$ ,  $4f^6(^7F_0)$ ;  $\text{Gd}^{3+}$ ,  $4f^7(^8S_{7/2})$ ], or none at all [ $\text{La}^{3+}$ ,  $4f^0(^1S_0)$ ;  $\text{Lu}^{3+}$ ,  $4f^{14}(^1S_0)$ ]. It has been found that the quadrupole crystal-field gradient (usually denoted  $A_2^0$ ) is adequate to orient nuclei of all four ions in the rare-earth ethyl sulfate lattice and that  $\text{La}^{3+}$  and  $\text{Eu}^{3+}$  can also be oriented in the "double-nitrate" lattice, because of the amplification of  $A_2^0$  through "antishielding"<sup>2</sup> in the rare-earth ions. The cooling salts neodymium ethyl sulfate (NES) and cerium magnesium nitrate (CMN) were used. A detailed account of the  $\text{Eu}^{3+}$ -in-NES work has been given.<sup>3</sup> A preliminary report of some of the work reported here has also been made.<sup>4</sup> Applications of

antishielded quadrupole crystal-field gradients for nuclear orientation have recently been made to  $\text{Am}^{243}$ ,<sup>5</sup> and to  $\text{Cs}^+$ .<sup>6</sup> It appears that this method should permit orientation of nuclei of several other elements.

In this paper we give a detailed account of orientation experiments on  $\text{La}^{140}$  in NES and CMN,  $\text{Eu}^{154}$  in CMN, and  $\text{Gd}^{159}$ ,  $\text{Lu}^{177}$  and  $\text{Lu}^{177m}$  in NES. Section II contains a brief description of the method. In Sec. III the data for each isotope are presented and discussed. Finally, in Sec. IV a short discussion is given of the applicability of this method for orienting nuclei.

### II. EXPERIMENTAL

The angular distribution of gamma radiation from an oriented assembly of nuclei not subject to intermediate-state reorientation is given in general terms by

$$W(\theta) = \sum_{\nu \text{ even}} B_{\nu} U_{\nu} g_{\nu} F_{\nu} P_{\nu}(\cos\theta). \quad (1)$$

Here we have used the nomenclature of Frankel, Shirley, and Stone<sup>7</sup> where  $B_{\nu}$  are the orientation parameters,<sup>8</sup>  $P_{\nu}(\cos\theta)$  are the Legendre polynomials, and  $F_{\nu}$  are the angular-correlation " $F$  coefficients."<sup>9</sup> The  $U_{\nu}$  describe the effect of the intermediate (unobserved) transitions, and  $g_{\nu}$  are attenuation factors which account for the finite solid angle subtended by the detector. The terms in Eq. (1) usually fall off rapidly with increasing  $\nu$ . For all cases we are to consider here the  $\gamma$ -ray distribution can be adequately described by the first two terms only, i.e.,

$$W(\theta) = 1 + B_2 U_2 g_2 F_2 P_2(\cos\theta). \quad (2)$$

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<sup>1</sup> B. Bleaney, J. M. Daniels, M. A. Grace, H. Halban, N. Kurti, F. N. H. Robinson, and F. E. Simon, Proc. Roy. Soc. (London) **A221**, 170 (1954).

<sup>2</sup> R. M. Sternheimer, Phys. Rev. **84**, 244 (1951); **95**, 736 (1954).

<sup>3</sup> D. A. Shirley and C. A. Lovejoy, Bull. Am. Phys. Soc. **6**, 512 (1961); B. R. Judd, C. A. Lovejoy, and D. A. Shirley, Phys. Rev. **128**, 1733 (1962).

<sup>4</sup> J. Blok and D. A. Shirley, J. Chem. Phys. **39**, 1128 (1963).

<sup>5</sup> N. J. Stone (private communication).

<sup>6</sup> J. Lubbers, A. R. Miedema, and W. J. Huiskamp, Physica **31**, 153 (1965).

<sup>7</sup> R. B. Frankel, D. A. Shirley, and N. J. Stone, Phys. Rev. **136**, B577 (1964).

<sup>8</sup> R. J. Blin-Stoyle and M. A. Grace, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 555.

<sup>9</sup> M. Ferentz and N. Rosensweig, Argonne National Laboratory Report ANL-5324, 1955 (unpublished).

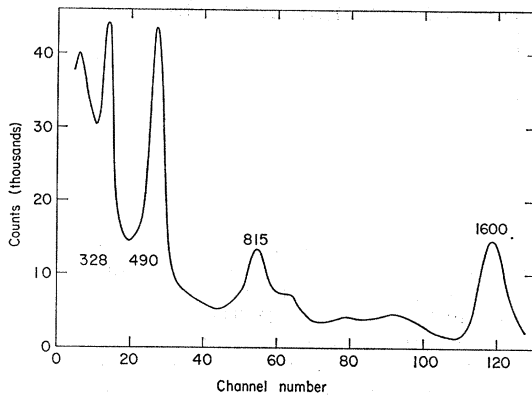


FIG. 1. Gamma-ray spectrum of  $\text{La}^{140}$  with NaI(Tl) detectors. Energies are indicated in keV.

