

The Gamma-Ray Distribution From Oriented Cerium-141

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The distribution of gamma-radiation emitted in the decay of Ce^{141} nuclei aligned in neodymium ethylsulfate single crystals has been remeasured for temperatures as low as 0.0162 °K and has been found to be considerably more anisotropic than previously thought. The discrepancy is explained on the basis of preferential scattering of the plane-polarized radiation in the Dewar system. Similar measurements with Ce^{141} nuclei oriented in a polarized cerium ethylsulfate crystal have been made. The hyperfine constant for Ce^{141} in the neodymium ethylsulfate lattice is deduced as $A = 0.0305 \pm 0.003 \text{ cm}^{-1}$, and the $E2/M1$ ratio for the 145-keV gamma ray in Pr^{141} is $+0.068 \pm 0.008$.

1. Introduction

In order to use the gamma-ray distribution $[1]^2 W(\theta) = 1 + \sum A_i f_i P_i(\cos \theta)$ from aligned or polarized Ce^{141} as a secondary thermometer in experiments involving beta-ray distributions [2] or heat conduction in rare earth ethylsulfate crystals, we have re-examined this distribution experimentally. We find a considerably greater anisotropy than that reported by previous investigators [3, 4], and we find that scattering may offer an explanation for the discrepancy. Moreover, we are able to make an experimental determination of the hyperfine constants for cerium isotopes in concentrated neodymium ethylsulfate. Experiments with Ce^{141} in concentrated cerium ethylsulfate show that this system can produce almost total alignment when a small magnetic field is applied.

2. Experimental Procedure

2.1. General

The Ce^{141} used in this investigation was fission-product material which contained Ce^{144} and Pr^{144} as impurities. Conversion electron spectra observed with a thick-lens beta-ray spectrometer were used, together with data on the conversion coefficients [5], to find the contribution of the 134-keV Ce^{144} gamma-ray to the experimental spectrum of the 145-keV gamma-ray of Ce^{141} .

Approximately 1 microcurie of the activity was grown into each of four single crystals, each weighing 2 to 3 g. Two crystals contained only neodymium ethylsulfate and the activity. In another, 1 percent of the neodymium was replaced by stable cerium. The fourth crystal was concentrated cerium ethylsulfate. An inactive layer was grown around each crystal.

Although the experimental procedure used with the cerium ethylsulfate crystal was somewhat different from that used with the other specimens, the

gamma-ray detectors and the counting procedures were the same in each case. Three gamma-ray detectors, usually utilizing 2 in. long by 2 in. diam NaI cylinders, were mounted in a horizontal array into which the Dewar assembly was swung after adiabatic demagnetization of the specimen. Each detector was monitored by a single-channel pulse height analyzer, whose output was switched at 1 min intervals from one to another of a pair of scalers. The output of one detector was also recorded by a 100-channel pulse height analyzer during the orientation period. After a counting time of from 3 to 10 min, the sample was warmed to the helium bath temperature in order to take a similar sampling of the isotropic radiation for normalization. The ratio of the "cold" to "warm" counting rates was then corrected for background, attenuation due to the finite solid angle subtended by the detector, and the gamma ray impurity.

2.2. Neodymium Ethylsulfate Host Crystals

The cryogenic apparatus used for the three neodymium ethylsulfate crystals has been described in detail by de Klerk and Hudson [6]; a useful modification has been the use of He^3 exchange gas, i.e. gas introduced to provide thermal contact between the sample and the pumped He^4 bath during the magnetization portion of the cycle. Pumping off the He^3 gas produced thermal isolation for demagnetization in approximately 3 min, an order of magnitude improvement over the use of He^4 . Each crystal was mounted with the c -axis horizontal in order that the horizontal 22-kilogauss field could be applied for maximum cooling effect. The data of Meyer [7] were used to deduce the temperatures reached for the various values of magnetic field and bath temperature; in these runs, the final temperature varied between 0.1 °K and 0.016 °K. In general, two of the gamma-ray detectors were placed to receive radiation emitted parallel to the c -axis, and the third sampled radiation emitted perpendicular to this alignment axis. Warming of the specimen could be detected by change in the gamma-ray counting rate and by change in the paramagnetic susceptibility; there was, however, no systematic variation in the gamma-ray data during any counting period. Furthermore, the initial rate

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² Figures in brackets indicate the literature references at the end of this paper.

of warming from the lowest temperatures was difficult to assess by the mutual inductance bridge, since the perpendicular susceptibility appeared to change very little below 0.020 °K. For higher initial temperatures, the change in temperature during the "cold" counting period varied from about 5 percent at 0.026 °K to about 10 percent at 0.100 °K.

