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Alignment of Three Odd-*A* Rare-Earth Nuclei*

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The results of nuclear magnetic hfs alignment experiments on Ce^{139} are presented, and are discussed in relation to those previously obtained for Ce^{141} and Nd^{147}. It is shown that the gamma-ray anisotropies, ϵ, in the case of Ce^{139} are consistent with a decay sequence $3/2^+ \rightarrow 5/2^+ \rightarrow 7/2^+$, the gamma transition being $M1$ with ~4% admixture of $E2$. Even such a small admixture of $E2$ has an important effect upon the anisotropy through the interference term. Other factors which might affect the anisotropy are discussed: electron spin-spin interactions are important, whereas reorientation effects due to long lifetimes in the intermediate state are not. Interference is also important in the case of the 92-kev gamma ray following the decay of Nd^{147}, where ~3% admixture of $E2$ radiation is needed to explain our results. It seems likely also that there is a similar effect in the 142-kev gamma ray following the decay of Ce^{141}. The similarity of these mixtures (same magnitude and phase $\phi = 0$) may not be fortuitous, since in each case the gamma ray connects levels which are classified by the shell model as $d_{5/2}$ and $g_{7/2}$. In the cases of Ce^{141} and Nd^{147} the $g_{7/2}$ state lies uppermost and ϵ is reduced by the $E2$ admixture, whereas ϵ is increased in Ce^{139}, where the levels are inverted.

1. INTRODUCTION

NUCLEAR alignment experiments on Ce^{139} have been carried out by using the same methods as were used in the previously reported experiments[1] on Ce^{141} and Nd^{147}. We present here the results on Ce^{139}, which will be discussed in relation to those on Ce^{141} and Nd^{147}. It has been possible to draw further conclusions from the latter experiments, partly by comparison with present measurements, and partly from the recent determinations[2,3] of the fairly long half-lives of the 166-kev transition in La^{139} (1.5×10^{-9} sec) and of the 144-kev transition in Pr^{141} (1.9×10^{-9} sec). The effect of a long lifetime in an intermediate state can give rise to reorientation effects which, except under very special circumstances,[4] result in an attenuation of the anisotropy of the succeeding gamma ray. For example, the half-life of the 92-kev state of Pm^{147} is 2.44×10^{-9} sec,[5] and we assumed that the complete absence of anisotropy we observed[1] in its emission pattern could be ascribed to this cause. We found, however, significant anisotropies in the 144-kev gamma ray from Pr^{141*} and in the 166-kev gamma ray from La^{139*} which are now known to have half-lives comparable to that of Pm^{147*}.

As was already pointed out,[1] there are two further effects which could play a part in attenuating the anisotropies, namely: interference occurring as a result of a mixed multipole transition (i.e., between $M1$ and $E2$ radiation), and the effect on the population distribution over the initial nuclear magnetic substates of electron spin-spin interaction between the paramagnetic ions. We shall attempt to evaluate the relative importance of these three effects as far as the present state of the theory allows. We begin by describing the experiments and results on Ce^{139}.

2. EXPERIMENTS AND RESULTS ON Ce^{139}

140-day Ce^{139} decays entirely by *K*-capture,[6] predominantly to La^{139*}, and from there to the ground state by emitting a 166-kev gamma ray, which was the one observed in our experiments. The ground state of La^{139} has a measured spin of 7/2[7] and is classified as a $g_{7/2}$ state[8] by the shell model.

Now Ba^{139} decays by β decay to the same excited state of La^{139}; from the log *ft* value of this transition and the probable spin of Ba^{139}, La^{139*} has been assigned a $d_{5/2}$ configuration.[9] This is also in keeping with the $M1$ classification of the 166-kev gamma ray on the basis of *K*-conversion data.[6] Ce^{139} is expected to have a $d_{3/2}$ configuration, and this is supported by the fact that few (if any) transitions are observed directly to the ground state.[6] The decay scheme as given in reference 6 is shown in Fig. 1.

