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INTERMEDIATE STATE REORIENTATION OF COBALT-57 NUCLEI ALIGNED IN CERIUM ZINC NITRATE

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INTERMEDIATE STATE REORIENTATION OF COBALT-57 NUCLEI ALIGNED IN CERIUM ZINC NITRATE*

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Radiation pattern and linear polarization of gamma rays from decay of Co^{57} aligned at low temperatures by Bleaney's method in the Y-sites of cerium zinc 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 $\delta = \pm\sqrt{(E2/M1)}$ for the 123 kev M1 + E2 gamma transition and of an attenuation coefficient Q_k in the angular distribution $W(\theta) = \sum_{k\ even} Q_k A_k P_k(\cos\theta)$, are $\delta = +0.148 \pm 0.010$, $Q_2 = 0.81 \pm 0.10$ at $1/T^* = 157$. 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 another nuclear alignment experiment involving the same isotope.

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 simple behavior, thereby complicating severely the interpretation of the data and the extraction of precise nuclear information.[4] These non-ideal 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.

The influence of reorientation arises as follows: when oriented radioactive nuclei decay by a cascade of nuclear transitions, alteration of the radiation pattern anisotropy of one of the transitions may arise if the initial state of that, or any prior, transition is relatively long-lived. For example, if reorientation is due to hfs coupling of the intermediate state nucleus to its environment with strength ΔE of the order 10^{-17} erg, then the alteration may be appreciable if the mean lifetime of the intermediate state is of the order $(h/\Delta E)$, or about 10^{-9} sec. Such effects, of course, have also been discussed[5-7] for certain gamma-gamma angular correlations.

For the purpose of obtaining a quantitative estimate of the intermediate state effect in a particular case, we have studied alignment, in the cerium double nitrate latice, of Co^{57} nuclei, for which an indication of non-

ideal behavior already existed.[8] Our choice was also guided by the availability of sufficient auxiliary information on the nuclear decay scheme, spins, hfs couplings, etc., for this isotope which would enable us to isolate the departure from ideal behavior with a precision limited mainly by the counting statistics.

A certain amount of interest attaches to the particular case studied. Co^{57} decays by electron capture and undergoes axial alignment[9] in the Y-sites[10] of Ce double nitrate, for which the hfs coupling is extremely anisotropic (A>>B). The decay leads to the second excited state of Fe^{57}, which has a lifetime of the order of tens of nanoseconds preceding a mixed M1+E2 gamma transition, thus providing the possibility of intermediate state perturbation by hyperfine fields.

We shall show that the influence of intermediate state reorientation in this case is appreciable. In isolating this effect we have used quantitative theoretical corrections for spin-spin interactions. Though the experimental data is of only moderate precision, its agreement with the available theory of perturbation by static isotropic hfs coupling[11] is surprisingly good. These results should be of considerable utility in designing and interpreting future nuclear orientation experiments with cerium double nitrate, and, its is hoped, in stimulating further theoretical studies of intermediate state reorientation of aligned nuclei.

I. EXPERIMENTS

Our experimental apparatus and methods are quite standard[4], and have been described at length in an earlier publication.[12] 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 paragraphs we shall describe some additional details pertinent to the particular problem studied.

Approximately 20 microcuries of Co^{57} activity[13] produced by proton bombardment of nickel 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 Osborn's demagnetization factors[14] we computed the shape-corrected magnetic temperatures $T^* = T^*(obs.)+0.29\times10^{-3}$°K, which should be practically equal to T above 6 mdeg K.[2]

As no magnetic field was applied following adiabatic demagnetization, axial alignment at very low temperatures by the Bleaney method[9] was produced for Co^{57} in the Y-sites of Ce-Zn nitrate[15]. The X-sites in this crystal, being essentially isotropic[15], 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[15], the value which we shall adopt in this paper.

