Intermediate-State Reorientation of Nuclei Aligned in Cerium Double Nitrate*

W. W. STROHM, JR.† AND R. C. SAPP‡
Department of Physics and Astronomy, The University of Kansas, Lawrence, Kansas
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Radiation patterns and linear polarization of gamma rays from decay of Co^{57} and Ce^{141} aligned at low temperatures by Bleaney's method in cerium double nitrate were studied for evidence of intermediate-state reorientation in this crystal. Influence of spin-spin interactions should be negligible on theoretical grounds. Experimental results, expressed in terms of the mixing ratio $\delta=\pm(E2/M1)^{1/2}$ for the $M1+E2$ gamma transitions and of an attenuation coefficient Q_k in the angular distribution $W(\theta)=\Sigma\, Q_k A_k P_k(\cos\theta)$, are as follows: 123-keV gamma from Co^{57} in Ce-Zn nitrate Y sites, $\delta=+0.148\pm0.010$, $Q_2=0.81\pm0.10$ at $1/T^*=157$; 145-keV gamma from Ce^{141} in Ce-Mg nitrate, $\delta=+0.085\pm0.013$, $Q_2=1.08\pm0.08$ at $1/T^*=150$. Comparisons are made with Steenberg's theory of intermediate-state reorientation by static, isotropic hfs coupling in the hard-core limit, and with results of other nuclear alignment experiments involving the same isotopes.

INTRODUCTION

CERIUM double nitrate single crystals have seemed ideal media for low-temperature nuclear orientation of radioactive nuclides in divalent and trivalent ions of the iron and rare-earth groups.[1–3] Unfortunately, every experiment carried out with this salt has shown significant departures from ideal behavior, thereby complicating the interpretation of the data and the extraction of precise nuclear information.[4] These nonideal effects have been thought to result from spin-spin interaction with the cerium ions and, in several cases, reorientation in moderately long-lived intermediate states of the nuclear decay cascade. Such effects as the latter have, of course, also been discussed for certain gamma-gamma angular correlations.[5–7]

To obtain quantitative estimates of the intermediate-state effect in the cerium double nitrate lattice, we have studied alignment of Co^{57} and Ce^{141} nuclei, for which some indications of nonideal behavior already existed.[8,9] Our choice was also guided by the availability of sufficient auxiliary information on the decay schemes, nuclear spins, and hfs couplings for these isotopes to enable us to isolate the departure from ideal behavior with a precision limited mainly by the counting statistics.

Additional interest attaches to the contrasts between the two cases studied. One (Co^{57}) undergoes axial alignment[10] in the Y sites[11] of Ce double nitrate, and decays by electron capture; the other (Ce^{141}) undergoes planar alignment[10] in the trivalent sites and decays by electron emission. Both decays lead to excited states with lifetimes of the order of nanoseconds preceding mixed $M1+E2$ gamma transitions, thus providing the possibility of intermediate state perturbation by extranuclear fields.[12]

We shall show that the influence of intermediate-state reorientation in Co^{57} is appreciable, while in Ce^{141} it is practically negligible. In isolating this effect we have used quantitative theoretical estimates indicating negligible corrections for spin-spin interactions. The experimental results, though of moderate precision, compare surprisingly well with an available theory of perturbation by static isotropic hfs coupling due to Steenberg.[12] These results, and the procedures illustrated, should be of considerable utility in designing and interpreting future nuclear orientation experiments with cerium double nitrate and other paramagnetic salts.

I. EXPERIMENTS

Our experimental apparatus and methods are quite standard,[4] and have been described at length in an earlier publication.[13] Experimental precautions of extrapolating all bridge and counter readings back to the time of demagnetization were observed in order to eliminate the influence of inhomogeneous warm-up of the sample. In the following two sections we shall describe some additional details pertinent to each individual case.

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† Present address: Monsanto Research Corporation, Mound Laboratory, Miamisburg, Ohio.
‡ Alfred P. Sloan Foundation Fellow.
[1] A. H. Cooke, H. Duffus, and W. P. Wolf, Phil. Mag. **44**, 623 (1953).
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[5] H. Frauenfelder, in *Beta and Gamma Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), p. 531.
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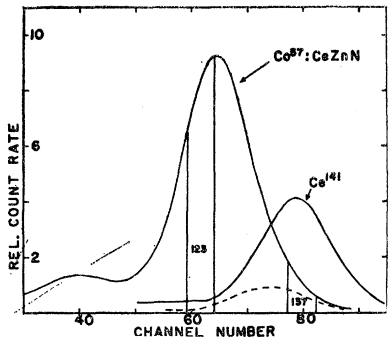

FIG. 1. Multichannel analysis of the gamma spectrum from the decay of Co^{57} in Ce-Zn nitrate at 1°K. The expected contribution of the 10% 137-keV gamma ray was constructed from the Ce^{141} 145-keV spectrum in the same counter. Channels chosen to study the 123 and 137 keV angular distributions are indicated.

A. Co^{57}

Approximately 20 μC of Co^{57} activity, produced by proton bombardment of nickel,[14] was grown into a single crystal of $Ce_2Zn_3(NO_3)_{12}\cdot 24H_2O$. Shape of the sample was that of a 1.1:1.1:0.90 cm ellipsoid with the crystal axis parallel to the minor axis. Magnetic susceptibility was measured along a major axis. Using appropriate demagnetization factors[15] we computed the shape-corrected magnetic temperatures $T^*=T^*(\text{meas})+0.29\times10^{-3}$°K; T^* should be practically equal to T above 6 mdeg K[2].