Thus only one observation angle (usually chosen as  $\theta=0$ ) is necessary. We usually placed an additional counter at  $\theta=\pi/2$  as well. The factors  $U_2$  and  $F_2$  can be readily determined from the angular momenta involved in the decay and the factor  $g_2$  has been calculated for NaI(Tl) scintillators.<sup>10</sup>

The energy levels of an ion governed by a spin Hamiltonian<sup>11</sup>  $\mathcal{H}$  may be obtained from the matrix whose elements are

$$H_{ij} = \langle S_z^i I_z^i | \mathcal{H} | S_z^j I_z^j \rangle. \quad (3)$$

The orientation parameters are then defined by<sup>8,12</sup>

$$B_\nu = (2I+1)^{1/2} \sum_{j=1}^N W(E_j) \times \left\{ \sum_{i=1}^N (a_{ij})^2 (-1)^{I-M_i} \langle IIM_i - M_i | II\nu 0 \rangle \right\}. \quad (4)$$

Here  $E_j$  is the  $j$ th eigenvalue,  $a_{ij}$  is the  $i$ th coefficient of the  $j$ th eigenvector in the  $|S_z I_z\rangle$  representation, and  $N$  is the size of the matrix. The population functions  $W(E_j)$  are given by the Boltzmann expression

$$W(E_j) = e^{-E_j/kT} / \sum_{i=1}^N e^{-E_i/kT}. \quad (5)$$

In practice we analyze the experiments implicitly, deducing  $B_2(T)$  from the measured  $W(\theta)$  and deriving hfs constants from  $B_2$ .

The samples, each consisting of a host cooling crystal of NES or CMN with the radioactive isotope isomorphously substituted into lattice sites, were cooled in a glass adiabatic demagnetization cryostat which has

<sup>10</sup> H. E. Gove and A. R. Rutledge, Chalk River Report CRP-755, 1958 (unpublished).

<sup>11</sup> A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1957).

<sup>12</sup> G. A. Westenbarger and D. A. Shirley, Phys. Rev. **123**, 1812 (1961).

TABLE I. Gamma-ray anisotropies observed in the decay of  $\text{La}^{140}$  in CMN at  $1/T=500$ . The data are corrected for background, finite solid angle, and the short half-life of the source.

Gamma energy (keV)	$\frac{3}{2}B_2U_2F_2$
490	+0.0100(27)
815	-0.0180(26)
1597	+0.0194(10)

been described elsewhere.<sup>13</sup> The temperature of the crystal was determined from the susceptibility measured with a 20-cps mutual inductance bridge using the recently determined temperature scales of NES<sup>14</sup> and of CMN.<sup>15</sup> In a typical experiment, stray heating would warm the crystal from the lowest temperatures to the liquid-He bath temperature (about 0.96°K) in about 20 h in the case of NES and in about 90 min. in the case of CMN. For most nuclei, 3-in. by 3-in. NaI(Tl) detectors were used to detect the gamma rays. The extremely complex spectrum of 155-day  $\text{Lu}^{177m}$  required the use of lithium-drifted germanium detectors.

### III. ALIGNMENT RESULTS

#### A. $\text{La}^{140}$

The  $\gamma$ -ray spectrum of  $\text{La}^{140}$  obtained with NaI(Tl) detectors is shown in Fig. 1. The quantity  $\frac{3}{2}B_2U_2F_2$  in CMN at  $1/T \cong 500$  is given in Table I for each of the 490, the 815, and the 1597-keV transitions. From the decay scheme (Fig. 2), it follows that the 490 and the 1597-keV transitions are pure  $E2$  (with calculable  $F_2$ 's of  $-0.448$  and  $-0.597$ , respectively), while the 815-keV transition may be mixed  $M1$  and  $E2$ . Now  $B_2$  is the

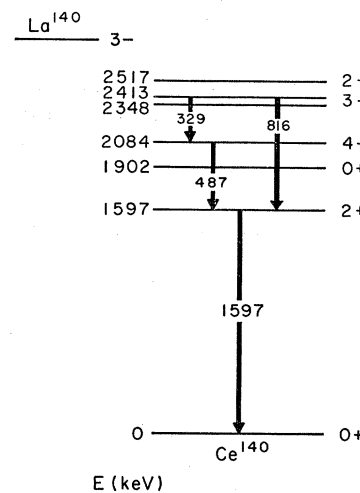


FIG. 2. Partial decay scheme of  $\text{La}^{140}$ .

<sup>13</sup> C. E. Johnson, J. F. Schooley, and D. A. Shirley, Phys. Rev. **120**, 2108 (1960).

<sup>14</sup> J. Blok, D. A. Shirley, and N. J. Stone, Phys. Rev. **143**, 78 (1966).