2.3. Cerium Ethylsulfate Host Crystals

The experimental arrangement in this case was dictated by the observation of Johnson and Meyer [8], indicative of an antiferromagnetic transition, that the adiabatic demagnetization process results in a minimum temperature of a cerium ethylsulfate specimen when the high initial field is reduced to 500 to 600 gauss, rather than to zero field. For an initial field of 22 kilogauss applied along the crystalline *c*-axis and a parallel residual field of 550 gauss, the final specimen temperature is calculated to be within the range 0.003 to 0.010 °K. In order to achieve optimum gamma-ray counting conditions in the present experiment, this residual field was supplied by a superconducting solenoid immersed in the helium bath. This coil consisted of 1,850 turns of 0.003 in. diam formvar-coated niobium wire, wound in two layers on the glass wall of the inner space of a light-guide cryostat designed for beta-ray distribution studies [9]. Since this solenoid produced a vertical field of no more than 1,700 gauss, it was necessary to use the large horizontal-field magnet for the initial field. In order to apply each field, in turn, along the crystalline *c*-axis, advantage was taken of the highly anisotropic *g*-values of cerium ethylsulfate [10]. The sample was suspended by a nylon thread passing horizontally through the crystal slightly above its center of gravity so that with zero applied field the *c*-axis was maintained vertical. Application of the 22-kilogauss field rotated the sample until the *c*-axis was horizontal. Upon demagnetization the crystal returned to the zero-field position, and it remained there when the solenoid was energized. The assembly was X-rayed under each of the above conditions to verify that the correct position was achieved.

As before, the gamma-ray counters were placed in a horizontal plane, which in this case corresponded to $\theta=90^\circ$ for each. Contrary to the observations with the neodymium ethylsulfate samples, the gamma-ray counting rate changed noticeably during succeeding 1-min periods, necessitating an extrapolation of the "cold" counting rate to the time of demagnetization, approximately 1 min before the start of counting.

3. Results

The angular dependence of the gamma-ray intensity from cerium-141 aligned in neodymium ethylsulfate was measured at approximately 0.0162 °K and found to be satisfactorily represented by the function

$$W(\theta) = 1 + (0.205 \pm 0.010)P_2(\cos \theta) + (0.006 \pm 0.018)P_4(\cos \theta)$$

where θ denotes the angle between the axis of alignment (in this case the crystalline *c*-axis) and the direction of gamma-ray emission. The coefficient of $P_4(\cos \theta)$ relative to that of $P_2(\cos \theta)$ can be calculated approximately, given an estimate of f_2 ; this ratio, A_4f_4/A_2f_2 , is calculated to be 0.024.

The temperature dependence of $W(0^\circ)$ for the neodymium ethylsulfate crystals is shown in figure 1. The data taken with the 1 percent cerium crystal could not be distinguished from that taken with the two essentially pure neodymium ethylsulfate crystals, and therefore it is also included in figure 1. The error flags denote the standard deviation of the mean of a number of 5 to 10 min counts at a given temperature. Overall, some one hundred such counts were made on the three samples. Corrections for detector solid angle, background, and cerium-144 impurity amounted to an average of 10 percent of A_2f_2 . The temperature scale was derived from the data of H. Meyer [7] for a magnetic field applied parallel to the crystalline *c*-axis.

The data of Cacho et al., [3] were taken with low specific activity cerium-141, so that the sample corresponded almost exactly with the present 1 percent cerium crystal. Since A_2f_2 was plotted by them as a function of $1/T^*$, approximate corrections for the nonspherical shape of the specimen and for Meyer's $T-T^*$ (sphere) relation have been made. The adjusted data, shown in figure 1, are in agreement with similar corrections made by Scurlock [11]. The triangular point in figure 1 represents the value of A_2 calculated from the experiments of Ambler, Hudson, and Temmer [4], using planar alignment of cerium-141 in cerium magnesium nitrate, multiplied by the f_2 deduced from the present experiments for the temperature 0.0166 °K.