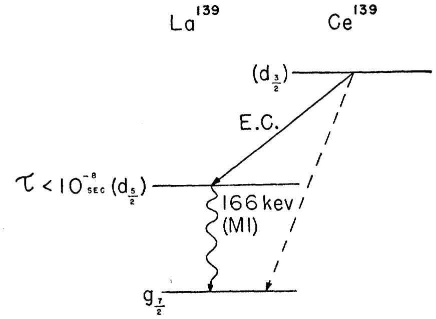

FIG. 1. Decay scheme of Ce^{139} as given in reference 6.

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[*] Supported, in part, by the Office of Naval Research.

[1] Ambler, Hudson, and Temmer, Phys. Rev. **97**, 1212 (1955).

[2] T. R. Gerholm and H. de Waard, Physica **21**, 601 (1955).

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The purpose of carrying out a nuclear alignment experiment with Ce^{139} was to check this decay scheme, especially since no angular correlation measurements are possible.

The isotope was prepared by the $La^{139}(d,2n)Ce^{139}$ reaction,[10] and our scintillation spectrum is shown in Fig. 2. It shows peaks corresponding to the 166-kev gamma ray and the La K x-ray following both K-capture and internal conversion; a small addition peak due to K x-rays (following K-capture) and gamma rays in coincidence can also be discerned. About 60 μC of activity was incorporated into single crystals of cerium magnesium nitrate. The principle of the nuclear alignment, the magnetic cooling techniques, and the gamma-ray counting methods employed, were the same as those previously described.[1]

Results collected from a number of demagnetizations are shown in Fig. 3 where we have plotted the gamma-ray anisotropy

$$\epsilon=[I(\tfrac{1}{2}\pi)-I(0)]/I(\tfrac{1}{2}\pi),$$

against $1/T^2$ where T is the absolute temperature; $I(\theta)$ is the intensity measured along a direction making an angle θ with the axis of quantization (the threefold symmetry axis of the crystal in this case). The value of ϵ at the lowest temperature (0.00308°K) was found to be $+0.105$ with a computed standard error of 0.005 (see Fig. 3). We recall that our salt produces alignment in a plane, i.e., the nuclear magnetic substates $m_I=\pm\frac{1}{2}$ are preferentially populated.

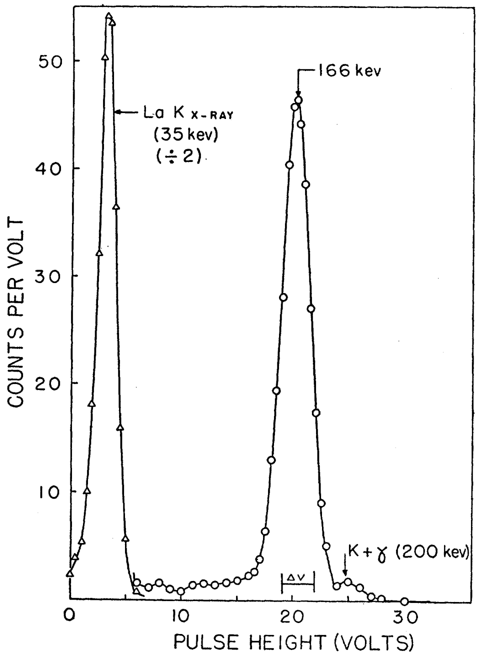

FIG. 2. Gamma-ray spectrum produced by Ce^{139} source. 1 in. ×1 in. NaI(Tl) crystal and 1-volt channel width. ΔV indicates actual channel width and position used during alignment experiments. Peak at 200 kev is addition peak due to coincidence of K x-ray following K-capture and subsequent 166-kev gamma ray. Note reduction factor of 2 in scale for the La x-ray peak at left.

