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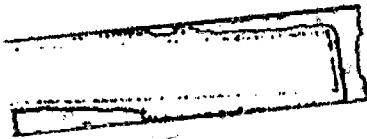
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ABSTRACT

Neodymium-147 was aligned and polarized at low temperatures in a neodymium ethylsulfate lattice. A saturation correction for susceptibility was verified. The effect of nondiagonal interactions on nuclear orientation was illustrated. Spin assignments of $5/2+$, $3/2+$, $5/2+$, and $5/2+$ were made for the excited states of Pm^{147} at 91, 410, 531, and 686 kev, respectively. Mixing ratios were obtained for six mixed γ rays in Pm^{147} . The magnitude of the amplitude mixing ratio $\delta(E2/M1)$ was found to be approximately proportional to γ -ray energy. Evidence was obtained that the β branches with end points at 0.23, 0.38, and 0.81 Mev are mostly of the $L=0$ type.

NUCLEAR ORIENTATION OF Nd^{147*}

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I. INTRODUCTION

Neodymium-147 and its daughter Pm^{147} are in a mass region where nuclear properties are not readily predicted from nuclear models. Considerable work had been done on the level scheme of Pm^{147} , and the level sequence of the four most heavily populated excited states has been known for some time.¹ Until recently, however, no spin assignments had been made for the highest three of these states. Recent angular correlation^{2,3} and conversion⁴ experiments yielded considerable information about the spins of these excited states and the multipolarities of several γ rays. A nuclear orientation experiment has been performed to fix these spin assignments, as well as to establish independently the multipolarities of six γ rays of mixed multipolarity.

Another purpose of this experiment was to check the validity of the saturation correction that must be made to the susceptibility of neodymium ethylsulfate, measured in a direction perpendicular to the c -axis of the crystal, in the presence of an external magnetic field parallel to this axis, in order to determine the absolute temperature. This correction is sometimes important in nuclear orientation experiments.^{5,6} In order to test it, one must consider the effects of nondiagonal interactions on alignment parameters. This is discussed in some detail in Sec. IV.

II. EXPERIMENTAL PROCEDURE

The apparatus and experimental procedure have been described elsewhere.⁵ Neodymium ethylsulfate was chosen as a lattice because the $T - T^*$ relationship is well known,⁷ and because the spin Hamiltonian for neodymium ions in this lattice is known. Polarization experiments were performed with the aid of an iron-free magnet drawing a current from storage batteries, and it was possible to measure the susceptibility of the crystal in a polarizing field rather accurately. The heat leak into the cryostat was kept low enough so that the change in the absolute temperature of the crystal was less than 3% during a typical 5-minute counting period. Only one count was taken per demagnetization, in order to minimize the effect of temperature inhomogeneities that develop as a demagnetized crystal warms up.

III. RESULTS

Gamma rays of energies 91, 276, 319, 398, 440, 531, and 686 kev were found to be anisotropic, and the angular distributions of these γ rays were studied as a function of temperature. The functional dependence of the angular distribution upon temperature was the same for all the γ rays investigated, with the individual anisotropies related by (+ or -) scale factors. All the angular distributions could be described by functions of the form $1 + \chi_2 P_2(\cos \theta)$ in the temperature range investigated.

Angular distributions of several γ rays at 0.020°K are shown in Fig. 1. The temperature dependence of the anisotropy for these γ rays, together with measurements taken in a polarizing field for the 531-kev γ ray, are shown in Fig. 2. The coefficients of the P_2 term, $B_2 U_2 F_2$, are listed in Table I together with certain derived quantities, for several γ rays, evaluated at $T = 0.025^\circ\text{K}$.

IV. DISCUSSION

A. The Alignment Parameters

The angular distribution of γ radiation from oriented nuclei is given by the expression⁸

$$I(\theta) = 1 + B_2 U_2 F_2 P_2(\cos \theta) + \dots, \quad (1)$$

where P_2 is a Legendre polynomial, U_2 and F_2 are specific nuclear parameters, and B_2 is the normalized second moment of nuclear spin projection along the orientation axis. Only the two terms above were necessary to fit the present data for all the γ rays investigated.