As no magnetic field was applied following adiabatic demagnetization, axial alignment of Co^{57} at very low temperatures by anisotropic hfs coupling was produced in the Y sites of Ce-Zn nitrates.[16] The X sites in this crystal, being essentially isotropic,[16] should yield negligible alignment in zero field and, hence, an isotropic gamma background. Although the crystallographic $X:Y$ site ratio is 2, cobaltous ions appear to have a slight affinity for Y sites; the ion $X:Y$ ratio deduced from paramagnetic resonance intensities is 1.8 ± 0.2,[13] the value which we shall adopt in this paper.

Radiation patterns $W(\theta)$ were measured, after demagnetizations from 7.0 and 25 kOe to final temperatures of $1/T^*=157$ and 276, respectively, with single-channel analyzers spanning the 123-keV photopeak. Our 1-in.×1-in. cyclindrical-NaI(Tl) counters were shielded by lead collimators to suppress gamma radiation scattered from the tail of the cryostat, the attenuating effect of which is particularly serious for low-energy gamma rays.[17]

The apparent gamma isotropy observed at all temperatures by the Oxford group[8] for Co^{57} in a Tutton salt was found also for the double nitrate. Although exactly zero anisotropy can, in principle, result from $M1+E2$ coherent mixing alone, it is clear that 137-keV $E2$ radiation, which accounts[18] for 10% of the decays from the 137-keV excited state of Fe^{57} and is not resolved by our scintillation counters, could significantly contribute to the apparent absence of anisotropy. In fact, small anisotropies of the order of a few percent, and of the expected signs, were detected when the windows of the single-channel analyzers were narrowed to span portions of the photopeak in which either 123 or 137-keV gamma radiation should predominate. Accordingly, we undertook multichannel analysis of the line shape in order to isolate contributions of the two gamma rays, using an RCL 256-channel analyzer kindly made available to us by Professor R. W. Krone.

Our procedure is illustrated by Fig. 1. Two five-channel portions of the spectrum, chosen to detect principally 123- and 137-keV radiations, respectively, were studied for anisotropy as the sample warmed up. The expected 137-keV line at 1°K was constructed by scaling down, in energy and intensity, the spectrum of Ce^{141} in the same counter (Fig. 1). The 137-keV background in the 123-keV portion was negligible; on the other hand, the temperature-dependent correction to the 137 keV portion from the 123-keV background amounted to about 45%.

The angular distributions at $1/T^*=157\pm8$ are shown in Fig. 2 for the two gamma rays. We shall concentrate on the results at this temperature because the interpretation of the radiation pattern is more straightforward just above the magnetic transition temperature of Ce-Zn nitrate.[13] Least-squares fits to the patterns with Legendre-polynomial expansions yield the following

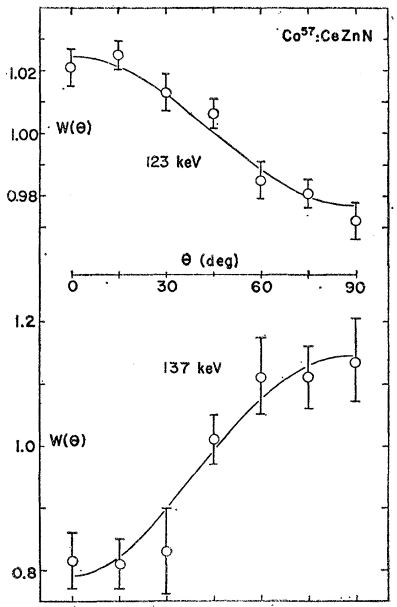

FIG. 2. Radiation patterns $W(\theta)$ of the 123 keV (upper curve) and 137 keV (lower curve) gamma transitions following decay of Co^{57} in Ce-Zn nitrate at $1/T^*=157$. The solid lines are the least-squares fits to Legendre polynomial expansions quoted in the text.

[14] Supplied by Nuclear Science and Engineering Corporation, Pittsburgh, Pennsylvania.

[15] J. A. Osborn, Phys. Rev. **67**, 351 (1945).

[16] J. W. Culvahouse, W. Unruh, and R. C. Sapp, Phys. Rev. **121**, 1370 (1961).

[17] The severity of the scattering effect for low energy gammas was first called to our attention by Dr. D. D. Hoppes of the National Bureau of Standards.

[18] A. T. G. Ferguson, M. A. Grace, and J. O. Newton, Nucl. Phys. **17**, 9 (1960).

results at this temperature:

$$W(123)=1+(0.0324\pm0.0038)P_2,$$

$$W(137)=1-(0.216\pm0.045)P_2-(0.023\pm0.052)P_4.$$

A Compton polarimeter of the type first described by Metzger and Deutsch[19] was utilized for determing the degree of linear polarization of the gammas, and hence the mixing ratio $\delta=\pm(E2/M1)^{1/2}$ of the 123-keV transition. The magnitude and sign of δ have a strong influence on estimating the reorientation effect from data on this gamma ray. As operation of this type of polarimeter has often been described in the literature, we shall omit further description of our particular experimental arrangement except to mention our use of a NaI(Tl) crystal, $\frac{1}{2}$ in. in diameter by 1 in. long, as a scatterer, instead of an organic material.[20] To our knowledge this is the first time that use of sodium iodide for this purpose at so low a gamma energy as 123 keV has been reported; we found that it works quite satisfactorily, in spite of the increased absorption of scattered gamma rays due to the high photo-electric cross section of NaI. The spectrum of Compton electrons in the scatterer in coincidence with gammas scattered into the side crystal showed a hump in the appropriate energy range, with little of the lower energy background often seen in Compton spectrometers.[21] Another advantage of NaI is that its photopeak gives a better energy calibration of the scattering counter than does the Compton edge typically encountered in organic scintillators.