<sup>15</sup> R. B. Frankel, D. A. Shirley, and N. J. Stone, Phys. Rev. **140**, A1020 (1965).

same for all transitions. Estimating  $U_2 \cong 0.8$ , we find  $F_2(815) = +0.56 \pm 0.08$ . We may compare this result with the angular correlation data for the same gamma ray. Since the 815-keV transition is the first in the cascade for angular correlations and "last" for nuclear orientation, it follows that agreement requires the same magnitude but opposite sign of the  $E2/M1$  mixing ratio,  $\delta$ .<sup>12,16</sup> The angular correlation results of various authors are shown in Table II. We observe fair agreement with Refs. 17 and 18 for our value of  $\delta = -0.11(5)$ . For Refs. 19, 20, and 21 no such agreement is possible, however. The absence of a large  $A_4$  term in the angular-correlation data precludes the alternate choices of strong  $E2$  mixing.

$\text{La}^{3+}$  has no  $4f$  electrons, thus no magnetic hfs interactions are possible. The spin Hamiltonian is simply

$$\mathcal{H} = P[I_z^2 - \frac{1}{3}I(I+1)]. \quad (6)$$

From this Hamiltonian and the value of  $B_2$  measured for the 1597-keV transition we find  $P = +0.110(16) \times 10^{-4} \text{ cm}^{-1}$ . Using nuclear-magnetic-resonance techniques Edmonds<sup>22,23</sup> has determined  $P$  for  $\text{La}^{139}$  in the double nitrate at various temperatures and extrapolating his data to  $T = 0^\circ\text{K}$  we find  $|P_{139}| = 0.186 \times 10^{-4} \text{ cm}^{-1}$ . Abragam and Chapellier<sup>24</sup> have measured  $P_{139} > 0$  in the double-nitrate lattice. Combining this sign with our result, and noting that  $P \propto Q/I(2I-1)$ , we find  $Q_{140} = +0.114(17) \text{ b}$ . We have used  $Q_{139} = +0.27$ ,<sup>25</sup>  $I_{140} = 3$ , and  $I_{139} = \frac{7}{2}$ .

Alignment of  $\text{La}^{140}$  in NES gave only a very small anisotropy, but with the aid of very fast stabilized counting apparatus we determined the anisotropy of the 1597-keV transition quite accurately. We find  $\frac{3}{2}B_2U_2F_2 = -0.0045(4)$  at  $1/T = 88$ . This leads to  $P = -1.42(12) \times 10^{-5} \text{ cm}^{-1}$  when we use  $U_2 \cong 0.8$  as

TABLE II. Mixing ratio for the 815-keV transition in  $\text{Ce}^{140}$  measured by angular-correlation techniques.

Reference	$F_2$	Acceptable values of $\delta$	Agreement with nuclear alignment?
17	+0.166(15)	+0.045,	yes
19	+0.016(15)	-0.09,	no
20	-0.006(9)	-0.09,	no
21	+0.125(32)	$ \delta  < 0.02$ ,	no
18	+0.186	+0.06,	yes

<sup>16</sup> S. Ofer, Phys. Rev. **114**, 870 (1959).

<sup>17</sup> W. W. Black and A. C. G. Mitchell, Phys. Rev. **132**, 1193 (1963).

<sup>18</sup> R. C. Bannerman, G. W. Lewis, and S. C. Curran, Phil. Mag. **42**, 1097 (1951).

<sup>19</sup> W. H. Kelley and M. L. Wiedenbeck, Phys. Rev. **102**, 1130 (1956).

<sup>20</sup> H. H. Bolotin, C. H. Pruett, P. L. Roggenkamp, and R. G. Wilkinson, Phys. Rev. **99**, 62 (1955).

<sup>21</sup> G. R. Bishop and J. P. Perez y Jorba, Phys. Rev. **98**, 89 (1955).

<sup>22</sup> D. T. Edmonds, Phys. Rev. Letters **10**, 129 (1963).

<sup>23</sup> D. T. Edmonds (private communication, 1964).

<sup>24</sup> A. Abragam and M. Chapellier, Phys. Letters **11**, 207 (1964).

<sup>25</sup> Y. Ting, Phys. Rev. **108**, 295 (1957).

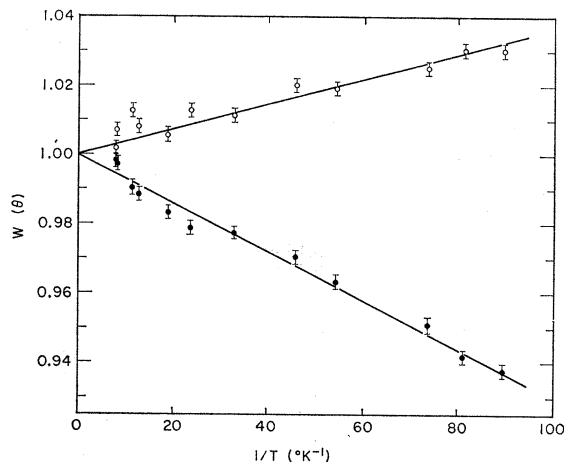


FIG. 3.  $W(\theta)$  for  $\text{Eu}^{154}$  in NES at  $\theta = 0^\circ$  (filled circles) and  $\theta = 90^\circ$  (open circles). The linear dependence on  $1/T$  is clearly illustrated.

before. Edmonds' value for  $\text{La}^{139}$  extrapolated to  $T = 0^\circ\text{K}$  is  $|P_{139}| = 1.98 \times 10^{-5} \text{ cm}^{-1}$ . We thus find  $Q_{140} = +0.140(19) \text{ b}$  from the NES data. An average of the two results yields  $Q_{140} = +0.127(13) \text{ b}$  as the best value.