An interesting feature of the gamma-ray distribution was the change in $W(\theta)$ with decreasing energy of the detected photon. The spectrum measured at 0.016 °K and an isotropic comparison spectrum taken 1 °K are shown in figure 2. $W(0^\circ)$ was 1.205 ± 0.010 for photon energies between approximately 140 and 170 keV; between 140 and 125 keV, however, the intensity decreased uniformly until at 115 keV it reached a minimum value of 0.80 ± 0.04 . Correspondingly, $W(90^\circ)$ was 0.90 ± 0.01 at the photopeak energy, rising to a maximum of 1.10 ± 0.05 at approximately 115 keV. This reversal in the anisotropy at lower energies was demonstrated to be a scattering phenomenon by the use of a collimating system consisting of a 1 cm hole in a gold foil surrounding the vacuum jacket and tin-lined lead cones which were placed in front of the detector crystals. Multichannel analysis of the photon spectrum under these conditions showed no reversal of the anisotropy around 115 keV. It should be pointed out that most of the data shown in figure 1 were taken with single-channel analyzers and that, to avoid the attenuation due to this scattering effect, the discriminator thresholds were set at about 135 keV. This selection made the counting rate sensitive to small shifts in the pulse height, as is evidenced by the relatively large standard deviations shown on the data of figure 1.

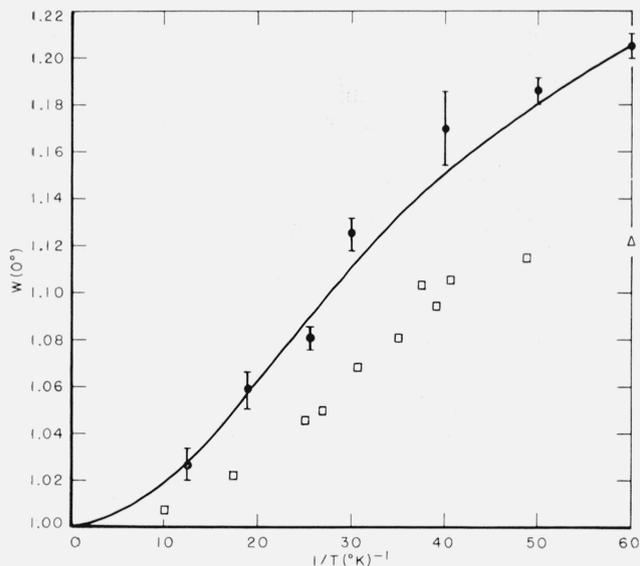


FIGURE 1. The temperature dependence of $W(0^\circ)$ (which equals the quantity $(1+A_2f_2)$ in this case) for Ce^{141} in neodymium ethylsulfate.

●, present data;
 □, Cacho et al., [3];
 △, calculated from data of Ambler, Hudson, and Temmer [4];
 —, theoretical curve (see discussion sec. 4).