The linear polarization P of the 123+137 keV radiation was determined at $\theta=75°$ and at the same temperature as were the patterns in Fig. 2. The same side counter was placed alternately in (N_{II}), and perpendicular to ($N_{\perp}$), the plane of the alignment axis and the scattering counter axis for successive demagnetizations to $1/T^*=157$. Coincidence rates, averaged over the temperature interval $1/T^*=75$ to 157 to improve statistics and normalized by dividing by the rates at 1°K, were extrapolated to $1/T^*=157$ by fitting with empirical curves of the form $1+a_2B_2(T)+a_4B_4(T)$, where $B_k(T)$ are the orientation parameters tabulated in Ref. 4. This extrapolation, illustrated in Fig. 3, was carried out to minimize inhomogeneous temperature distribution effects; it offers a procedural compromise between, on the one hand, using unextrapolated rates which necessarily correspond to a large temperature interval and, on the other, making a linear extrapolation which might well overestimate the initial influence of temperature inhomogeneity on the coincidence rates. Normalization corrects for possible small residual misalignment of the polarimeter axis. The results are

$$N_{\text{II}}/N_{\perp}=0.693\pm0.090 \text{ at } 1/T^*=157.$$

The statistical error quoted also spans the uncertainties in normalization and extrapolation procedures, but does not incorporate them.

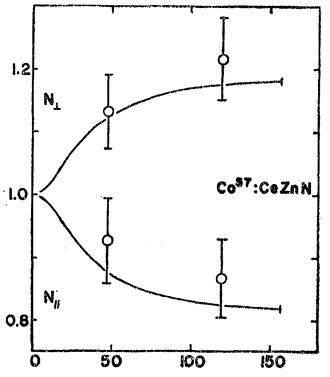

FIG. 3. Normalized coincidence rates N from Compton polarimeter at $\theta=75°$ for (123+137)-keV gamma radiation from Co^{57} in Ce-Zn nitrate following magnetic cooling to $1/T^*=157$ and subsequent warmup. $N_{\perp}$ corresponds to side counter perpendicular to plane of gamma-ray direction and alignment axis; N_{II} is for side counter in this plane. The empirical curves, with the appropriate theoretical temperature dependence, are used to extrapolate the data to conditions just after cooling.

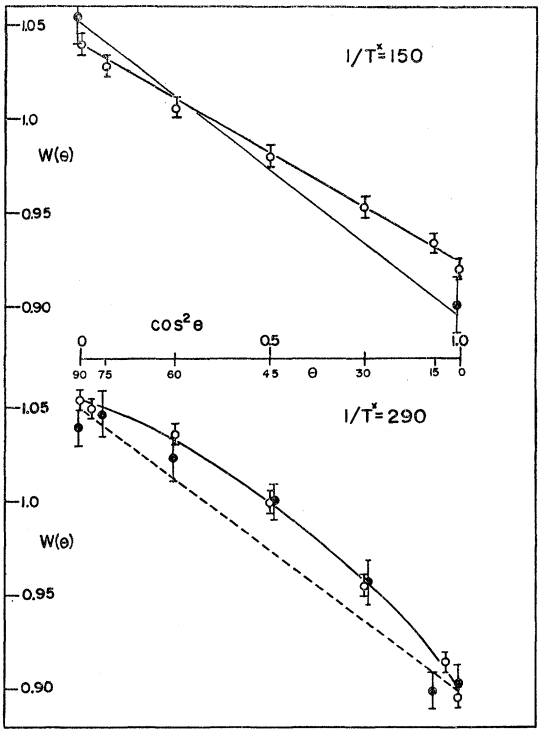

FIG. 4. Radiation patterns $W(\theta)$ of 145-keV gamma rays from decay of Ce^{141} in Ce-Mg nitrate at $1/T^*=150$ (upper curves) and 290 (lower curves). The open circles are data taken with pulse-height selector windows spanning full peak of spectrum; the solid points correspond to data with windows across only the upper half of the spectrum to minimize detection of scattered radiation.

[19] F. Metzger and M. Deutsch, Phys. Rev. **78**, 551 (1950); see also the excellent review article by L. W. Fagg and S. S. Hanna, Rev. Mod. Phys. **31**, 711 (1959).

[20] This was suggested to us by Professor P. S. Jastram of Ohio State University.

[21] See, for example, H. J. Rose, F. Uihlein, F. Riess, and W. Trost, Nucl. Phys. **36**, 583 (1962).

Finally, we obtain P from the relation

$$P=(N_{\parallel}-N_{\perp})/(N_{\parallel}+N_{\perp})\bar{r},$$

where $\bar{r}=[\bar{\sigma}(80,90)-\bar{\sigma}(80,0)]/[\bar{\sigma}(80,90)+\bar{\sigma}(80,0)]$ is the analyzing factor for our polarimeter geometry. Here $\bar{\sigma}(\bar{\beta},\bar{\psi})$ is the Compton cross section averaged over angular ranges of the scattering angle β and azimuthal angle ψ determined by the crystal geometries and pulse-height selector settings of the scattering counter and the side counter. Performing a simple unweighted average over the angles, which in our case were estimated to be $\beta=80°\pm(20°\pm5°)$, $\psi=0°$, $90°\pm(25°\pm5°)$, we obtain $\bar{r}=0.76\pm0.04$. Therefore, $P=-0.240\pm0.040$ at $1/T^*=157$, with the quoted uncertainty determined by counting statistics; the uncertainty in $\bar{r}$ has not been folded in, but lies within this range.

