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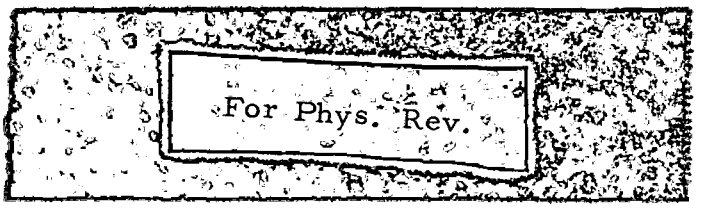

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ABSTRACT

Hyperfine structure, apparently of pure quadrupolar form, was observed in the Eu^{+3} ion in a neodymium ethylsulfate lattice. It has twice the predicted magnitude and opposite sign. Several crystal field effects were considered in attempting to explain the discrepancy, but none were successful. Pseudo-quadrupole effects are shown not to be of primary importance because the coupling constant is negative. For Eu^{152} we find $P^{152} = -(6.7 \pm 0.5) \times 10^{-4}$ cm^{-1} and for Eu^{154} $P = -(8.3 \pm 0.7) \times 10^{-4}$ cm^{-1}. The spin and parity assignments of 2- for the 1531-kev state in Sm^{152} and the 1400 and 1723-kev states in Gd^{154}, as well as the electric dipole multipolarities of the radiations depopulating these states, were confirmed. The quadrupole moment of Eu^{154} was found to be 3.24 ± 0.37 barns, assuming the interaction to be purely electric quadrupole.

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I. INTRODUCTION

One of the significant trends in chemical and atomic physics in the past few years has been the increasing awareness, both experimental and theoretical, of small hyperfine structure effects which cannot be explained by the very simplest models, involving only valence electrons in pure, noninteracting hydrogenlike orbitals. These effects are observed, for example, in some internal magnetic fields, in antishielding, and in the influence on hyperfine structure of higher-order crystal-field interactions and deviations from Russell-Saunders coupling.

We report herein a case in which such subtle effects are clearly present: namely, the existence of hyperfine structure in the ground state of Eu^{+3}. This ion has (in the usual approximation) the electronic configuration $4f^6$, from which it follows by Hund's rule that the ground state is the singlet ^7F_o, with no hyperfine structure possible. The optical spectrum of Eu^{+3} in an ethylsulfate lattice has been thoroughly analyzed by Judd[1], who confirmed this state and assigned several others.

Elliott[2] has shown, using second-order perturbation theory, that a weak electric quadrupole coupling should arise in Eu^{+3} in an ethylsulfate lattice through the V_2^o term in the crystal-field potential, which connects the ground state with the Stark level characterized as $|J=2, J_z = 0\rangle$. He considered the influence of other small quadrupole-like interactions (by this we mean quadrupole and pseudoquadrupole interactions) within the usual crystal-field theoretical framework and found that the contributions of such interactions

were orders of magnitude smaller than this second-order V_2^0 effect.

We have looked for this interaction experimentally in nuclear orientation experiments on radioactive europium isotopes in an ethylsulfate lattice. Hyperfine structure was observed, at least predominantly quadrupolar and of the expected order of magnitude, but of different magnitude and sign.

In the following, the experiments and interpretation are described in some detail. Several nuclear parameters, which are of particular importance in establishing the sign of the coupling constant, must first themselves be independently established. In this process some new nuclear information is gained. Finally, possible explanations of the hyperfine structure are discussed.

II. EXPERIMENTAL

Neodymium ethylsulfate was chosen as a lattice because of its usefulness in nuclear alignment experiments. The crystal field parameters for europium are expected to be nearly the same in this lattice as in europium ethysulfate, from which they were derived by Judd.

The apparatus has been briefly described elsewhere.[3] Care was taken to minimize the heat leak into the sample and to insure that the entire single crystal of neodymium ethylsulfate was at an essentially uniform, constant temperature during the counting period. At the lowest temperatures the average temperature of the crystal changed by only about 1% during a five-minute counting period.

The gamma-ray spectra of Eu^{154} and a mixture of Eu^{152} and Eu^{154} are shown, for reference, in Figure 1. Partial decay schemes showing portions relevant to our discussion are drawn in Figure 2 and 3. In Figures 4 and 5 are shown the temperature dependences of the angular distributions of the

1.277 Mev γ-ray from Gd^{154} and the 1.409 Mev γ-ray from Sm^{152}, following the decay of the oriented europium parents. Finally in Table 1 the anisotropies of the γ-rays from both samples at .02°K are listed and compared with the level schemes in Figures 2 and 3.