### B. $\text{Eu}^{3+}$

The alignment of  $\text{Eu}^{154}$  and  $\text{Eu}^{152}$  in NES has been reported previously.<sup>3</sup> We have repeated the experiment, finding  $\frac{3}{2}B_2U_2F_2 = -0.098(7)$  at  $1/T = 90$  for the 1277-keV transition of  $\text{Eu}^{154}$ . The larger anisotropy reported earlier<sup>3</sup> arose from a combination of errors in the old temperature scale for NES and an unrealistically high background correction applied to the data. For  $\text{Eu}^{3+}$  the spin Hamiltonian is represented by Eq. (6) to a good approximation. The best evidence for this is the linearity of  $B_2(T)$  in  $1/T$ . This is shown in Fig. 3. We find  $P = -4.17(28) \times 10^{-4} \text{ cm}^{-1}$  from the measured anisotropy.

The alignment of  $\text{Eu}^{154}$  in CMN similarly gave  $\frac{3}{2}B_2U_2F_2 = +0.019(1)$  at  $1/T = 500$  for the 1277-keV transition. The corresponding quadrupole coupling constant is  $P = +0.15(1) \times 10^{-4} \text{ cm}^{-1}$ .

### C. $\text{Gd}^{159}$

The temperature dependence of the anisotropy of the prominent 362-keV gamma ray of  $\text{Gd}^{159}$  is shown in Fig. 4. This gamma ray is a pure  $E1, \frac{5}{2}^- \rightarrow \frac{3}{2}^+$  transition in  $\text{Tb}^{159}$  preceded by a pure Gamow-Teller  $\frac{3}{2}^- \rightarrow \frac{5}{2}^-$  beta decay from  $\text{Gd}^{159}$ . It is therefore simple to calculate  $U_2 = +0.749$  and  $F_2 = +0.374$ . The spin Hamiltonian for  $\text{Gd}^{3+}$  in the ethylsulfate may be written as<sup>26</sup>

$$\mathcal{H} = g\beta\mathbf{H} \cdot \mathbf{S} + B_2^0P_2^0 + B_4^0P_4^0 + B_6^0P_6^0 + B_6^6P_6^6 + A\mathbf{S} \cdot \mathbf{I} + P[I_z^2 - \frac{1}{3}I(I+1)] + CS_zT_z. \quad (8)$$

<sup>26</sup> K. D. Bowers and J. Owen, Repts. Progr. Phys. **18**, 304 (1955).

TABLE III. Gamma-ray anisotropies for  $\text{Lu}^{177m}$  in NES. The average temperature of the measurements was  $1/T=81$ . The error in the last figure is indicated in parentheses.

Gamma energy (keV)	$\text{Hf}^{177}$ gamma rays		Gamma Energy (keV)	$\text{Lu}^{177}$ gamma rays	
	$B_2U_2F_2$	$B_2U_2F_2$		$B_2U_2F_2$	$B_2U_2F_2$
	Uncorrected	Background corrected		Uncorrected	Background corrected
105.4	+0.091(5)	+0.37(16)	121.6	-0.055(6)	-0.24(14)
113.0	+0.016(5)	+0.048(24)	268.4	-0.175(20)	-0.175(20)
128.5	+0.154(6)	+0.46(23)	318.8	-0.197(18)	-0.197(18)
153.3	+0.210(6)	+0.48(13)	413.6	-0.230(21)	-0.230(21)
174.4	+0.121(7)	+0.27(9)			
208.4	-0.090(5)	-0.090(5)			
228.5	-0.220(8)	-0.220(8)			
249.7	-0.138(11)	-0.138(11)			
281.8	-0.216(11)	-0.216(11)			
296.1	-0.138(43)	-0.138(43)			
327.4	-0.210(16)	-0.210(16)			
378.2	-0.303(13)	-0.303(13)			
418.2	-0.223(18)	-0.223(18)			
465.0	-0.216(64)	-0.216(64)			

The energy spacing of the electronic levels is shown in Fig. 5. In the absence of a magnetic field all terms in Eq. (8) are negligibly small compared to  $B_2^0$  which is given in Ref. 26 as  $B_2^0=0.0067 \text{ cm}^{-1}$ . The operator  $P_2^0$  is defined as  $[3S_z^2-S(S+1)]$ . With this Hamiltonian the doublet with  $S_z=\pm\frac{1}{2}$  will lie lowest by  $0.04 \text{ cm}^{-1}$  as shown in Fig. 5. The  $\text{Nd}^{3+}$  neighbors set up a dipole-dipole field at the  $\text{Gd}^{3+}$  site which can be handled in a straightforward way,<sup>12,13</sup> through the term  $CS_2T_z$  in Eq. (8). This Hamiltonian has been used to calculate the temperature dependence of  $B_2$  in terms of  $P$ . For this calculation we have used  $A=0.5\times 10^{-3} \text{ cm}^{-1}$ ,<sup>27</sup>  $g=1.99$ ,  $S=\frac{7}{2}$ , and  $I=\frac{3}{2}$ . An IBM 7094 computer performed the numerical calculations, giving a satisfactory fit to the experimental data for  $P=-1.44(5)\times 10^{-3} \text{ cm}^{-1}$ .