B. Ce^{141}

A spherical single crystal of $Ce_2Mg_3(NO_3)_{12}\cdot 24H_2O$ containing initially about 10 μC of Ce^{141}, produced by neutron bombardment at Oak Ridge of CeO_2 and converted to cerous nitrate, was studied. The specimen shape should insure equality of T^* and T at $1/T^*=150$. Planar alignment in zero magnetic field was induced after magnetic cooling below 1°K. No multichannel analysis was employed in this experiment, but the technique of setting the pulse selector windows on the upper half peak to discriminate against scattered radiation was utilized. Collimation would have been preferable because the high threshold decreases the counter stability and the counting rate unnecessarily.

The angular distributions of 145-keV gamma radiation with the full peak and upper-half-peak window settings, after 7.0 and 25 kOe demagnetizations to $1/T^*=150$ and 290, respectively, are shown in Fig. 4. We find that at $1/T^*=150$, the pattern is well accounted for by a P_2 term alone—consistent with the coefficient of P_4 being very small when the $E2$ admixture is small. On the other hand, a most interesting result is that the pattern at $1/T^*=290$ departs strongly from a P_2 distribution; nor can it be fitted by the addition of a P_4 term. Note that we were careful to verify that scattering is not causing this. Unfortunately, because of our lack of understanding of the magnetically ordered state in Ce-Mg nitrate, we are unable to say whether this pattern might be a consequence of spin-spin interaction.[13] Therefore, we shall concentrate our analysis on the higher temperature pattern:

$$W(145)=1-(0.106\pm0.011)P_2.$$

At $\theta=90°$ the linear polarization of the 145-keV gamma was determined using a chronologically earlier version of the Compton polarimeter discussed in Sec. IA. The principal difference lay in the use of a 1-in.×1-in. Pilot B scatterer. The window on the scattering counter was set across a low hump in the expected energy range of the coincidence spectrum as a function of electron energy, and the entire coincidence output in this range, corrected only for accidentals, was taken to be a measure of the polarization. In this experiment a large low-energy background, which we tentatively ascribe to multiple scattering, was present in addition to the hump. However, multiple scatterings which could satisfy the energy limitations imposed by the pulse height selectors ought to have roughly the same polarization sensitivity as does the single Compton scattering process upon which the polarimeter's operation is based. Therefore, we applied no multiple-scattering corrections or background subtractions to our data.

Applying the same averaging, normalizing, and extrapolating procedures as were discussed for Co^{57} previously to the relative rates $N_{\parallel}=1.200\pm0.060$, $N_{\perp}=0.783\pm0.040$ in the interval $1/T^*=66-150$, we found

$$N_{\parallel}/N_{\perp}=1.65\pm0.10 \text{ at } 1/T^*=150.$$

Scattering angle ranges for this experiment were estimated to be $\beta=90°\pm(15°\pm5°)$, $\psi=0°$, $90°\pm(32°\pm5°)$, which yield $\bar{r}=0.71\pm0.04$. Thus, we obtain $P=+0.350\pm0.033$ at $1/T^*=150$.

II. DISCUSSION OF RESULTS

Our analysis of the experiments just described will be based on the expression for the angular distribution of linearly polarized $M1+E2$ gamma radiation from aligned nuclei in the form given by Blin-Stoyle and Grace[4]: the relative intensity per unit solid angle emitted in a direction making an angle θ with the axis of alignment and with plane of polarization at an angle ϕ with the plane containing θ is

$$W(\theta,\phi)=1+\sum_k \lambda_k B_k U_k Q_k \times[F_k' J_k P_k(\theta)+F_k'' J_k'' P_k^{(2)}(\theta)\cos 2\phi]. \quad (1)$$

In this formula $k=2,4$. B_k is the temperature-dependent orientation parameter calculated, assuming thermal equilibrium, from the Boltzmann populations of the hfs energy levels characterized by the spin Hamiltonian

$$H=AI_zS_z+B(I_xS_x+I_yS_y). \quad (2)$$

A number of useful special cases are tabulated in Ref. 4.

The factor U_k represents the effect of an unobserved preceding transition—in the present cases, a beta transition—and are proportional to Racah coefficients. F_k' and F_k'' are factors dependent on the parameters of the gamma transition, and quadratically on the degree of coherent mixing δ. They are easily evaluated using the tables of Ferentz and Rosenzweig.[22]

In Eq. (1) the $P_k(\theta)$ and $P_k^{(2)}(\theta)$ are the Legendre and associated Legendre functions, respectively. J_k and J_k'' are factors correcting the intensity distribution for effects of finite detector acceptance solid angle; the graphs presented by Stanford and Rivers[23] are useful in estimating these corrections.

[22] M. Ferentz and N. Rosenzweig, Argonne National Laboratory Report ANL-5324, 1955 (unpublished).

[23] A. L. Stanford and W. K. Rivers, Rev. Sci. Instr. **30**, 719 (1959).

Spin-spin interactions are introducted by the temperature-dependent factor λ_k. This mode of representation contains the implicit assumption that the interaction of the ion containing the aligned nucleus with all its neighbors neither destroys the axial symmetry of the nuclear radiation pattern nor introduces another axis into the pattern. (A separate investigation of the cylindrical symmetry of the radiation pattern substantiated this assumption to within statistics.)