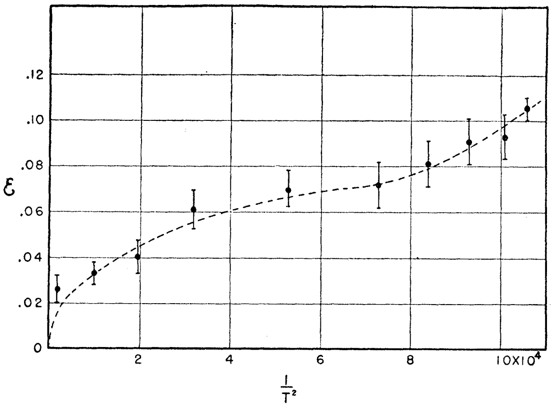

FIG. 3. Anisotropy (ϵ) *vs* reciprocal of square of absolute temperature ($1/T^2$) for 166-kev gamma ray of La^{139}. Points represent averages of 9 demagnetizations. Note smaller statistical error at lowest temperature (see reference 1).

3. DISCUSSION

(a) Idealized Case

The theoretical angular distribution in the case where we neglect spin-spin interaction, reorientation effects, and multipole mixtures, is easily obtained from the following expression[11]:

$$I(\theta)=1+B_2(J_0,T)F_2(LJ_2J_1)U_2(jJ_0J_1)P_2(\cos\theta),$$

where J_0 is the spin of Ce^{139}, J_1 the spin of the 166-kev excited state of La^{139}, J_2 the spin of the ground state of La^{139}, j the angular momentum carried away during K-capture, and L is the multipolarity of the 166-kev gamma ray. The parameters F_ν have been tabulated[12] in connection with γ-γ angular correlation and the parameters B_ν and U_ν are given in reference 11. The effect of the population distribution over the initial nuclear magnetic substates, $W(M)$, enters only into the term B_2; hence the latter is the only term which depends upon the temperature, T.

If the hyperfine splitting of the paramagnetic ion due to the nucleus under investigation had been directly determined, say from paramagnetic resonance measurements (i.e., if the constants A and B in the spin-Hamiltonian were known) the calculation of the various $W(M)$'s would be straightforward.

Unfortunately, paramagnetic resonance measurements have not yet been carried out with Ce^{139}, so that we are forced to estimate the values of A and B from the theory of the Ce^{+++} ion[13,14] and the systematics of

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[10] Ce^{139} was obtained from the Oak Ridge National Laboratory, Oak Ridge, Tennessee.

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[12] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).

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[14] B. R. Judd, Proc. Roy. Soc. (London) **A227**, 552 (1953).

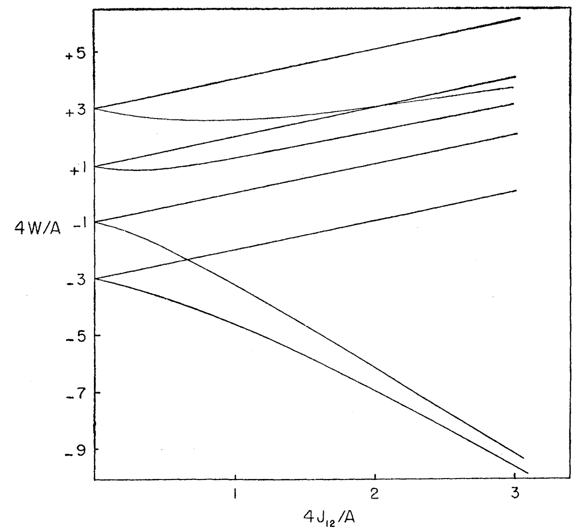

FIG. 4. Energy-level diagram for two paramagnetic ions. Each ion has $S=\frac{1}{2}$, one has hyperfine interaction $AS_ZI_Z(I=\frac{3}{2})$, the other has no hfs. Ions are coupled together by an antiferromagnetic exchange term $4J_{12}\bar{S}_1\cdot\bar{S}_2$, and there are eight unresolved doublets. Their energy, W, is plotted as $4W/A$ *vs* $4J_{12}/A$.

the nuclear moments of odd-neutron nuclei. This value may only be approximate but, as it turns out, is probably adequate for our present purposes. The appropriate formulas were quoted in our previous paper,[1] the result being that $B\gg A$. Further, assuming the magnetic moment of Ce^{139} to be 0.75 nm, we obtain $B=0.021$ cm^{-1}. With this value of B we obtain a theoretical value of $\epsilon=+0.095$ at 0.00308°K. Now this is already smaller than the observed value, even before we correct for attenuation by electron spin-spin interaction. The presence of the latter effect is inferred from the measurements on the 530-kev gamma ray (pure electric quadrupole) following one branch of the decay of Nd^{147}; here the anisotropy was appreciably attenuated, and this could not be accounted for by interference or reorientation. Indeed, we believe that this effect causes attenuation in all our measured anisotropies, and present the following semiquantitative argument in support of this contention.