No observable effect was seen when  $\text{Gd}^{159}$  was aligned in CMN. We can place an upper limit on the anisotropy

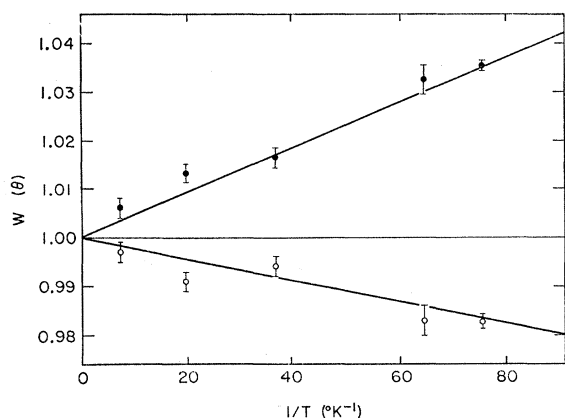


FIG. 4.  $W(\theta)$  versus  $1/T$  for  $\text{Gd}^{159}$  in NES at  $\theta=0^\circ$  (filled circles) and  $\theta=90^\circ$  (open circles).

<sup>27</sup> W. Low, Phys. Rev. **103**, 1309 (1956). We have used the hfs constants for stable  $\text{Gd}^{156}$  and  $\text{Gd}^{157}$  in estimating the correction.

of  $|B_2U_2F_2|<0.004$  at  $1/T=500$ . It is possible, of course, that the  $\text{Gd}^{3+}$  actually does not grow substitutionally into the CMN lattice. For the heavier rare earths, there is always the possibility that most of the activity is merely incorporated in "brine holes" in the crystal interior rather than being an integral part of the lattice. In such a case, the ions would not feel the crystal field and no alignment would be expected.

#### D. $\text{Lu}^{177}$ (7d)

The complete  $\text{Lu}^{177}$  decay scheme is shown in Fig. 6.<sup>28,29</sup> The temperature dependence of anisotropy of the

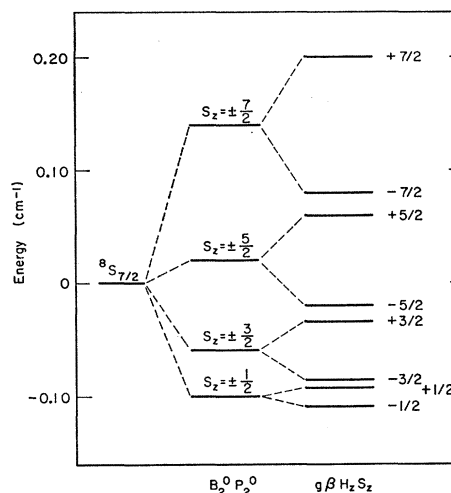


FIG. 5. Spacing of the electronic levels of the  $\text{Gd}^{3+}$  ion in an NES lattice. The terms of the spin Hamiltonian responsible for the splitting are indicated along the abscissa. Here,  $H_z$  represents the dipole-dipole field of 184 G due to the  $\text{Nd}^{3+}$  neighbors.

<sup>28</sup> P. Alexander, F. Boehm, and E. Kankleit, Phys. Rev. **133**, B284 (1964).

<sup>29</sup> J. Blok and D. A. Shirley, Phys. Letters **13**, 232 (1964).