Finally, we represent intermediate state reorientation by a temperature-dependent factor Q_k, which should also depend on the perturbing mechanism and on the mean life of the intermediate state τ. Again, this assumes that the cylindrical symmetry of the radiation pattern is not altered by the reorientation mechanism.

In this paper we shall employ conventional definitions of anisotropy ϵ and degree of linear polarization P, which are:

$$\epsilon=[W(90,45)-W(0,45)]/W(90,45), \quad (3)$$

$$P(\theta)=[W(\theta,90)-W(\theta,0)]/[W(\theta,90)+W(\theta,0)]. \quad (4)$$

We now proceed to the interpretation of results for the two nuclides individually.

A. Co^{57} in Ce-Zn Nitrate

The radiation pattern $W(\theta,45)$ of the 137-keV gamma may be written

$$W(137)=yW(Y,137)+(1-y)W(X,137)$$
$$=1+y\lambda_2B_2U_2Q_2F_2J_2P_2+y\lambda_4B_2U_4Q_4F_4J_4P_4, \quad (5)$$

where y=fraction of Co^{57} in Y sites$=0.357\pm0.025$; B_k are the orientation parameters[4] for Co^{57} in the Y sites at $1/T=157$, using $I=\frac{7}{2}$, $A(Y)/2k=0.0209°K$, $B(Y)=0$ in Eq. (2); $J_2=0.974$, $J_4=0.920$ in our counter geometry; $F_2(137)=-0.53452$, $F_4(137)=-0.61721$. We assumed the beta decay to be of the allowed unfavored type[24,25] $\frac{7}{2}(1)\frac{5}{2}$, for which $U_2=0.875$, $U_4=0.581$.

The pattern of Co^{57} radiation from the isotropic X sites has been set equal to unity in Eq. (5), which contains the presumptions that neither spin-spin interactions nor intermediate state reorientation disturb the spherical symmetry of the X ion at the temperature of this experiment. The first can be justified empirically, in that the anisotropy of Co^{60} gamma rays showed negligible departure from the ideal curve down to the transition point of Ce-Zn nitrate.[13] The second presumption is at least plausible from Steenberg's theory[12] of reorientation, which predicts no effect from the mechanism he treated (static isotropic hfs coupling) if the X-site environment for both the initial and intermediate nuclear states is indeed spherically symmetric. Finally, we set $\lambda_2(Y)$ and $\lambda_4(Y)$ equal to unity, since $A(Y)\gg B(Y)$ makes this ion relatively insensitive to interactions.[13]

Comparing the theoretical pattern at $1/T=157$,

$$W(137)=1-0.239Q_2P_2-0.0848Q_4P_4, \quad (5')$$

with the observed pattern quoted in Sec. IA, we obtain $Q_2=0.90\pm0.19$, $Q_4=0.27\pm0.60$. The rather large uncertainties are consequences of the relatively large subtractive corrections for 123-keV radiation in the 137-keV channels, and of the lower number of counts recorded in these channels.

For the 123-keV gamma ray,

$$W(123)=yW(Y,123)+(1-y)W(X,123)$$
$$=1+y\lambda_2B_2U_2Q_2F_2'J_2P_2+y\lambda_4B_4U_4Q_4F_4'J_4P_4, \quad (6)$$

where all the symbols have the same meaning as before; $F_2'(123)=(0.37417-1.89737\delta-0.19090\delta^2)/(1+\delta^2)$, $F_4'=0.70539\delta^2/(1+\delta^2)$. In order to get an idea of the value of δ, let us first consider the condition that the 123-keV pattern be isotropic, i.e., $F_2'=0$; the corresponding $\delta=+0.19$. Next, consider the combined pattern

$$W(123+137)=bW(123)+(1-b)W(137). \quad (7)$$

Here the intensity branching fraction of 123-keV radiation $b=0.90\pm0.02$.[18] This pattern would be isotropic at $1/T=157$ if $F_2'=-(1-b)F_2/b=0.0620$, implying $\delta=+0.16$. Of course, these approaches offer no chance to determine Q_2, and, in fact, represent rough approximations to the real situation. Therefore, we determine δ more accurately by combining the polarimeter and angular distribution data.

The total linear polarization at angle θ and temperature T is

$$P(123+137)=-y\{\lambda_2B_2U_2Q_2J_2''$$
$$\times[bF_2''-(1-b)F_2/2]P_2^{(2)}(\theta)+\lambda_4B_4U_4Q_4J_4''$$
$$\times[bF_4''+(1-b)F_4/12]P_4^{(2)}(\theta)\}. \quad (8)$$

This is the quantity determined because the two gammas cannot be resolved in the polarimeter. In this relation we have used the following additional quantities:

$$J_2''=0.993, \quad J_4''=0.980;$$
$$F_2''=(0.18708+0.31623\delta-0.09545\delta^2)/(1+\delta^2),$$
$$F_4''=-0.05878\delta^2/(1+\delta^2).$$

Actually, for $|\delta|$ near 0.16 the terms in $W(123)$ and $P(123+137)$ involving Q_4 are so small that this quantity remains undetermined.

Using our values of $W(123)$ and P from Sec. IA we obtain $\delta=+0.148\pm0.010$, $Q_2=0.81\pm0.10$, or $\delta=-12.1$, $Q_2=-2.94$; the latter are not compatible with predominantly $M1$ radiation. The value of δ disagrees with that of the Oxford group[8] (0.19 ± 0.02) but is in excellent agreement with a recent determination (0.15 ± 0.035) by de Waard and van der Woude.[26] The statistical range of Q_2 spans also the uncertainty in y. Q_2 is much closer

[24] L. S. Cheng, Rev. Mod. Phys. **28**, 45 (1956).
[25] J. G. Dash, R. D. Taylor, D. E. Nagle, P. P. Craig, and W. M. Visscher, Phys. Rev. **122**, 1116 (1961).