Radiation patterns $W(\theta)$ were measured, after demagnetizations from 7.0 and 25 koer to final temperatures of $1/T^* = 157$ and 276, respectively, with single channel analyzers spanning the 123 kev photopeak.[4] Our one-by-one inch cylindrical 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.[16]

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 apparent that 137 kev E2 radiation, which accounts for 10%[17] 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 isotropy. In fact, small anisotropies of the order of a few per cent, 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 was expected to predominate. Accordingly, we undertook multichannel analysis with an RCL 256 channel analyzer (kindly made available by Prof. R. W. Krone) of the line shape in order to isolate contributions of the two gamma rays.

Our procedure is illustrated by Fig. 1. Two five-channel portions of the spectrum, chosen to detect principally 123 or 137 kev radiation, respectively, were studied for anisotropy as the sample warmed up. The expected 137 kev

line at 1°K was constructed by scaling down 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 $1/T^* = 157 \pm 8$ 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 straight forward just above the magnetic transition temperature of Ce-Zn nitrate.[2,12] Least squares fits to the patterns with Legendre polynomial expansions yield the following results at this temperature:

$$W(123) = 1 + (0.0324 \pm 0.0038)P_2$$

$$W(137) = 1 - (0.216 \pm 0.045)P_2 - (0.023 \pm 0.052)P_4.$$

A Compton polarimeter of the type first described by Metzger and Deutsch[18] was utilized for determining the degree of linear polarization of the gamma rays, and hence the mixing ratio $\delta = \pm(E2/M1)$ of the 123 kev transition. The magnitude and sign of δ have a strong influence on estimating the reorientation effect from data on the 123 kev gamma ray. As operation of this type of polarimater has often been described in the literature, we shall mention only a few salient peculiarities of our experimental arrangements.

The chief point of interest was our use of a Na I(Tl) crystal, one-half inch in diameter by one inch long, as a scatterer[19], instead of an organic material. 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, the coincidence counting rate being only slightly lower than that for a one by one inch Pilot B scatterer we had previously employed, in spite of the increased absorption of scattered gamma rays due to the high photoelectric cross-section of NaI. The coincidence spectrum of Compton electrons in the scatterer showed a single hump at the energy corresponding to scattering into the side counter, with little of the lower energy background often seen in Compton spectrometers[20]. 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 the same temperature as were the patterns in Fig. 2. The same side counter was placed alternately in ($N_{//}$), 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 reference 4. This type of 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 influence of temperature inhomogeneity on the coincidence rates. The results are

$$N_{//}/N_{\perp} = 0.693 \pm 0.090.$$

The statistical error quoted also spans the uncertainty in normalization and extrapolation procedures.

Finally, we obtain P from the relation

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

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 $\bar{\beta}$ and azimuthal angle $\bar{\psi}$ determined by the 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 we estimate to be $\beta = 80° \pm (20° \pm 5°)$, $\psi = 0°, 90° \pm (25° \pm 5°)$, we obtain $\bar{r} = 0.76 \pm 0.04$. Therefore, $P = -0.240 \pm 0.040$, with the quoted uncertainty determined by counting statistics; the uncertainty in $\bar{r}$ has not been folded in, but lies within this range.

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

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

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

$$\mathcal{H} = A I_z S_z + B(I_x S_x + I_y S_y). \quad (2)$$

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

The factor U_k represent the effect of unobserved preceding transitions—in the present cases, beta transitions—and are simply proportional to Racah coefficients. F_k' and F_k'' are factors dependent on the parameters of the gamma transition, and particularly on the degree of coherent mixing $\delta = \pm(E2/M1)^{1/2}$. They are easily evaluated using the tables of Ferentz and Rosenzweig.[21]

In equation (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[22] are useful in estimating these corrections.

Spin-spin interactions are introduced 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 does not destroy the axial symmetry of the radiation pattern. (A separate investigation of the cylindrical symmetry of the radiation pattern substantiated this assumption.)

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 that cylindrical symmetry of the radiation pattern is not altered.

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

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

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

We now proceed to the results and their interpretation.