III. NUCLEAR INFORMATION

The angular (directional) distribution of γ-radiation following the decay of oriented nuclei is given by an expression of the form[4]

$$W(\theta) = 1 + B_2 U_2 F_2 P_2(\cos\theta) + \ldots . \qquad (1)$$

The dots represent higher-order terms not present in this experiment. Thus only the two terms in Eq. (1) are significant in interpreting these experiments. The first term gives the isotropic intensity, normalized to unity. The term in $P_2(\cos\theta)$, where θ is the angle from the crystalline c axis, describes the anisotropic component. The parameter B_2 is temperature-dependent and is the same for all radiation from a given isotope, being a function only of properties of the parent nucleus. The reorientation parameter U_2 describes the effect of preceeding (usually unobserved) transitions on the orientation. With each γ-ray is associated a distinct F_2 which is a function only of the initial and final spins and of the multipolarity of the transition.

In treating the data to study the level schemes of the daughters we first effectively eliminated B_2 and P_2 from the angular distributions by comparing, for different γ-rays, data taken at the same temperature and at the same angle. Thus the anisotropic component of the angular distribution of each gamma ray is proportional to U_2F_2 for that gamma ray. Knowing one U_2F_2 reliably from other information on the decay scheme, one can then obtain the other U_2F_2's by direct comparison of the anisotropic components of angular

distribution (i.e. the coefficients of P_2). The problem, then, is to establish one U_2F_2 reliably for the decay of each isotope, Eu^{152} and Eu^{154}.

In both spectra one γ-ray stood out as the best from which to derive quantitative results: the 1277-kev γ-ray of Gd^{154} and the 1409-kev γ-ray of Sm^{152} (Figure 1). In each case the photopeak was clearly resolved and the background was quite low. Moreover these γ-rays exhibited the largest anisotropies. Unfortunately the published work was not unanimous in the multipolarity assignments for these two γ-rays or in the spins of the states from which they are emitted. In each case the high energy γ-ray decays to the 2+ state of the ground-state rotational band, and angular correlation measurements have been made on both.[5,6,7,8] In Gd^{154} polarization correlation measurements were also available.[9] The angular correlation coefficients were identical within experimental error, and the other features of the transitions are very similar, each being populated by allowed beta decay from a parent 3- state and decaying to the 2+ first-excited state; thus we shall discuss them together.

The angular correlation data left only two possible combinations for the multipolarities of the high energy γ-rays and the spins of the states from which they proceed: (a) a spin of 2 and essentially pure dipole multipolarity, or (b) a spin of 3 and multipolarity of 85% dipole, 15% quadrupole, with relative phase $\delta < 0$. A determination of the multipolarities of these γ-rays from conversion coefficients would be expected to decide between these two combinations, and indeed the most precise measurements show pure E1 multipolarity in both cases,[10,11] clearly indicating alternative (a) above. On the other hand an earlier measurement gave $\alpha_K \approx 1.7 \times 10^{-3}$ for the 1277-kev γ-ray,[12] consistent with alternative (b). Experimental and theoretical[13] conversion coefficients are given in Table 2.

It is always desirable to overdetermine a set of physical quantities by obtaining one more equation than there are parameters, for only by so doing can one find errors which would otherwise go unnoticed. In this case a very definitive choice can be made between (a) and (b) above from the signs of the F_2's of the 1277- and 1409-kev γ-rays alone. The signs may easily be established by comparing the F_2's of these γ-rays with the F_2's for other γ-rays, of known multipolarity, in the daughter nuclei. In fact it is easily shown (by direct calculation) that the U_2's are all positive, and it thus suffices to compare the signs of the U_2F_2 products. This is done in Table 1, from which it is clear that the F_2's of the high energy γ-rays are negative, which is possible only for alternative (a) above. This then constitutes an independent confirmation of alternative (a).

Angular correlation data[8] but no conversion coefficients are available for the 1600 kev γ-ray in Gd^{154}. Our data (Table 1) establish $F_2 < 0$ for this γ-ray, leading, in a manner similar to that discussed above, to a unique assignment of 2- for the 1723-kev state and E1 multipolarity for the 1600 kev γ-ray.