[26] H. de Waard and F. van der Woude, Phys. Rev. **129**, 1342 (1963); the apparent disagreement in sign of δ results from their use of a sign convention for the relative phase of $M1$ and $E2$ matrix elements opposite to that which we employ.

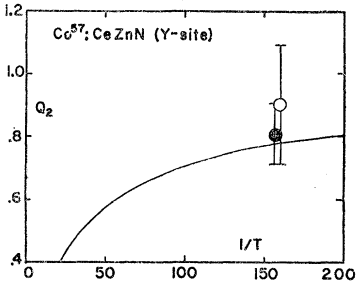

FIG. 5. Attenuation coefficient Q_2 due to intermediate state reorientation following decay of Co^{57} in Y sites of Ce-Zn nitrate at $1/T^*=157$, compared with a curve computed from Steenberg's theory of reorientation by static, isotropic hfs coupling in the hard-core limit. The open circle is Q_2 from the 137-keV data; the closed circle represents Q_2 from 123-keV results.

to unity for Ce-Zn nitrate at this temperature than in the case of the Tutton salt at a higher temperature ($Q_2=0.11$) [8]; at least part of this difference appears to be due to the discrepancy in δ values, however. Although the temperature dependence of Q_2 would also be of some interest for comparison with theory, we have not been able to get reliable data at higher temperatures because of the small heat capacity of Ce-Zn nitrate and the consequent faster warmups.

These values of Q_2 at $1/T=157$ for Co^{57} in the double nitrate Y site are compared in Fig. 5 with a curve computed from Steenberg's theory[12] of intermediate state reorientation by static hfs coupling with the assumption of isotropic ($A'=B'$) hyperfine interaction between the electron shell and the nucleus in the intermediate state (second excited state of Fe^{57}); the coupling is assumed sufficiently strong that for $\tau=12\times10^{-9}$ sec[18] the "hard-core limit" (precession angle $\alpha=\pi A'\tau/h\rightarrow\infty$) is approached. The contact hfs interaction between the nucleus and an s electron or hole would have such properties.

Equation (21) of Ref. 12 gives the intermediate substate populations to order $(1/kT)^2$ in this limit:

$$(N_m+N_{-m})/N=\tfrac{1}{3}\{1+(1/24)(12m^2-35)\times[1.607\eta^2-(\eta/6)(2.571+1.607\eta)+\cdots]\}, \quad (9)$$

with $m=\frac{1}{2}, \frac{3}{2}, \frac{5}{2}$ and $\eta=A(Y)/2kT=+0.0209/T$. Since in the intermediate state U_2B_2 is proportional to $(4/25)\sum m^2(N_m+N_{-m})/N-7/15$, we can formulate the expression $Q_2=(U_2B_2)^*/(U_2B_2)$, where the asterisk refers to the calculation including the correction term $(\eta/6)(2.571+1.607\eta)$.

The agreement shown in Fig. 5 is surprisingly good in view of the limitation of the theory to a "high"-temperature approximation which may not yield an accurate answer at $1/T=157$ (in this case our use of the *ratio* Q_2 may serve to extend somewhat the temperature range of the theory). If we were to accept this agreement, it would suggest that the mechanism described by the theory—static isotropic hfs coupling—may be responsible for the attenuation. Such a coupling could arise from $1s$, $2s$, or $3s$ holes formed during the transfer of the extra cobalt $3d$ electron to fill a hole in the iron K, L, or M shell following electron capture decay. In view of the short K-shell hole lifetimes estimated by Tolhoek *et al.*,[27] the effective holes would probably have to be in the outer shells. It does seem that this mechanism should be important in electron capture decays such as that of Co^{57}.

Nevertheless, it must be recognized that dynamic effects of nuclear recoil and/or electron shell de-excitation might be able to induce transitions between hyperfine levels during the intermediate-state lifetime. Such mechanisms, the quantitative effects of which have not yet been evaluated theoretically for oriented nuclei, might also be able to account for the observed attenuation. If these were operative on Co^{57} nuclei in X sites, they might destroy the isotropy of this contribution and shift the estimate of Q_2. In the absence of theories of attenuation by these mechanisms or of a more accurate treatment of the static hfs coupling effect at low temperatures, we are prevented from reaching a firm conclusion about the perturbing mechanism in this experiment.

B. Ce^{141} in Ce-Mg Nitrate

For the 145-keV ($M1+E2$) angular distribution,

$$W(145)=1+\lambda_2B_2U_2Q_2F_2'J_2P_2+\lambda_4B_4U_4Q_4F_4'J_4P_4, \quad (10)$$

we obtain B_k for $I=\frac{7}{2}$ as a function of $|B|/2kT=0.00908/T$ from the tables of Ref. 4. Here we have used the resonance values[28] of $|B|$; $|A|$ in Eq. (2) was too small to observe but is probably less than 10% of $|B|$.[28] Thus, neglect of A should not lead to errors larger than 1%. We note that B is proportional to the nuclear moment of Ce^{141} and hence is negative in this case, since $\mu(Ce^{141})$ is indicated to be negative from the sign of the 580-keV beta anisotropy.[29] The beta decay factor $U_2=0.970$ was obtained from the angular distribution of 440-keV beta particles,[29] the transition being classified as first-forbidden with $\Delta I=0$ and contributions principally of tensor rank zero. This value of U_2 implies $U_4=0.932$.