(b) Effect of Electron Spin-Spin Interaction

We wish to know the effect of interactions between ions on the population distribution over the initial nuclear magnetic substates. The Hamiltonian for the system is

$$\mathcal{H}=\sum_i B(S_{xi}I_{xi}+S_{yi}I_{yi})+\sum_{j>i} G(S_iS_j),$$

where $G(S_iS_j)$ represents the interaction between the ith and jth ions and may be of an exchange or dipolar type, or both. The problem has been treated by approximate methods,[15,16] by obtaining the first few terms in an expansion of the nuclear alignment parameter, $\langle I_z^2\rangle$ as a power series in B/kT. Although it was shown that, up to terms in $1/T^2$, $\langle I_z^2\rangle$ is unaffected by the interaction term, the expansion is only useful when B/kT and $G/kT\ll 1$. This does not hold in our case, however, and we now wish to show that we should expect attenuation at the lowest temperatures.

Consider two ions, one with hfs $AS_{z1}I_{z1}+B(S_{x1}I_{x1}+S_{y1}I_{y1})$, and the other with no hfs. Suppose, first, that these are coupled together by an isotropic antiferromagnetic exchange term, $4J_{12}\mathbf{S}_1\cdot\mathbf{S}_2$. For the particular case of $I=\frac{3}{2}$, $S=\frac{1}{2}$, and $A\gg B$, we have plotted the energy levels of the system in Fig. 4 for increasing values of $4J_{12}/A$. It will be seen that the spacing between the lowest levels steadily decreases as J_{12} increases. For large J_{12} the situation corresponds to the fact that in the antisymmetrical state there is, to first order, no hfs. This effect, at low temperatures, would reduce the nuclear alignment and so attenuate the anisotropy. A similar situation holds for $B\gg A$, except for the additional effect that, since S_{z1} and I_{z1} are not good quantum numbers, mixing between various nuclear levels due to the spin-spin interaction will occur. In the salt we used, namely cerium magnesium nitrate, exchange interaction is known to be negligibly small,[17] but there is still the effect of dipolar coupling. This is complicated by the fact that the ions are magnetically anisotropic, and that the interaction depends upon the relative positions of the ions. The case of $A\gg B$ and $g_{\parallel}\gg g_{\perp}$ (for both ions) is exceptional in that the full hfs persists even for large interactions and no mixing occurs between the nuclear levels. It should be pointed out that in the nuclear alignment experiments where the results have given good agreement with the ideal theoretical case this situation has obtained.[18-22] In the present case, however, where $B\gg A$ and $g_{\perp}\gg g_{\parallel}$, mixing between nuclear levels does occur, and it is reasonable to suppose that if the magnitude of the interaction is comparible with the hfs, the nuclear population distribution would be modified at low temperatures in such a way as to decrease the degree of alignment. In other words, the criterion given by Bleaney,[23] that the hfs should be well resolved, is not fulfilled.

In our case, in the absence of interactions, the lowest energy level has energy $W=-B$ and can be written in $|S_z,I_z\rangle$ quantization:

$$(1/\sqrt{2})\{|-\tfrac{1}{2},+\tfrac{1}{2}\rangle-|+\tfrac{1}{2},-\tfrac{1}{2}\rangle\}.$$

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[16] N. R. Steenberg, Phys. Rev. **93**, 678 (1954).

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[23] B. Bleaney, Phil. Mag. **42**, 441 (1951).