As discussed before, the 1277- and 1409-kev γ-rays have the most reliably interpreted anisotropies, lying as they do high in the level scheme and in the γ-ray spectrum. The preceding radiation in each case is an allowed beta transition of the type 3-(L=1)2- for which U_2 is +0.828. For the γ-rays themselves F_2 is -0.418. Thus B_2 is readily evaluated using the data in Figures 4 and 5, and is found to be

$$B_2(152) = +(3.45 \pm 0.28)\times 10^{-3}\ T^{-1} \qquad (2a)$$

$$B_2(154) = +(4.28 \pm 0.34)\times 10^{-3}\ T^{-1} \qquad (2b)$$

A T^{-1} temperature dependence of B_2 for low degrees of alignment characterizes hyperfine structure of the quadrupole form. In Figures 4 and 5 we have

fitted our data with curves of this form (straight lines as plotted). The fit, which is quite good, though not excellent, constitutes the chief evidence that the hyperfine structure Hamiltonian has, at least predominantly, a quadrupolar form:

$$\mathcal{H} = P[M^2 - 1/3\ I(I+1)], \qquad (3)$$

where M is the nuclear-spin magnetic quantum number. Combining this with the expression for B_2:[4,14]

$$B_2 = \Sigma \frac{[3M^2 - I(I+1)]\ W(M)}{[1/5\ I(I+1)(2I-1)(2I+3)]^{1/2}}, \qquad (4)$$

we obtain B_2 in terms of P. On comparison with Eqs. 2 we find

$$P_{152} = -\ (6.7\pm0.5)\times10^{-4}\ \text{cm}^{-1}, \qquad (5a)$$

$$P_{154} = -\ (8.3\pm0.7)\times10^{-4}\ \text{cm}^{-1}. \qquad (5b)$$

We note that these values are slightly different from those reported earlier,[15] the major change being an increased magnitude for P_{154}. The present value was obtained using a sample of pure Eu^{154}, thus eliminating the background corrections which were necessary in the earlier experiment because of the presence of Eu^{152}. The change in P_{152} follows from improved thermometry.

If we assume that the quadrupole moment is proportional to P for each isotope, we can derive the ratio

$$Q_{154}/Q_{152} = 1.24 \pm 0.10 \qquad (6)$$

Alpert was given the ratios $|Q_{152}/Q_{151}|$ and $|Q_{152}/Q_{153}|$ as 2.75 ± .17 and 1.08 ± .07, respectively.[16] Krebs and Winkler have recently measured Q_{151} and Q_{153} as +0.95±0.1 and +2.42±0.20 barns, respectively.[17] Combining all these data we find

$$|Q_{154}| = 3.24 \pm 0.37 \text{ barns}$$

It is of course the intrinsic, rather than the spectroscopic, quadrupole moments which are of interest in nuclear theory. Bohr and Mottelson have given the relationship between these two quantities.[18] Making appropriate substitutions and solving their expression explicitly for the intrinsic quadrupole moment Q_o, we obtain

$$Q_o = Q \frac{I+1}{I} \frac{2I+3}{2I-1} \qquad (7)$$

The quadrupole moments derived from the above discussion are listed in Table 3, and intrinsic quadrupole moments derived therefrom are shown in Figure 6. The error bars in Figure 6 give the standard deviation for all sources of error. The relative magnitudes of the Q_o's are known with somewhat better accuracy. In fact the relative Q_o magnitudes are on very firm grounds, inasmuch as they depend only on ratios of hyperfine structure constants. Thus the relative magnitudes would not be subject to change if, for example, one of the sets of measurements (Refs. 16, 17, or this work) should be found to be subject to an antishielding correction heretofore not considered.

Thus we can interpret the trend in Fig. 6 with some confidence as indicating a rather sharp break in Q_o between 88 and 90 neutrons, with a slower, monotonic rise in the three heaviest isotopes. This is in good accord with other data on collective nuclei in this region, and indicates both that the deformation increases with neutron number for these four isotopes and that there is no significant "odd-even" effect.

It should be borne in mind that there are two important assumptions leading to the derivation of Q_{154}; namely (1) that the hyperfine-structure Hamiltonian is adequately represented by Eq. (3), and (2) that Q_{154} is proportional to P_{154}. We believe these assumptions to be true, as there is good evidence,

experimental and theoretical, for them. It is always advisable, however, when using moment values, to bear in mind the assumptions which went into their derivations.