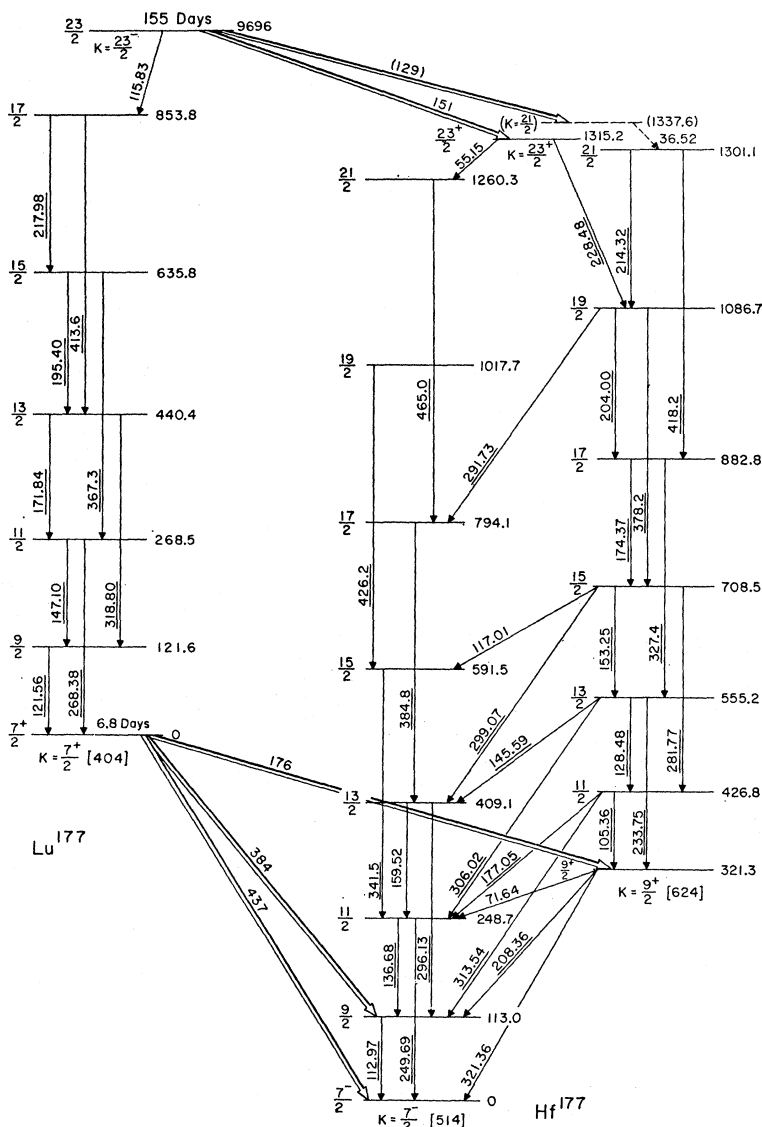


FIG. 6. Decay scheme of  $\text{Lu}^{177}$  from Refs. 28 and 29. Only the underlined transitions were observed in the Ge (Li) spectrum.

208-keV and the 113-keV gamma rays from 7-day  $\text{Lu}^{177}$  aligned in NES are shown in Figs. 7 and 8. We note that the 208-keV peak has pure  $E1$  multipolarity, while the

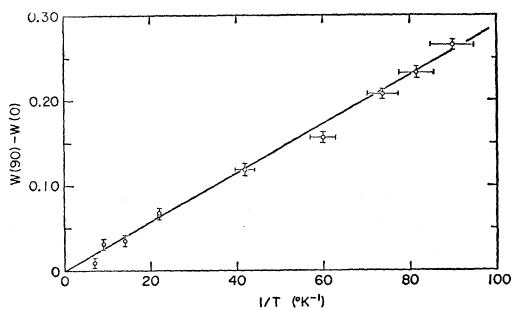


FIG. 7.  $-\frac{3}{2}B_2U_2F_2$  for the 208-keV gamma ray of  $\text{Lu}^{177}$  aligned in NES. The linear dependence on  $1/T$  is evident.

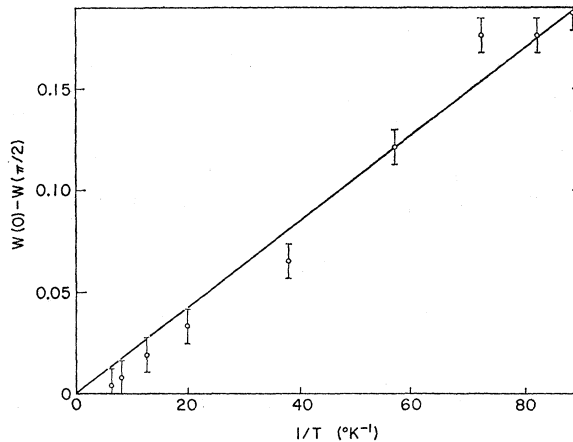


FIG. 8.  $\frac{3}{2}B_2U_2F_2$  for the 113-keV gamma ray of  $\text{Lu}^{177}$  aligned in NES showing the characteristic linear dependence on  $1/T$ .

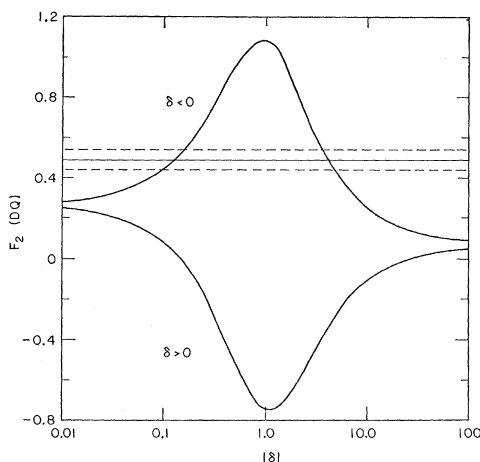


FIG. 9. Theoretical  $F_2$  versus  $\delta$  for the mixed transition  $9/2^-(M1,E2)7/2^-$ , in  $\text{Hf}^{177}$ . The experimental  $F_2$  is shown as a horizontal line with error limits indicated by the dashed lines.