The next higher level is a doublet with energy $W=-\sqrt{3}B/2$ and can be written

$$(1/\sqrt{2}\{|\pm\tfrac{1}{2},\pm\tfrac{1}{2}\rangle-|\pm\tfrac{1}{2},\pm\tfrac{3}{2}\rangle\}.$$

An interaction with another ion with $g_\perp \gg g_{\parallel}$ will cause mixing between these levels, i.e., the nuclear magnetic substates $\pm\frac{3}{2}$ and $\pm\frac{1}{2}$ will be mixed in the lowest level.

To show that the mixing might be appreciable we observe that, in the absence of interactions, the spacing between the two lowest hyperfine levels is $0.134B \simeq 0.0028$ cm^{-1}. The magnitude of the interaction can be estimated from the width of the stable Ce^{+++} resonance line, which has been reported[17] to be Gaussian in shape with a half width of 40 gauss (0.0034 cm^{-1}). Thus the line broadening due to the interactions is greater than the spacing of the lowest hyperfine levels, and we should expect appreciable mixing of nuclear magnetic levels. We conclude that the alignment, and hence the anisotropy, is likely to be attenuated in our experiments.

(c) Interference Due to $M1$-$E2$ Mixtures

Returning now to the results of Ce^{139}, we have seen that the observed anisotropy is significantly larger than would be expected on the basis of a pure $M1$ transition. An explanation for this can be readily found, however, if it is assumed that the gamma transition contains a small admixture of $E2$ radiation, for there is an interference term which produces a considerable change in the anisotropy. It should be pointed out that in previous nuclear alignment experiments, e.g., Co^{60},[18,19] Co^{58},[20] Co^{56},[12] Mn^{54},[22] the final nucleus is even-even and therefore has zero spin in the ground state. Hence transitions to the ground state can never involve mixed radiation. Furthermore, most even-even nuclei have a spin of 2^+ in their first-excited states,[24] so that no dipole radiation occurs.

In general the angular distribution for a mixed transition is given by

$$\begin{aligned}I(\theta)=&\sum_\nu B_\nu(J_0,T)F_\nu(LJ_2J_1)U_\nu(jJ_0J_1)P_\nu(\cos\theta)\\&+\delta^2\sum_\nu B_\nu(J_0T)F_\nu(L'J_2J_1)U_\nu(jJ_0J_1)P_\nu(\cos\theta)\\&+2\delta\sum_\nu(-1)^{J_1-J_2-1}B_\nu(J_0,T)\\&\times[(2J_1+1)(2L+1)(2L'+1)]^{\frac{1}{2}}G_\nu(LL'J_2J_1)\\&\qquad\times U_\nu(jJ_0J_1)P_\nu(\cos\theta),\end{aligned}$$

where $L'=L+1$, the parameters G_ν are tabulated by Biedenharn and Rose,[12] and δ is the ratio of the reduced (real) matrix elements[12] of $(L+1)$ to L radiation, i.e., the ratio of their intensities is equal to δ^2. In our case $L=1$ and $L'=2$.

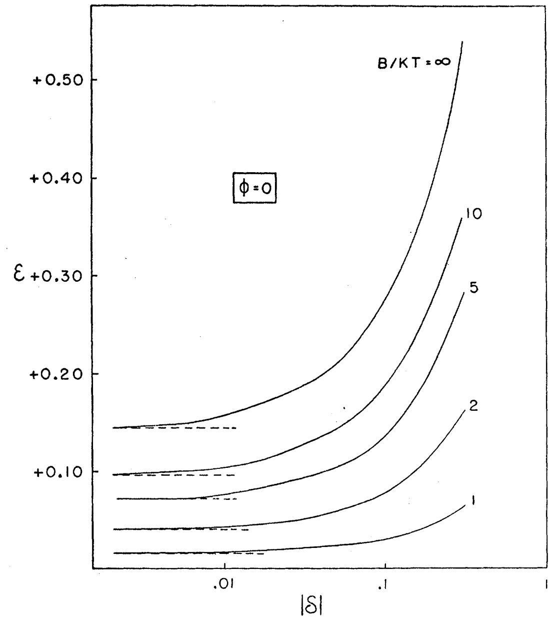

FIG. 5. Curves showing gamma-ray anisotropies, ϵ, for the decay scheme $3/2\to5/2\to7/2$(Ce^{139}), as a function of $|\delta|$ for various values of B/kT. B is the hfs constant; k, Boltzmann's constant; T, absolute temperature; δ, ratio of the reduced matrix elements for $E2$ to $M1$ radiation. Relative phase ϕ is taken to be zero.