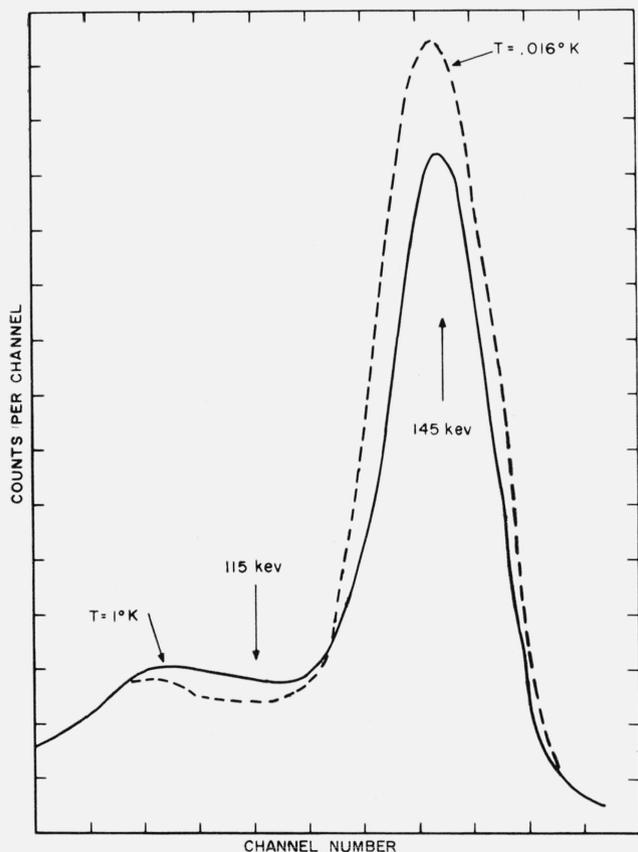


FIGURE 2. The energy spectrum of gamma-radiation from Ce^{141} in neodymium ethylsulfate observed at $\theta=0^\circ$ without collimation.

—, isotropic spectrum taken at $T=1^\circ K$;
 - - -, spectrum taken at $T=0.016^\circ K$.

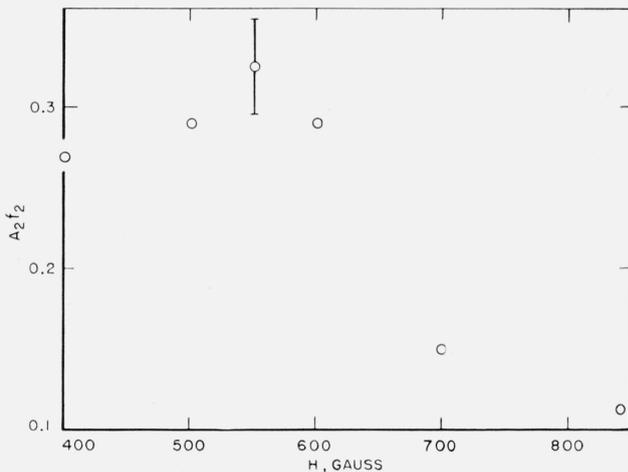


FIGURE 3. The coefficient of $P_2(\cos \theta)$ for gamma-radiation from Ce^{141} oriented in cerium ethylsulfate, as a function of magnetic field applied parallel to the crystalline c-axis.

The results of the cerium ethylsulfate experiments are illustrated in figure 3, in which the experimental A_2f_2 is plotted against applied field. The maximum value of A_2f_2 is seen to be about 0.325 ± 0.03 . The corresponding magnitude of f_2 can be estimated, assuming the hyperfine interaction to be similar to that deduced for neodymium ethylsulfate as the host lattice, to be nearly 0.571, the value characteristic of complete alinement. The fact that $f_2 \leq 0.571$ allows a lower limit of 0.508 to be placed on A_2 . This limit A_2 was used in the analysis of the neodymium ethylsulfate data that follows.

4. Discussion

The change in sign of the experimental anisotropy at energies below about 130 keV can be accounted for by noting that the gamma-rays emitted from the aligned cerium-141 are plane-polarized, and considering the effect of this polarization on subsequent scattering of the photons from the vacuum jacket and the Dewar walls (see fig. 4). With a typical (uncollimated) counting geometry the radiation de-

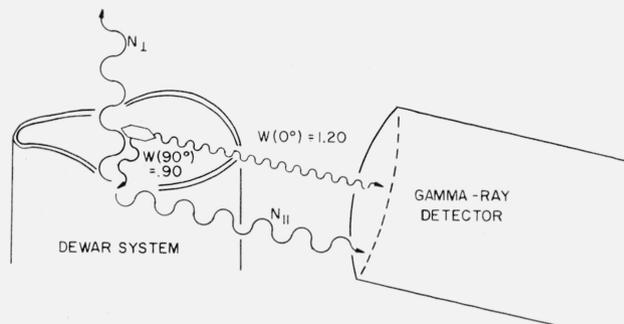


FIGURE 4. Schematic diagram of source, Dewar system, and detector placed at $\theta=0^\circ$.