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

$$W(137) = y\,W(Y,137)+(1-y)W(X,137)$$

$$= 1 + y\lambda_2B_2U_2Q_2F_2J_2P_2 + y\lambda_4B_4U_4Q_4F_4J_4P_4$$

where y = fraction of Co^{57} in Y-sites = 0.357 ± 0.025; B_k are the orientation parameters[4] for Co^{57} in the Y-sites at 1/T = 157, using I = 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 [23,24] 7/2(1)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, which contains the presumption that neither spin-spin interactions nor intermediate state reorientation disturbs 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[12]. A somewhat tenuous justification of the second presumption must be made <u>a posteriori</u> from Steenberg's theory[11] of reorientation, which predicts no effect from the mechanism he treated (static isotropic hfs coupling) if both the initial and intermediate states of the X-ion are indeed spherically symmetric. Finally, we shall set $\lambda_2(Y)$ and $\lambda_4(Y)$ equal to unity, since A(Y)>>B(Y) makes this ion rather insensitive to interactions.[12]

Comparing thetheoretical pattern at 1/T = 157,

$$W(137) = 1 - 0.239\ Q_2P_2 - 0.0848\ Q_4P_4$$

with the observed pattern quoted in Section I, we obtain Q_2 = 0.90 ± 0.19, Q_4 = 0.27 ± 0.60. The relatively poor precision with which the coefficients of P_2 and P_4 were

measured resulted from the low intensity of 137 kev rays and the large 123 kev correction.

For the 123 kev gamma,

$$W(123) = y\,W(Y,123) + (1-y)W(X,123)$$

$$= 1 + y\lambda_2 B_2 U_2 Q_2 F_2' J_2 P_2 + y\lambda_4 B_4 U_4 Q_4 F_4' J_4 P_4,$$

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 123 kev pattern, for which the P_2 coefficient vanishes if $\delta = +0.19$. Next, consider the combined pattern:

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

Here the intensity branching fraction of 123 kev radiation $b = 0.90 \pm 0.02$[17]. 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. Therefore, we use the polarimeter data to determine δ.

The total linear polarization at angle θ and temperature T is

$$P(123 + 137) = -y\{\lambda_2 B_2 U_2 Q_2 J_2'' [bF_2'' - (1-b)F_2/2] P_2^{(2)} + \lambda_4 B_4 U_4 Q_4 J_4'' [bF_4'' + (1-b)F_4/12] P_4^{(2)}\}.$$

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: $J_2'' = 0.993$, $J_2'' = 0.980$; $F_2'' = (0.18708 + 0.31623\delta - 0.09545\delta^2)/(1 + \delta^2)$, $F'' = -0.05878\delta^2/(1 + \delta^2)$. Actually, for $/\delta/$ near +0.16

the terms in W(123) and P(123 + 137) involving Q_4 are quite small, so that this quantity remains undetermined.

Using our values of W(123) and P from Section I we obtain $\delta = +0.148 \pm 0.010$, $Q_2 = 0.81 \pm 0.10$. The value of δ disagrees with that of the Oxford group (0.19)[8], but is in excellent agreement with a recent determination by de Waard and van der Woude[25]. This more precise Q_2 value is in satisfactory agreement with Q_2 from W(137). It is much closer to unity than in the case of the Tutton salt at a higher temperature (Q_2=0.11)[8]. Although the temperature dependence of Q_2 would also be of some interest for comparison with theory, we have not been able to obtain 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. 4 with a curve computed from Steenberg's theory[11] 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}) so strong that for $\tau = 12 \times 10^{-9}$ sec[17] the "hard-core limit" (procession angle $\alpha = \pi A'\tau/h \to \infty$) is reached. The contact hfs interaction of an s-electron or hole would have such properties.