In this experiment $J_2=0.970$, $J_4=0.903$. The gamma transition factors are now

$$F_2'=(0.32733-1.8898\delta-0.07794\delta^2)/(1+\delta^2) \quad \text{and} \quad F_4'=0.63673\delta^2/(1+\delta^2).$$

Here again the smallness of δ precluded detection of the $k=4$ term.

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For the linear polarization we write

$$W(90)P(90)=-3\lambda_2 B_2 U_2 Q_2 F_2'' J_2'' - (15/2)\lambda_4 B_4 U_4 Q_4 F_4'' J_4'', \quad (11)$$

with $J_2''=0.983$, $J_4''=0.950$; $F_2''=(0.16366+0.31497\delta-0.03897\delta^2)/(1+\delta^2)$ and $F_4''=-0.05306\delta^2/(1+\delta^2)$.

Using the experimental data of Sec. IB and the numbers just quoted, we can solve simultaneously for the values of $\lambda_2 Q_2(1/T=150)$ and δ: $\lambda_2 Q_2=1.08\pm0.08$, $\delta=+0.085\pm0.013$; the possible solution $\delta=-49\pm5$, $\lambda_2 Q_2=-4.6\pm0.4$ is excluded by the mainly $M1$ character of the gamma rays.

For a precise value of the factor λ_2 we have used a theoretical approach. As the high temperature approximation[30,31] cannot yield a reasonable answer at $1/T=150$, we calculated the effect on Ce^{141} of a Zeeman term $g_1\beta HS_x$ added to Eq. (2), where H represents the local magnetic field due to the neighboring Ce ions. We have previously found this model useful for Co in Ce double nitrate.[13,16] In the present case it can only be considered an approximation to the actual distribution of local fields experienced by Ce ions in the paramagnetic state at any instant. The value $H=50$ Oe was taken to be typical of the rms field in the paramagnetic state, as reflected in the resonance line widths.[1,16]

The matrix of $(\frac{1}{2})g_1\beta H(S_+ + S_-)+(\frac{1}{2})B(S_+I_- + S_-I_+)$ on the basis $|S_z\rangle|I_z\rangle$ was diagonalized by an IBM-650 computer, yielding eigenvalues and eigenstates from which $(N_m+N_{-m})/N$ can be computed. Now $B_2(T)$ is proportional to $(4/49)\sum m^2(N_m+N_{-m})/N-3/7$, so we form the ratio $\lambda_2(T)=B_2(H,T)/B_2(0,T)$. In this discussion we assume a distribution in the directions of H in the plane perpendicular to the crystal axis[13,16] such that $\langle I_x\rangle=0=\langle I_y\rangle=\langle I_xI_y\rangle$, $\langle I_x^2\rangle=\langle I_y^2\rangle$, etc., so that $\langle I_z^2\rangle$, or B_2, fully specifies the (axial) orientation of the nuclear ensemble.[31,32]

The result is found to be, for $H=50$, that $\lambda_2=1.00\pm0.01$ in the entire temperature range $1/T=100$ to 300. That spin-spin interactions should be small in this case may be seen on the following physical grounds. Attenuation results from energy level shifts and state admixtures caused by H; these will be of second order in $g_1\beta H/B\sim1/3$, and thus of the order 10%. Furthermore, the lowest energy level spacing is increased by H so that the population of the ground state at temperature T is increased. This happens to cancel, approximately, in this temperature range the attenuation due to increased admixture of higher states into the ground state through the H term. In this connection note that the mixing of the nuclear substates is already relatively extreme because $B\gg A$.[10] Therefore, even though the local field model is not realistic in detail for Ce-Ce interactions, we expect it to be a sufficiently reliable index of the effectiveness of interactions. In this case, it suggests that the disturbance should not be more than 1 or 2% at most. Accordingly, we shall take $\lambda_2=1$ in our analysis, and thus $Q_2=1.08\pm0.08$. As for Co^{57}, we were unable to obtain values of Q_2 at higher temperatures.

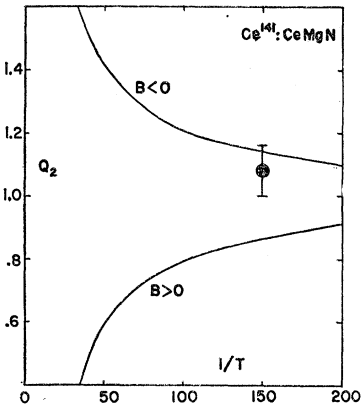

FIG. 6. Intermediate state reorientation coefficient Q_2 from decay of Ce^{141} in Ce-Mg nitrate at $1/T^*=150$, compared with curves calculated from Steenberg's theory of static, isotropic hfs reorientation in the hard core limit, for hfs constant $B>0$ (lower curve) and $B<0$ (upper curve).