[Radiation emitted in the $\theta=90^\circ$ direction and scattered by the Dewar system through an angle of 90° can impinge on the uncollimated detector. Indicated values of $W(0^\circ)$ and $W(90^\circ)$ are those observed at $T=0.016^\circ K$ for the collimated beams.]

defined as N_{\parallel} [12], taking the scattering medium as the Dewar system, could impinge directly upon the detector placed along the axis of alignment. A photon beam of 145 keV must be scattered through 90° in order to be degraded to an average energy of 115 keV according to the simple Klein-Nishina relation [13]; thus, without considering polarization of the gamma-rays, the ratio of "cold" to "warm" counting rates measured at 115 keV energy is seen to be less than unity. The (cold/warm) counting rate for the scattered beam identified as N_{\parallel} in figure 4 would in fact be 0.90, but taking account of the detector solid angle and energy resolution raises this ratio to about 0.94. One can include the effect of preferential scattering of the radiation emitted at $\theta=90^\circ$ due to its plane-polarized nature by use of the measurement of $N_{\perp}/N_{\parallel}=1.58$ at $T=0.025^\circ\text{K}$ [3]. In this case the value of N_{\parallel} , normalized to unity for isotropic, unpolarized radiation, is 0.78 [12]. This is in agreement with the experimental measurement at that energy. It is seen that for photons losing more or less energy than those included in N_{\parallel} , the normalized intensity would be nearer unity, again in accord with experiment. A similar argument holds for measurements made with the gamma-ray detector placed to receive radiation emitted perpendicular to the c -axis.

The scattering phenomenon takes on particular significance in the present experiment because the small energy loss in 90° scattering places the "reverse anisotropy" region adjacent to the energy region of interest. Inclusion of some or all of this scattered radiation can be shown by multichannel analysis to reduce the measured value of A_2f_2 by a factor as high as 0.58. The discrepancy between the present data and that of the previous investigators [3] is of just this magnitude, and may in fact lie in the various gamma-ray pulse height analyzer settings. It might be pointed out that no attenuation factor for imperfections in the crystal lattice has been taken account of in the analysis of the present data. As was observed before, the presence of 1 percent cerium in the neodymium ethylsulfate crystal was found to have no measurable effect on the alignment results.

From the data taken with the cerium ethylsulfate crystal, $(A_2f_2)_{\max}=0.32\pm 0.03$. Since $f_2=0.571$ for complete alignment, $A_2\geq 0.30/0.57=0.52$. Assuming the zero-field Hamiltonian in neodymium ethylsulfate $\mathcal{H}=AS_zI_z+B(S_xI_x+S_yI_y)+CS_zT_z$, where T_z is the total spin component of the two nearest neighbor cerium ions [14] and where $B\sim 0.05A$, the energy levels and consequently the parameter $f_2(T)$ of the system can be calculated. From a comparison of the calculated temperature dependence of f_2 for various values of the hyperfine parameter A with the experimental A_2f_2 curve, taken with the limit on A_2 deduced from the cerium ethylsulfate data, the hyperfine constant $A=0.0305\pm 0.003\text{ cm}^{-1}$. If one assumes that $A=(\text{constant})\mu_N/I$, then using the value of the nuclear magnetic moment of Ce^{141} determined by Kedzie et al., [15] corrected by use of the more accurate value of $\langle r^{-3} \rangle$ due to Judd and Lindgren [16], one obtains the value 0.091 ± 0.01 for the constant. The constant thus calculated is

somewhat larger than the value calculated by Haag et al., [17] on the basis of the theory of Elliott and Stevens, [18] for cerium ethylsulfate.

The value of f_2 calculated at $T\sim 0.0162^\circ\text{K}$ is 0.414 ± 0.025 ; this number, taken with the experimental A_2f_2 at the same temperature yields for A_2 the value 0.495 ± 0.03 . The calculated temperature dependence of A_2f_2 for $A_2=0.495$ and $A=0.0305\text{ cm}^{-1}$ is shown in figure 1. Taking the attenuating effect due to the unobserved beta decay to be represented by the factor 0.97 [19], we find for the 145 keV gamma-ray of Pr^{141} an $E2$ to $M1$ mixing ratio [12] $\delta=+0.068\pm 0.008$.

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NOTE: Since this paper was prepared, data on the gamma-ray distribution from Ce^{141} oriented in neodymium ethylsulfate has been noted in the thesis of J. N. Haag (UCRL 9872, September 29, 1961 (unpublished)). These data are in close agreement with the corresponding data of the present authors.

5. References

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