IV. THE FIELD GRADIENT IN Eu^{+3} ETHYLSULFATE

In discussing the possible hyperfine structure in europium ethylsulfate, Elliott considered several types of interaction in several orders of perturbation theory, using the ground state singlet ^7F_o and the higher levels ^7F_J ($1 \leqslant J \leqslant 6$), split into Stark doublets and singlets by the C_{3h} symmetry of the crystal field, as basis functions. Of the interactions which Elliott considered, those which are important in this experiment, in which the external magnetic field was zero, are given by

$$\mathcal{H} = A_2^{\,o} (3z^2 - r^2) + (2\beta\beta_N\mu_N/I) \langle r^{-3} \rangle \vec{N} \cdot \vec{I} \qquad (8)$$

where the first term is that part of the crystal-field potential with $Y_2^{\,o}$ symmetry. The largest contribution to the hyperfine structure was obtained by Elliott using this term in second order perturbation theory. He showed that this would give a coupling constant (Eq. 3) $P = +1.5Q \times 10^{-4}$ cm^{-1}, with Q in barns. This should be corrected to $+1.2\,Q \times 10^{-4}$ cm^{-1} to account for the newer values of $\langle r^{-3} \rangle$ for 4f electrons,[19] which would give 44.8 Å^{-3} for Eu^{+3} rather than the value of 57 Å^{-3} used by Elliott.[2] He also found that the direct interaction of the first term in Eq. 8 gives a P of the order of 10^{-6} cm^{-1}, and that the second term in Eq. 8 should produce a pseudoquadrupole coupling with $P' \sim 10^{-7}$ cm^{-1}. Clearly then the second-order crystal-field term should dominate. In particular P should have the same sign as Q, and for prolately-deformed nuclei, with $Q > 0$, P should be positive.

In fact, as discussed in Section III, P is clearly negative. Two main paths are possible in explaining this, either (1) the quadrupole moments of Eu^{152} and Eu^{154} are actually negative, or (2) the crystal field calculation is inadequate. We cannot say that the quadrupole moments are positive from any direct experimental evidence, but there is a large systematic body of information available for nuclei in this region which strongly suggests that these nuclei are prolately deformed, as are all the neighboring nuclei. Thus we reject alternative (1) above as highly unlikely, and the discussion below is based on alternative(2).

Two approaches which may be taken within the framework of crystal-field theory are the consideration of departures from Russell-Saunders coupling and of configuration mixing, particularly of the type f^5p. B. R. Judd has kindly made rough calculations of these effects which indicate that they are at least one or two orders of magnitude smaller than 10^{-4} cm^{-1}. Internal magnetic fields from the neighboring Nd^{+3} ions can produce a magnetic hyperfine interaction, which is, however, easily shown to be of the order of 10^{-6} cm^{-1}.

It is interesting that any pseudoquadrupole interactions, arising from the second term in Eq. 8 in second-order perturbation theory, will contribute only positive components to P in the ground state. Thus the negative sign of the experimental coupling constant establishes, on purely empirical grounds, that magnetic contributions to P are relatively unimportant.

An effect which is not encompassed by the crystal-field theoretical approach is antishielding. This is a possibility which should be treated theoretically for 4f electrons. Very unusual antishielding factors would be required, but these can probably not be ruled out a priori. Murakawa has empirically estimated an antishielding effect of about 2.5 for 4f electrons[20] which, if applicable to Eu^{+3}, would not explain our results. However as

Murakawa pointed out some of the assumptions on which his derivation was based are very questionable, and the factor applicable to Eu^{+3} could be quite different.

We wish to emphasize that the observed coupling constants, of the order of 10^{-3} cm^{-1}, while small on an absolute scale, are orders of magnitude larger than any theoretical effects of the right sign. If we naively used the value $P = +1.2\,Q \times 10^{-4}$ cm^{-1}, which is the only theoretical effect approaching the experimental results in magnitude, we would obtain a value of -6.9 barns for Q_{154}, which would be completely incompatible with nuclear systematics and the collective model.

Clearly we cannot explain the origins of this hyperfine structure within the framework of the usual crystal field theory. We cannot exclude the possibility that some unanticipated antishielding factor is responsible, and indeed in the absence of any actual evidence to the contrary, this seems the best guess available at present. This would seem to be an interesting and possibly fruitful problem for detailed theoretical investigation, and a theoretical solution would be most welcome. To help stimulate such an investigation, we might point out that the collective nuclear model is on very firm ground, and that this model implies that the intrinsic quadrupole moment, Q_o, of these (europium) nuclei should be in the range +(6-9) barns. Thus this case is very unusual among quadrupole coupling problems in that the quadrupole moment is reliably calculated from nuclear systematics and the coupling constant is known, leaving no ambiguity about the field gradient.