Eq. (21) of reference 11 gives the intermediate substate populations to order $(1/kT)^2$ in this limit:

$$(N_m + N_{-m})/N = (1/3)\left\{1 + (1/24)(12m^2 - 35)\left[1.607\eta^2 - (\eta/6)(2.571 + 1.607\eta) + \ldots\right]\right\}$$

with $\eta = A(Y)/2kT = 0.0209/T$. 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. 4 is surprisingly good in view of the limitation of the theory to "high" temperature approximation which may not yield an accurate answer at $1/T = 157$ (in this regard, our use of the ratio Q_2 may be serving to extend the temperature range of the theory somewhat). This agreement 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 shells following electron capture decay. In view of the short K-shell hole lifetimes estimated by Tolhoek et al.[26], the effective holes would probably have to be in the outer shells. It seems reasonable that this mechanism ought to be one of the most important in electron capture decays such as Co^{57}.

However, it must be recognized that dynamic effects of recoil and/or electron shell de-excitation might be able to induce transitions between hyperfine levels during the intermediate state lifetime. Such mechanisms, the effects of which have not yet been evaluated theoretically for oriented nuclei, might be shown also to be able to account for the observed attenuation. Thus, we cannot yet reach a conclusion about the mechanism which is causing the attenuation.

III. SUMMARY

We have investigated an instance in which nuclear alignment of Co^{57} in cerium zinc nitrate might be disturbed by intermediate state reorientation, and evidence for a $(19 \pm 10)\%$ attenuation at $1/T = 157$ was presented. These results should be compared with those for the Tutton salt[8], which are $\delta = +0.19$ and an 89% attenuation at $1/T = 80$.

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 available also for comparison before drawing any firm conclusions as to the dominant mechanisms.

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FOOTNOTES

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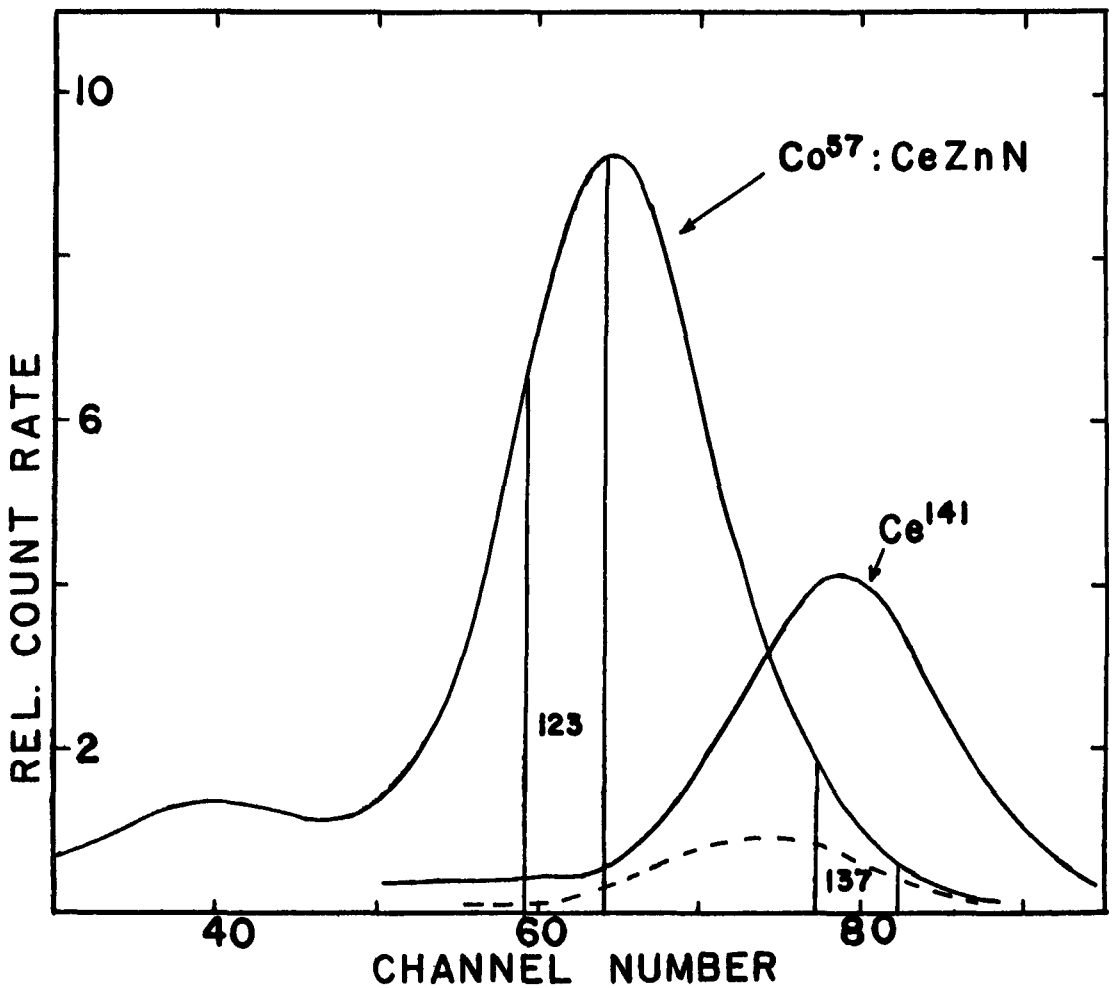
FIGURE CAPTIONS