In Fig. 6 we show the comparison of Q_2 with Steenberg's theory for static isotropic hfs coupling in the hard-core limit, as would be appropriate if, for example, the electron configuration changed to $4f^25s^15p^6$. The computation proceeds as for Co^{57}, except that now

$$(N_m+N_{-m})/N=\tfrac{1}{4}\{1-\tfrac{1}{8}(m^2-63/12)[\zeta^2-\tfrac{3}{8}\zeta+\cdots]\},$$

with $m=\frac{1}{2}, \frac{3}{2}, \frac{5}{2}, \frac{7}{2}$ and $\zeta=B/kT=-0.0182/T$. $Q_2(T)$ is calculated as before. Disturbance appears to be negligible but, due to the limited precision of the experiment, we cannot exclude a small enhancement at $1/T=150$ in accord with the Steenberg theory. Note that $\tau=3.9\times10^{-9}$ sec here,[33] about one third of the lifetime of the Fe^{57} state discussed above. Also as in the case of Co^{57}, the accuracy of the Steenberg theory is questionable at this temperature, and dynamic perturbations such as recoil and electron shell excitation should also be considered.

Our data on Ce^{141} presented above differ markedly from those reported earlier by Ambler, Hudson, and Temmer,[9] whose results had indicated serious attenuation of the alignment. In our study the following experimental improvements were introduced: (i) one single crystal of spherical shape, instead of several natural crystals, minimized the $T-T^*$ difference and the possibility of crystal misorientation; (ii) many demagnetizations from different initial conditions and extrapolation of bridge and counter readings back to the time of demagnetization eliminated the influence of inhomogeneous temperature distribution in the sample; (iii) study of the full radiation pattern prompted us to focus our attention on an intermediate temperature where the angular distribution offered the most straightforward interpretation; (iv) the gamma linear polarization measurement on the same sample determined the $E2/M1$ mixing ratio independently; (v) experimental precautions minimized scattering from the tail of the cryostat which can seriously attenuate the gamma

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anisotropy. On the theoretical side we had the benefit of additional information on the spin and hfs coupling of Ce^{141} and on the inner beta transition which has recently become available.

Our conclusion that disturbance of this alignment of Ce^{141} in Ce-Mg nitrate is small, essentially zero, is in agreement with a recent report of the Oxford group.[34] To their comments concerning the probable influence of temperature inhomogeneity on Ambler, Hudson, and Temmer's data, we should like to add the following: since even the lowest temperature point ($\epsilon=12\%$) in Ref. 9 lies considerably under our data ($\epsilon=14\%$) at $1/T^*=290$, it seems possible that scattering played a role in attenuating Ambler, Hudson, and Temmer's results. Also, the complete radiation pattern at the lowest temperature, where they did their study, shows a departure from explainable behavior, possibly due to onset of magnetic ordering.[13]

Finally, it is of interest to compare Ce^{141} alignment in double nitrates with that in ethyl sulfates.[34–37] Since g_1 is small in the latter crystal, Ce^{141} is relatively insensitive to spin-spin interactions ($\lambda_2=1$). The experimental results have customarily been fitted assuming $Q_2=1$. However, in principle Ce^{141} could be susceptible to intermediate state reorientation in this salt by, e.g., the Steenberg mechanism,[12] as was Co^{57} in the Y site of Ce-Zn nitrate. It is important to note that, because the hfs constants for Ce^{141} in Nd ethyl sulfate have not been directly measured by paramagnetic resonance, A and B as well as δ (and Q_2) have to be determined from the alignment data. As the purely nuclear parameter δ should be the same in all experiments on Ce^{141}, the agreement among experiments with different salts is satisfying (see Table I below).

TABLE I. Summary of experimental values for mixing ratio δ and intermediate state reorientation coefficient Q_2 from nuclear alignment experiments in double nitrate (D.N.), ethyl sulfate (E.S.), and Tutton salt (T.S.) crystals.

Nuclide	Crystal	$\delta=\pm(E2/M1)^{1/2}$	$Q_2(1/T^*)$	Ref.
Co^{57}	Ce-Zn D.N.	$+0.148\pm0.010$	0.81 ± 0.10 (157)	[a]
(123 keV)	Cu-Rb T.S.	$+0.19\ \pm0.02$	0.11 (80)	8
Ce^{141}	Ce-Mg D.N.	$+0.085\pm0.013$	1.08 ± 0.08 (150)	[a]
(145 keV)	Nd E.S.	$+0.08\ \pm0.02$	(1)[b]	34,35
	Ce-Mg D.N.	$(+0.08\ \pm0.02)$[b]	(1)[b]	34
	Nd, Ce E.S.	$+0.068\pm0.008$	(1)[b]	36
	Nd E.S.	$+0.066\pm0.022$	(1)[b]	37

[a] This work.
[b] Assumed value.

III. SUMMARY

We have investigated two instances in which nuclear alignment in cerium double nitrate might be expected to be disturbed by intermediate state reorientation. In Co^{57} evidence for a $(19\pm10)\%$ attenuation at $1/T=157$ was presented; a possible enhancement of no more than $(8\pm8)\%$ was found for Ce^{141}. These results are displayed, along with values from other nuclear alignment experiments, in Table I.

A theory of reorientation by static, isotropic hfs coupling, due to Steenberg, agrees reasonably well with the experimental results, surprisingly so in view of the high temperature approximation of the calculation. Although this mechanism is a most reasonable one, it would be desirable to have theoretical treatments of nuclear recoil and electron shell de-excitation also available for comparison before attempting to identify the dominant mechanism or mechanisms in the case of Co^{57}.

Note added in proof. The high temperature approximation can be eliminated by using Eqs. (16) and (17) of Ref. 12, instead of Eq. (21). For Co^{57} at $\eta=3.28$, $\alpha=0.10$, we find $Q_2=0.82$, $Q_4=0.40$; both are in excellent agreement with experiment. For Ce^{141} at $\zeta=-1.36$, $\alpha=\infty$, $Q_2=+0.94$; if a small enhancement is occurring, it cannot be due to static hfs coupling.

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