The parameter B_2 varies with temperature and is functionally dependent on the hyperfine-structure energy-level spacing. To calculate this level spacing, all the terms in the relevant spin Hamiltonian must be known. Thus B_2 does not depend on nuclear properties alone, and considerable information is required about nonnuclear interactions in order to determine nuclear moments from nuclear orientation experiments. Although a good approximation to the spin Hamiltonian relevant to nuclear orientation experiments on a given isotope may be obtained from the paramagnetic-resonance spin Hamiltonian of a stable isotope of the same element in a magnetically dilute isomorphous crystal, additional terms may be necessary to account for quadrupole interaction⁵ or dipole-dipole interactions.^{5,6,9} The former is sometimes not detectable by resonance experiments, whereas the latter is usually important in concentrated paramagnetic salts.

If some of the terms in the spin Hamiltonian are not diagonal in the $(S'S_z'II_z)$ representation, it becomes necessary to solve secular equations to determine the energy levels and to diagonalize the representation in energy with the new base vectors, expanded in terms of those of the $(S'S_z'II_z)$

representation, in order to calculate the orientation parameters. Consider, for example, an n -fold representation with energy levels E_i ($1 \leq i \leq n$) whose base vectors $|i\rangle$ may be expanded in terms of the base vectors $|j\rangle$ of the $(S'S_z'I_{I_z})$ representation, thus: $|i\rangle = a_{ij} |j\rangle$. Then the expression for the orientation parameters¹⁰ becomes

$$B_k = (2I+1)^{1/2} \sum_i W(i) \sum_j a_{ij}^2 C_{M_j - I, k}^{I, 0} (-1)^{I-M_j}. \quad (2)$$

In the Gorter-Rose method of nuclear polarization^{11, 12} the appropriate spin Hamiltonian is partially diagonalized in the $(S'S_z'I_{I_z})$ representation by application of a magnetic field H_z , which reduces the influence of the off-diagonal terms on the energy level spacing. It should be remembered, however, that I_z is not a good quantum number in applied fields of several hundred gauss, and the effect of off-diagonal terms must be taken into account in the interpretation of nuclear orientation data.

For this experiment the spin Hamiltonian is

$$\mathcal{H} = g_{\parallel} \beta H S_z + A S_z I_z + B(S_x I_x + S_y I_y) + C S_z T_z. \quad (3)$$

The corresponding energy-level diagram is shown in Fig. 3. Following Elliott and Stevens,¹³ the quadrupole interactions term was estimated and found to be negligible. The values of A and B were obtained by comparison of paramagnetic-resonance data for Nd^{147} in lanthanum magnesium nitrate¹⁴ with data for Nd^{143} in lanthanum magnesium nitrate and in lanthanum ethylsulfate.¹⁵ This yields $A = 0.0289 \pm 0.0001 \text{ cm}^{-1}$ and $B = 0.01512 \pm 0.00005 \text{ cm}^{-1}$.¹⁴ The last term in $\mathcal{H}$ takes account of dipole-dipole interactions between the Nd^{147} ion and its two nearest neighbor neodymium ions in the lattice. The total spin operator for these two neighbors is denoted by T , and T_z can have any of the values 1, 0, or -1. At high

temperatures the relative populations of these three values approach 1:2:1. At lower temperatures and in a magnetic field, the relative populations must be weighted with the appropriate Boltzmann factors. The value of C is not adjustable, but is calculated from crystallographic¹⁶ and resonance¹⁵ data. For this experiment $C = 0.056 \pm 0.001 \text{ cm}^{-1}$. The effects of next-nearest, etc., neighbors on the alignment parameters can easily be shown to be negligible.¹⁷ This interaction was observed in early paramagnetic-resonance experiments.¹⁸