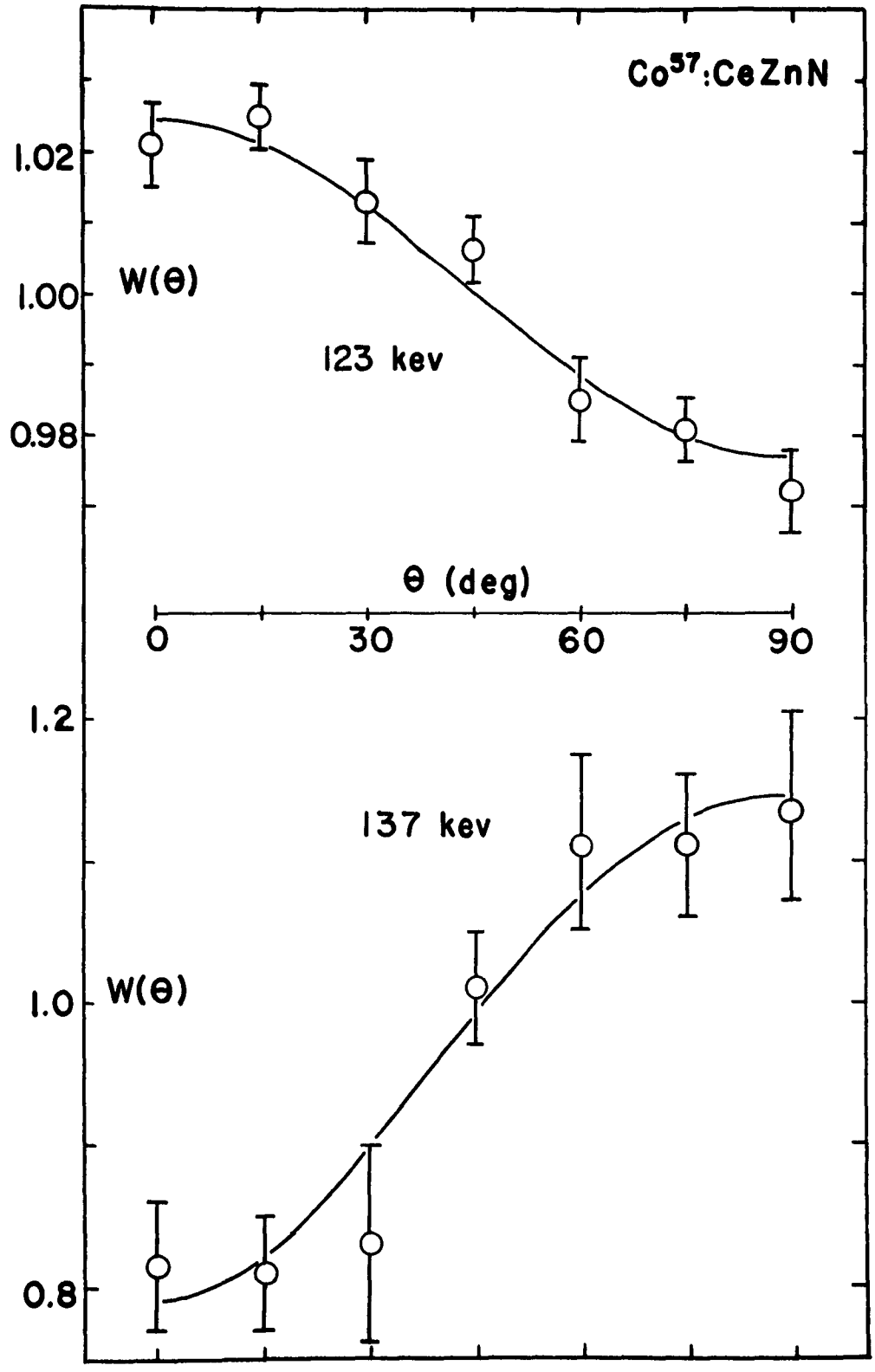
Fig. 1. Multi-channel analysis of the gamma spectrum from the decay of Co^{57} in Ce-Zn nitrate at 1°K. The expected contribution of the 10% 137 kev gamma ray was constructed from the Ce^{141} 145 kev spectrum in the same counter. Window settings used to study the 123 and 137 kev angular distributions are indicated.