ACKNOWLEDGMENTS

We thank Dr. C. E. Johnson who originated this problem and participated in its early stages, Professor J. O. Rasmussen who has shown continued interest in this and related problems, and Dr. R. J. Elliott, who has contributed several helpful discussions. We are especially indebted to Dr. B. R. Judd who explained some of the subtler crystal field effects and provided quick and reliable estimates of their magnitudes, and to Dr. Seymour S. Alpert who allowed us to quote his data prior to publication.

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* Work done under the aupices of the U. S. Atomic Energy Commission.

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19. B. R. Judd and I. P. K. Lindgren, Phys. Rev. 122, 1802 (1961).

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Table I. Angular distribution coefficients.

Isotope	E_γ (kev)	Spin sequence[a]	$B_2U_2F_2$ at .02°K	$B_2U_2F_2$ (calc) P < 0	$B_2U_2F_2$ (calc) P > 0
Sm^{152}	345	2 (Q) 0	-0.045 ± 0.010	-0.046[b]	+0.023[c]
Gd^{152}	965	2 (Q) 2	+0.015 ± 0.010	+0.010	-0.005
"	1087, 1113	2 (Q) 0, 3 (Q) 2	-0.038 ± 0.010	-0.042	+0.021
"	1409	2 (D) 2	-0.060 ± 0.005	(-0.060)	(-0.060)
Gd^{154}	121	2 (Q) 0	-0.029 ± 0.005	-0.038[d]	+0.019
"	248	4 (Q) 2	-0.050 ± 0.016	-0.062	+0.031
"	593	2 (D) 3	small, > 0	+0.021	-0.010
"	725, 759	2 (D) 2, 3 (Q) 4	-0.066 ± 0.017	-0.056	+0.028
"	875	2 (Q) 2	> 0	+0.023	-0.012
"	998, 1007	2 (Q) 0, 3 (Q) 2	small, < 0	-0.051	+0.026
"	1277	2 (D) 2	-0.074 ± 0.006	(-0.074)	(-0.074)[e]
"	1600	2 (D) 2	-0.057 ± 0.030	-0.074	+0.037

a) Dipole and Quadrupole are denoted by "D" and "Q".

b) Data for Sm^{152} and Gd^{152} are normalized to the 1409-kev γ-ray.

c) Normalized as in b), but using the sequence 3(.85D, .15Q) 2 required for P > 0 (see text).

d) Similar to b), but using the 1277-kev γ-ray for Gd^{154}.

e) As in c), but for the 1277-kev γ-ray.

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Table 2. Conversion coefficients.

Isotope	E	$10^3 \alpha_K$				Ref.
		exp.	E1	M1	E2	
Sm^{152}	1409	.499 ± .025	.47	1.50	1.05	10
Gd^{154}	1277	.72 ± .07	.63	2.1	1.48	11

Table 3. Quadrupole moments of europium

Isotope	Q	Q_o	Ref.
Eu^{151}	+0.95 (10)[a]	+2.66 (28)	17
Eu^{152}	2.61 (20)	6.26 (50)	16
Eu^{153}	+2.42 (20)	+6.78 (56)	17
Eu^{154}	3.24 (37)	7.78 (.88)	this work

a) Standard deviations are given in parentheses. We are responsible for assigning standard deviations to derived quantities.