113-keV peak is a mixed  $M1-E2$ ,  $9/2^- \rightarrow 7/2^-$  transition. Using  $U_2 = +0.925$  for the 208-keV transition and  $U_2 = +0.870$  for the 113-keV transition, we find on comparing the anisotropies a value  $F_2^{(113)} = +0.343(34)$ . In Fig. 9 is plotted  $F_2$  as a function of the mixing ratio for this transition. Since transitions in the  $7/2^-$  rotational band of  $\text{Hf}^{177}$  are characterized by a large degree of  $E2$  character,<sup>28</sup> we select the branch giving the larger  $\delta(E2/M1) = -4.7_{+0.4}^{-0.8}$ . From the expression<sup>28</sup>

$$\frac{1}{\delta^2} = \frac{(g_K - g_R)^2 (2I+2)(2I-2)}{Q_0^2 E_\gamma^2} (2.87 \times 10^6). \quad (9)$$

We calculate  $(g_K - g_R)^2 = 0.0012(2)$  using  $Q_0 = 6.74$ .<sup>30</sup> Since the sign of  $(g_K - g_R)$  is the same as that of  $\delta$ ,<sup>31</sup> we find  $(g_K - g_R) = -0.034(1)$  for the  $7/2^-$  [514] rotational band.<sup>32</sup> Using  $g_R = +0.215(14)$  we find  $g_K = +0.18(2)$  in good agreement with Bernstein and de Boer's value of  $g_K = 0.162(10)$ ,<sup>31</sup> but with the sign determined.

The  $\text{Lu}^{3+}$  ion has a completely filled  $4f$  shell so no magnetic interactions are possible. The Hamiltonian is thus the same as for  $\text{La}^{3+}$ , and from the anisotropy of the 208-keV gamma ray we find  $P = -6.7(3) \times 10^{-4} \text{ cm}^{-1}$ .

For  $\text{Lu}^{177}$  aligned in CMN the  $\gamma$ -ray anisotropy, if any, was too small to measure. If the activity actually grew into the rare-earth lattice sites, we may set  $|B_2 U_2 F_2| < 0.003$  at  $1/T = 500$  as an upper limit.

#### E. $\text{Lu}^{177m}$ (155 day)

We have aligned the long-lived isomer of  $\text{Lu}^{177}$  in NES using Ge (Li) detectors to measure the anisotropy of many of the gamma rays in this complex spectrum.<sup>29</sup>

<sup>29</sup> O. Hansen, M. C. Olesen, O. Skilbreid, and B. Elbek, Nucl. Phys. 25, 634 (1961).

<sup>31</sup> E. M. Bernstein and J. de Boer, Nucl. Phys. 18, 40 (1960).

<sup>32</sup> S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 29, No. 16 (1955).

In Table III we show the quantities  $B_2 U_2 F_2$  obtained by counting with one Ge (Li) detector parallel to the crystal axis. We note that there is no noticeable difference in anisotropy between the  $\text{Hf}^{177}$  and the  $\text{Lu}^{177}$  gamma rays. The decay scheme (Fig. 6) requires the cascades to go through two isomeric high- $K$  states in  $\text{Hf}^{177}$ . A technique involving rapid chemical separation from  $\text{Lu}^{177m}$  has shown that at least one of these has a half-life of  $t_{1/2} = 1.06$  sec.<sup>33</sup> It is interesting that there is no reorientation in this isomeric  $\text{Hf}^{177}$  intermediate state during its 1-sec lifetime.

From the anisotropy of the 413-keV gamma ray we have calculated the ratio of the nuclear quadrupole moments of  $\text{Lu}^{177m}$  and of the 7-day ground state,  $\text{Lu}^{177g}$ . For the 413-keV gamma ray of  $\text{Lu}^{177m}$  we have  $B_2 U_2 F_2 = -0.230(21)$  at  $1/T = 81$ . From this value we calculate  $P = -1.30(12) \times 10^{-4} \text{ cm}^{-1}$  using  $U_2 = +0.937$  and  $F_2 = -0.378$ . Since we found  $P = -6.7(3) \times 10^{-4} \text{ cm}^{-1}$  for the 7-day ground state, we obtain

$$\frac{Q^m}{Q^g} = \frac{[PI(2I-1)]_m}{[PI(2I-1)]_g} = \frac{-1.30 (23/2) 22}{-6.7 \frac{7}{2} \times 6} = 2.33(25).$$

Using  $Q_0^g = 7.8$  estimated by Townes<sup>34</sup> we find  $Q^g = 3.6$ . On this basis the quadrupole moment of the  $23/2$  level becomes

$$Q^m = 2.33 \times 3.6 = 8.4(10) \text{ b},$$

from which  $Q_0^m = 10.8(13) \text{ b}$ .

A comparison of the anisotropies of the  $I \rightarrow I-2$  and the  $I \rightarrow I-1$  transitions originating from the same state will yield the  $M1-E2$  mixing ratio for the  $I \rightarrow I-1$  transition. Since  $B_2$  and  $U_2$  are the same for the cascade and the crossover transitions, the ratio of

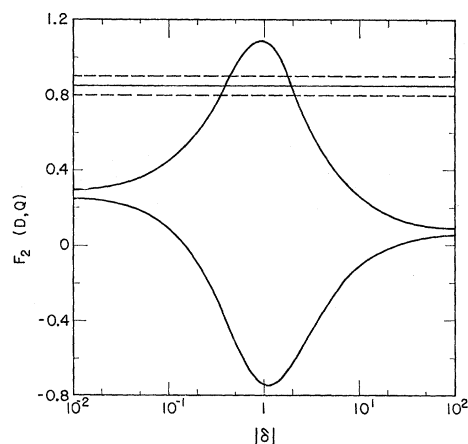


FIG. 10. Theoretical  $F_2$  versus  $\delta$  for the  $15/2^+(M1,E2)13/2^+$  transition in  $\text{Hf}^{177}$ . The experimental  $F_2$  with limits of error is shown as in Fig. 9.