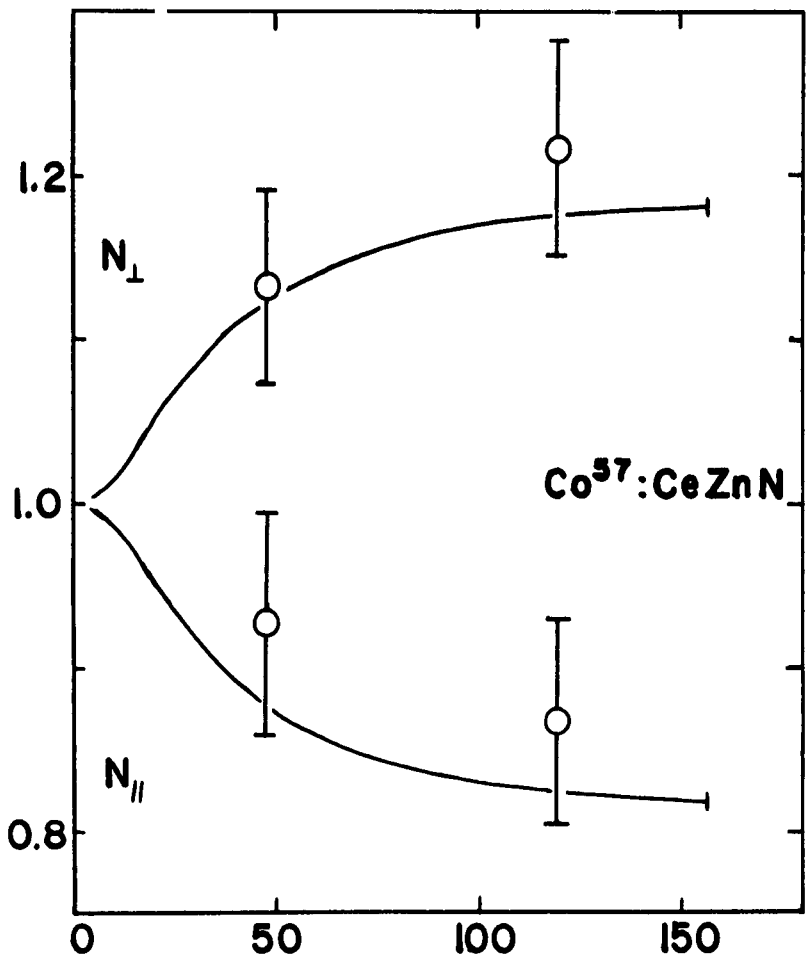
Fig. 2. Radiation patterns $W(\theta)$ of the 123 kev (upper curve) and 137 kev (lower curve) gamma transitions following decay of Co^{57} in Ce-Zn nitrate at $1/T^* = 157$. The solid lines are the least-squares fits to Legendre polynomial expansions as quoted in the text.