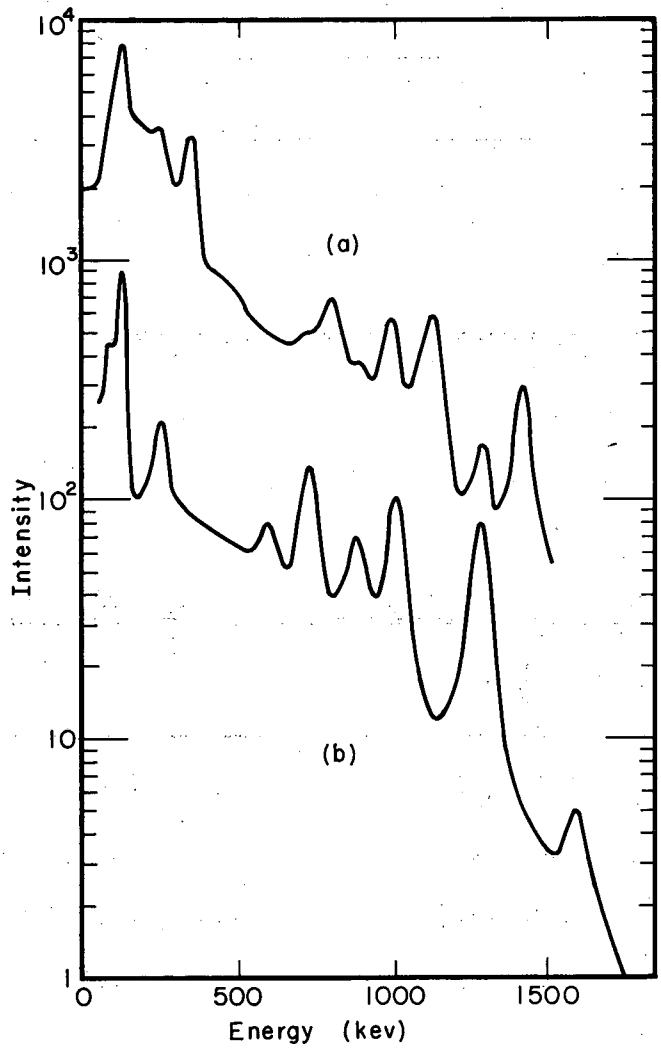

Fig. 1. Gamma-ray spectra of (a) Eu^{152} + Eu^{154}, (b) Eu^{154}. These spectra were taken with 3" x 3" NaI(Tl) detectors and samples consisting of the europium isotopes in single crystals of neodymium ethylsulfate. The γ-ray peaks may be identified in Figures (2) and (3).

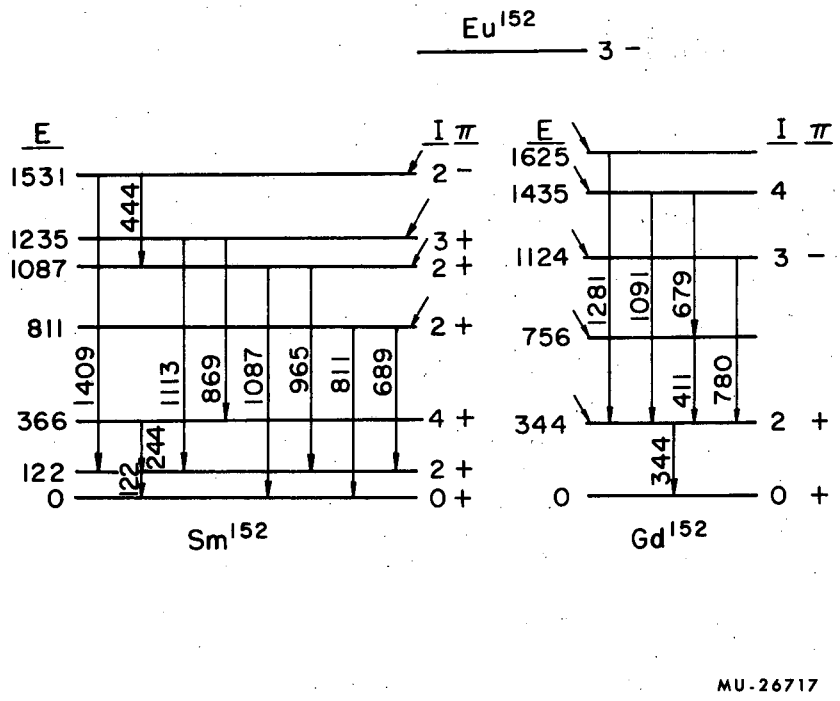

Fig. 2. Partial decay scheme of Eu^{152}, showing transitions of interest in this research. The 2- spin and parity assignment for the 1531 kev state of Sm^{152} was confirmed.