We have plotted in Fig. 5 the anisotropy, ϵ, as a function of δ (assumed small), for various values of B/kT. In the absence of any detailed knowledge of the solid-state effects, we do not know what effective value of B to use and we cannot get an accurate value of δ. Only a positive value of δ can *increase* the value of ϵ; referring to Fig. 5, we can see that a value of $+0.2$ would almost certainly be big enough to explain our results. Such a small admixture of $E2$ radiation would not conflict with either the K-conversion data,[6] or the Coulomb excitation data.[25]

We now return to discuss the former results on Ce^{141} and Nd^{147} and, in particular, the fact that with the 92-kev gamma ray following the decay of Nd^{147} no anisotropy was observed. It was pointed out above that this could be explained either by an $M1$–$E2$ mixture or by reorientation effects due to the long half-life of the 92-kev level of Pm^{147} ($t_{\frac{1}{2}}=2.44\times10^{-9}$ sec). But in any case, electron spin-spin interactions could not have been wholly responsible, since a significant anisotropy was observed with the 530-kev gamma ray in the alternate branch of the decay. Recently, the half-lives of La^{139*} following the decay of Ba^{139}, and of Pr^{141*} following the decay of Ce^{141} have been found to be 1.5×10^{-9} sec[2] and 1.9×10^{-9} sec[3], respectively. In these cases the half-lives are of the order of that of Pm^{147*}, and yet significant anisotropies were found.

(d) Effect of Reorientation in the Intermediate State

Reorientation effects have been considered by Steenberg[4] with regard to aligned nuclei, and by a number of authors[26,27] with regard to angular cor-

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[24] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).

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[27] H. Frauenfelder, *Beta and Gamma Ray Spectroscopy* edited by K. Siegbahn (North Holland Publishing Company, Amsterdam, 1955), Chap. 19, 531–599.

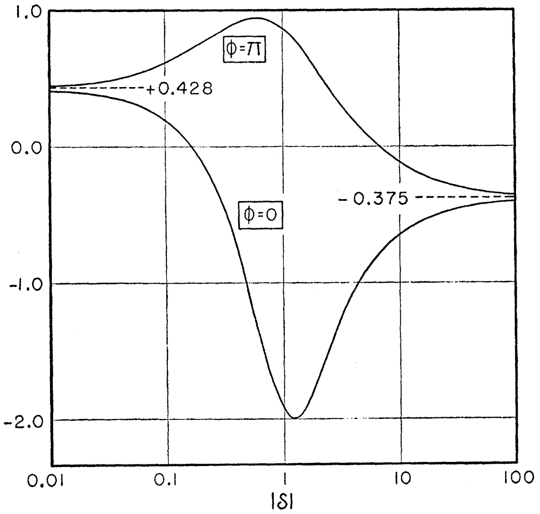

FIG. 6. Effect of $M1$–$E2$ interference on the anisotropy of the 92-kev gamma-ray of Pm^{147} for $T=0$ (see reference 1). Decay sequence $9/2\rightarrow7/2\rightarrow5/2$. Anisotropy, ϵ, is plotted *vs* $|\delta|$, the ratio of reduced (real) matrix elements of $E2$ to $M1$ radiation; note logarithmic scale. The two branches refer to the two possible relative phases (0 or π) of the matrix elements. Dotted line at $+0.428$ represents limiting anisotropy for pure $M1$ radiation; dotted line at -0.375 represents limiting anisotropy for pure $E2$ radiation. Note that ϵ vanishes (for small $E2$ admixture) near $\delta=+0.17$.