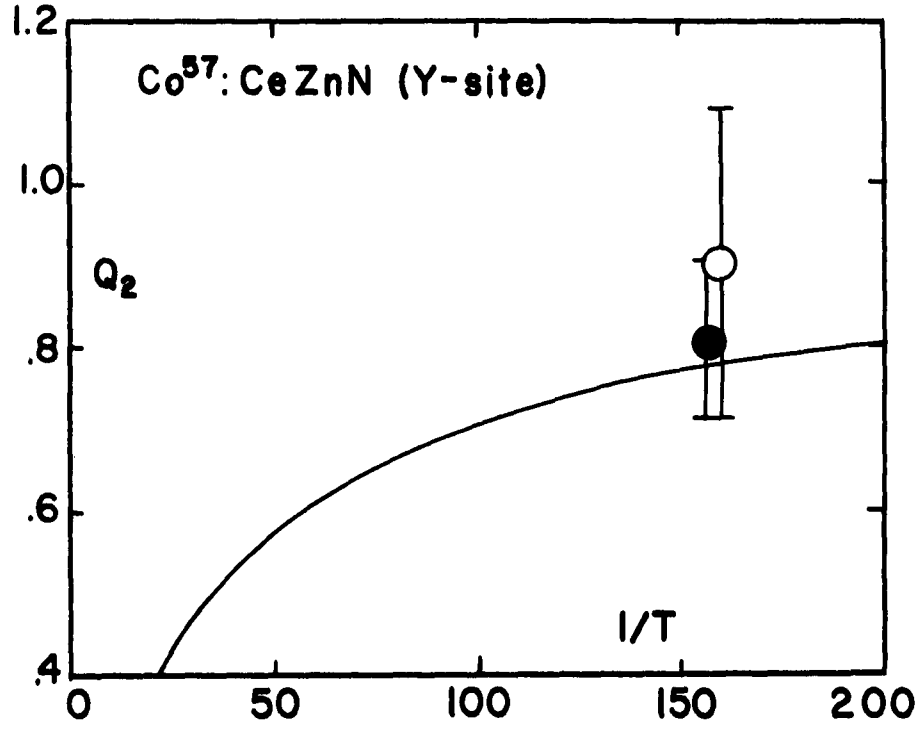
Fig. 3. Normalized coincidence rates N from Compton polarimeter at $\theta = 75°$ for 123 + 137 kev gamma radiation from Co^{57} in Ce-Zn nitrate following magnetic cooling to $1/T^* = 157$ and subsequent warmup. $N_{\perp}$ corresponds to side counter perpendicular to plane of gamma ray direction and alignment axis; $N_{//}$ 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 afterccooling.

Fig. 4. Attenuation coefficient Q_2 due to intermediate state reorientation following decay of Co^{57} in Y-sites of Ce-Zn nitrate at $1/T^* = 157$. The curve is computed from theory of reorientation by static, isotropic hfs coupling in the hard core limit.


10
6
2
REL. COUNT RATE
Co57 : CeZnN
Ce141
123
137
40
60
80
CHANNEL NUMBER


Co57:CeZnN
1.02
1.00
0.98
W(θ)
123 kev
θ (deg)
0
30
60
90
1.2
1.0
0.8
W(θ)
137 kev


1.2
1.0
0.8
$N_\perp$
$N_{//}$
Co^{57}:CeZnN
0
50
100
150


Co57: CeZnN (Y-site)
1.2
1.0
.8
.6
.4
Q2
1/T
0
50
100
150
200