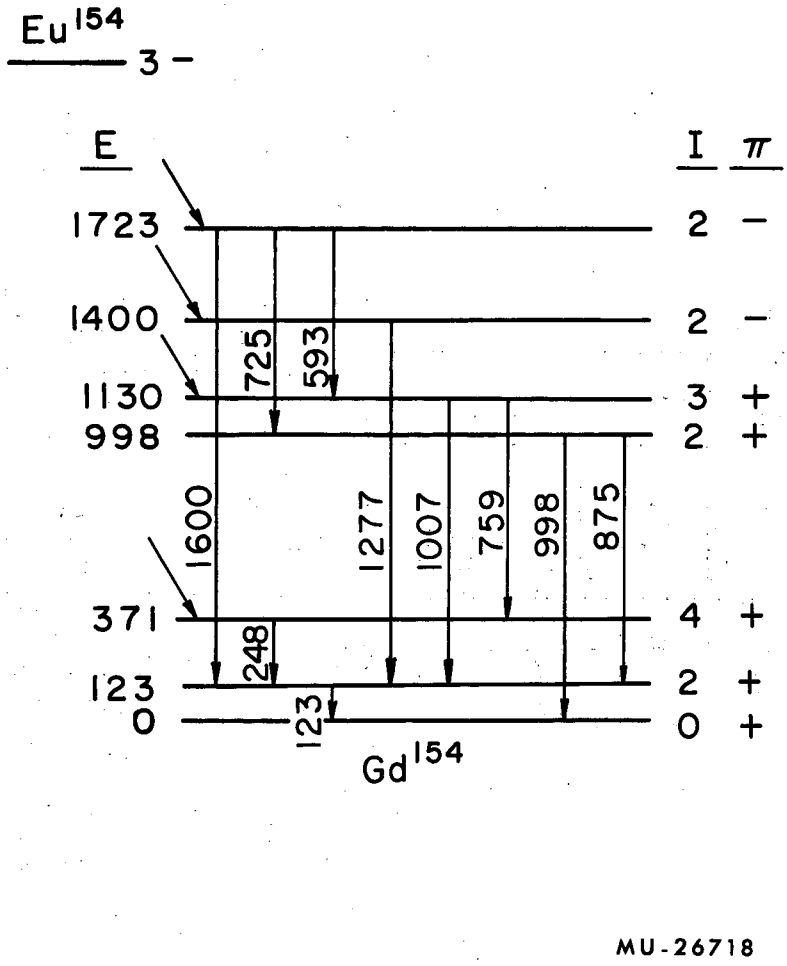

Fig. 3. Partial decay scheme of Eu^{154}. The spin assignments of the states at 1400 and 1723 kev in Gd^{154} were confirmed in this work.

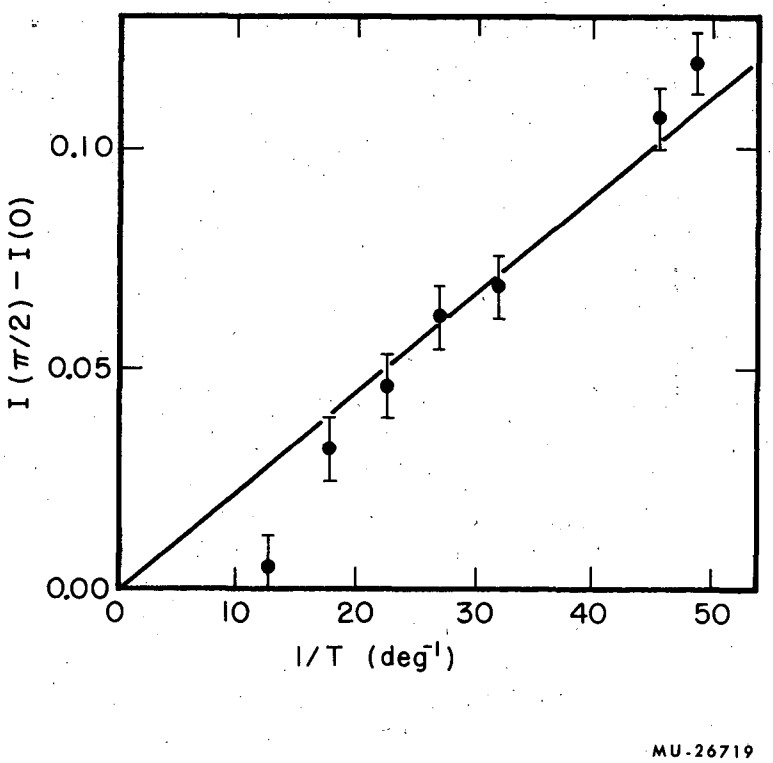

Fig. 4. Angular distribution function for the 1277-kev γ-ray of Gd^{154} following the decay of Eu^{154} oriented in neodymium ethylsulfate, as a function of reciprocal temperature. The function plotted is the difference between the normalized intensities at 90° and 0° from the crystalline axis.