relation. The main problem is to find the correct form of the interaction which causes reorientation. In our case the major term will be the coupling of the nuclear magnetic moment with the magnetic field due to the electrons in the outer unfilled shell. Immediately after β decay it is very probable that the configuration of electron shells is the same as before; the work of Migdal[28] and Feinberg[29] shows that, for heavier elements, the probability that the β particle ionizes or excites the outer electrons is very small. Thus the hfs constants of the initial and intermediate nucleus will differ only in so far as their nuclear moments differ, and we shall have $A/B=A'/B'$, where the primes refer to the intermediate nucleus. Of course, in order for the atom to remain neutral it must eventually gain an electron in the magnetic subshell, but the lifetime for this should be of the order of those for optical transitions, i.e., 10^{-8} sec. This, however, is longer than the gamma lifetime.

Assuming, then, that $A/B=A'/B'$, it follows that in the case of Ce^{141} where the spin change during β decay is $7/2\xrightarrow{0}7/2$ no reorientation *at all* takes place. In the case of Nd^{147} we get some reorientation but, since we have a stretched configuration $(9/2\xrightarrow{1}7/2)$, it is only very small. We may estimate the effect by using formula (20) in reference 4. We find that we would expect an attenuation of about 2% whereas we observe essentially complete attenuation.

We therefore consider the effect of $M1$–$E2$ interference. In the case of the 92-kev gamma ray from Pm^{147}, we have plotted in Fig. 6 ϵ *vs* δ for $T=0$. For $\delta=+0.17$ we would have complete isotropy at all temperatures; we suggest that this is the most likely explanation.

In the case of the 144-kev gamma ray following the decay of Ce^{141}, it is not possible to say definitely that interference is important, since the magnitude of the attenuation due to electron spin-spin interaction cannot be predicted. We find, however, that a small admixture of $E2$, again with δ positive, would cause appreciable attenuation.

CONCLUSIONS

An anisotropy of $+0.105\pm0.005$ was observed at 0.00308°K in the angular distribution of gamma rays following the decay of aligned Ce^{139} nuclei. This is in accord with the decay scheme shown in Fig. 1 provided we assume a small admixture of $E2$ radiation ($\delta^2\sim0.04$). The sign of the ratio, δ, of the reduced matrix elements for $M1$ and $E2$ radiation turns out to be positive (i.e., their relative phase is zero).

From a consideration of three effects which could attenuate the gamma ray anisotropy in the cases of the 92-kev gamma ray following the decay of Nd^{147} and the 144-kev gamma ray following the decay of Ce^{141}, we conclude that, despite the relatively long half-life in the intermediate state, reorientation effects are unlikely to have been significant. In the former case, we must again have interference between $M1$ and $E2$ radiation with $\delta^2\sim0.03$. In the latter case, interference could again be significant with δ also positive. Now δ is the ratio of two matrix elements and its sign is determined by their relative phase, which can only be taken to be 0 or π.[30] It is worthwhile pointing out that in two of the cases we consider, and probably the third also, δ is positive; at the same time, on the single-particle model the states which are connected by the gamma ray are in each case $g_{7/2}$ and $d_{5/2}$. (In the case of Ce^{139} these levels are inverted and the interference causes an *increase* in the anisotropy).

The effect of electron spin-spin coupling causes an attenuation of the gamma-ray anisotropy, but is difficult to estimate. If the exact effect were known, then the values of δ could be accurately determined. Thus nuclear alignment could be a useful tool in the accurate determination of mixing ratios. To this end we suggest that it would be worthwhile to repeat these experiments using a different paramagnetic salt, namely, one in which $A\gg B$ and $g_{\parallel}\gg g_{\perp}$ where it is expected that the attenuating effect of spin-spin interaction would be much smaller. It should also be possible, at least in the case of Ce^{141} and Ce^{139}, to determine the values of A and B in the spin-Hamiltonian directly by paramagnetic resonance methods.

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[28] A. Migdal, J. Phys. (U.S.S.R.) 4, 449 (1941).

[29] E. L. Feinberg, J. Phys. (U.S.S.R.) 4, 423 (1941).

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