<sup>33</sup> E. Bodenstedt, P. Meyer, L. Schanzler, M. Forker, H. Wagner, and J. Radeloff, Physikal. Verhandlungen 5, 87 (1965).

<sup>34</sup> C. H. Townes, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1958), Vol. 381, p. 377.

the anisotropies equals the ratio of the  $F_2$ 's. Although in principle one can do this for each pair of transitions in each of the three rotational bands populated in the decay of  $\text{Lu}^{177m}$ , the experimental resolution is such that only the 153–281 and the 174–327 pair have both members sufficiently well resolved to measure the anisotropy with good reliability. For the 153 keV,  $15/2 \rightarrow 13/2$  transition we have

$$\begin{aligned} F_2(153) &= \frac{+0.478}{-0.216} F_2(281) \\ &= \frac{+0.478}{-0.216} (-0.3856) = +0.853(54). \end{aligned}$$

Figure 10 shows the ranges of  $\delta$  satisfied by this value of  $F_2$ . The transition is expected to be largely  $M1$  from a comparison with the analogous state in  $\text{Hf}^{179}$ ; thus we favor the smaller value of  $\delta$ . We thus find  $\delta = -0.39(4)$  and from Eq. (9) we calculate  $(g_K - g_R)/Q_0 = -0.049(5)$ . Using  $Q_0 = 6.85$  from the analogous state in  $\text{Hf}^{179}$ , we find  $(g_K - g_R) = -0.335(34)$ .<sup>30</sup> Taking  $g_R = +0.203(34)$  from Bernstein and de Boer for  $\text{Hf}^{179}$  yields  $g_K = -0.13(5)$ . The theoretical value for this  $\frac{9}{2}^+ [624]$  band based on the Nilsson model is  $g_K = -0.35$ . Theory and experiment can be brought into agreement if we choose an effective spin  $g$  factor which is 50% of the free neutron value as suggested by Rasmussen and Chiao.<sup>35</sup>

For the 174 keV,  $17/2 \rightarrow 15/2$  transition we obtain in analogous fashion

$$\begin{aligned} F_2(174) &= \frac{+0.274}{-0.210} F_2(327) \\ &= \frac{+0.274}{-0.210} (-0.3776) = +0.493(52). \end{aligned}$$

<sup>35</sup> J. O. Rasmussen and L. W. Chiao, *Proceedings of the International Conference on Nuclear Structure, Kingston, Canada* (University of Toronto Press, Toronto, 1960), p. 646.

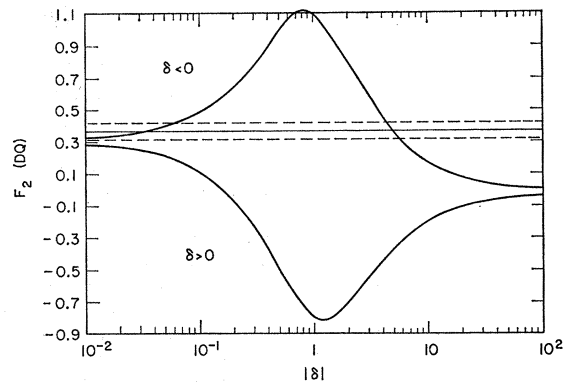


FIG. 11. Theoretical  $F_2$  versus  $\delta$  for the  $17/2^+(M1,E2)15/2^+$  transition in  $\text{Hf}^{177}$ . The experimental  $F_2$  with limits of error is shown as in Fig. 9.

Figure 11 shows the ranges of  $\delta$  satisfied by this value of  $F_2$ . Again using only the smaller value of  $\delta$  we have  $\delta = -0.13(3)$  from which  $(g_K - g_R)/Q_0 = -0.047(11)$ . Using  $Q_0 = 6.85$  and  $g_R = +0.203(34)$  as above we find  $g_K = -0.12(8)$ .

All members of a rotational band should have the same value of  $g_K$ . For the two cases calculated above we find that this is indeed the case for the  $\frac{9}{2}^+ [624]$  rotational band in  $\text{Hf}^{177}$ , to within the accuracy of our measurements.

#### IV. DISCUSSION

The specifically nuclear results are discussed above. Systematic trends for rare-earth ethyl sulfates are discussed elsewhere.<sup>36</sup> We conclude from the foregoing that quite appreciable quadrupole coupling constants can be expected for heavy ions through antishielding of the external crystal-field gradient in noncubic crystals. This nuclear-orientation mechanism<sup>37</sup> is thus of considerably more practical importance than it was previously thought to be.

<sup>36</sup> J. Blok and D. A. Shirley, *Phys. Rev.* **143**, 278 (1966).

<sup>37</sup> R. V. Pound, *Phys. Rev.* **76**, 1410 (1949).