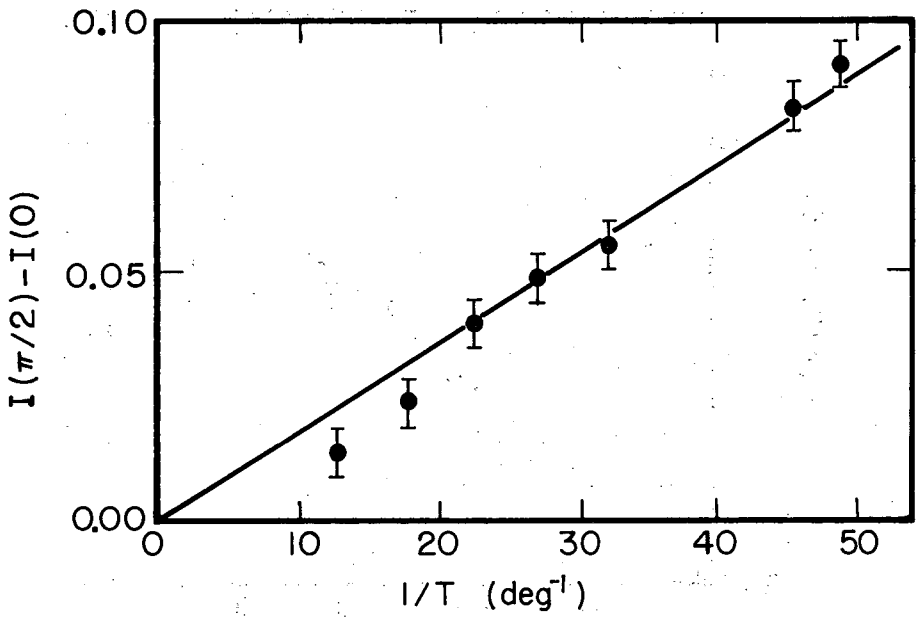

Fig. 5. Angular distribution function for the 1409-kev γ-ray of Sm^{152} following the decay of oriented Eu^{152}, as in Fig. 4.

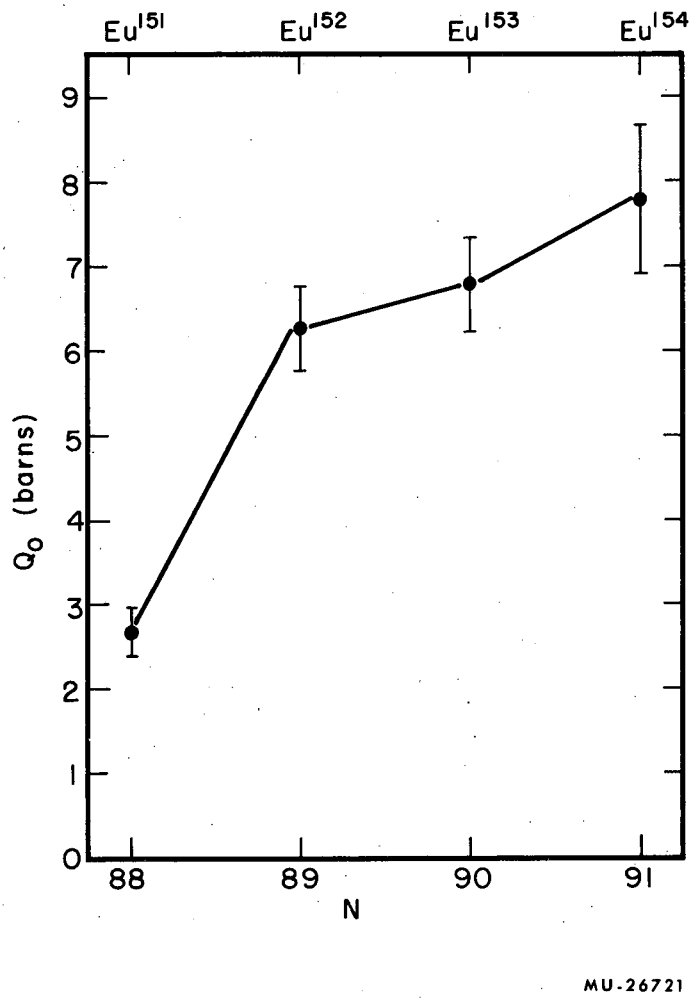

Fig. 6. Intrinsic quadrupole moments vs neutron number for the europium isotopes. Only in Eu^{151} and Eu^{153} are the signs of the Q_o's known; for the even isotopes the Q_o's are assumed to be positive. $(Q_o)_{152}$ and $(Q_o)_{154}$ are known to have the same sign. The total possible error is indicated in each case; the relative magnitudes are known more precisely.