When the magnetic susceptibility perpendicular to the c axis, $\chi_{\perp}$, is measured in the presence of an external field H along the c axis in order to determine the temperature in a nuclear polarization experiment, a saturation correction to $\chi_{\perp}$ must be made. For $S' = 1/2$ this correction takes the form⁵

$$\frac{\chi_{\perp}^H}{\chi_{\perp}^0} = \frac{\tanh(g_{\parallel} \beta H / 2kT)}{g_{\parallel} \beta H / 2kT} \quad (4)$$

This correction has been necessary in several investigations performed in this laboratory.^{5,6} It usually amounts to about 10% for moderate polarizing fields, and must be known accurately in nuclear moment determinations. In order to check the validity of Eq. (4) and to illustrate the variation of B_2 with external field in the presence of nondiagonal interactions, γ ray anisotropies were measured in the presence of polarizing fields of 100, 200, and 400 gauss along the c axis. The results, along with theoretical curves calculated from Eqs. (1) through (3), normalized empirically for the nuclear parameters $U_2 F_2$, are shown in Fig. 2. We conclude that Eq. (4) holds very well under these conditions.

B. Energy Levels in Pm¹⁴⁷

In addition to determining nuclear moments of the parent nucleus, the techniques of nuclear orientation may also be used to study the character of the β decay or to measure spins of excited states and determine multiplicities of γ rays in the daughter. In experiments involving γ -ray anisotropies, the quantity $B_k U_k F_k$ is measured directly and comparison with calculated values of B_k , for example, yields the product $U_k F_k$ for each γ ray. If the initial and final spins and multipolarity of the observed γ transition are known, F_k may be calculated and compared with the experimental value of $U_k F_k$ to obtain U_k , a reorientation parameter dependent on the angular momenta involved in the preceding unobserved transitions. Conversely, if U_k can be calculated, comparison with experiment yields F_k for the observed γ ray. This F_k function is a valuable parameter and cannot be obtained directly from angular correlation experiments alone, which measure only products of F_k 's.

In this experiment we have obtained $U_2 F_2$ products for several γ rays. Combining these with previous nuclear orientation data¹⁹ angular correlation data,^{2,3} and conversion data^{1,4} we have made spin assignments to the excited states of Pm¹⁴⁷ at 91, 410, 531, and 686 keV. Using the conversion data as a guide, we have assigned accurate M1-E2 mixing ratios to several γ rays. We also obtained evidence that three of the β transitions are mostly of the type $L=0$. In the following detailed discussion, frequent reference will be made to the F_2 parameters and the mixing ratios δ (E2/M1) for individual γ rays. It should be noted that a given mixed γ ray will in general have two values of δ (E2/M1) differing only in sign,^{20,21} and two values of F_2 . One set of values of F_2 and δ is associated with the γ ray when it is first

in a cascade and one when it is last. To distinguish these two sets of values we shall add the superscript "F" or "L", for "first" or "last" in a cascade, to the symbol in question. This notation is applicable to multiple cascades in which the first and last transitions have the same values of F_2 as if there were no intermediate transitions, with the latter serving only to attenuate the correlation through U_2 factors.^{22,23} In nuclear orientation experiments, the axis of quantization is determined by crystal axes rather than by the direction of propagation of preceding radiations, and F_k^F and δ^F may be determined separately for each γ ray. In the following discussion, the product $U_2 F_2$ was obtained for each γ ray by dividing the value of $B_2 U_2 F_2$ evaluated at $T = 0.025^\circ\text{K}$ (Table I) by the value $B_2(T = 0.025) = 0.277$, calculated from Eqs. (2) and (3).

The 91-keV Level

The evidence in favor of a spin and parity assignment $5/2+$ to this level is overwhelming. The $\log ft$ of 7.4 for β decay from the $5/2-$ ground state of Nd^{147} indicates a first-forbidden transition with $\Delta I = 0$ or 1. The predominantly M1 multipolarity of the 91-keV transition to the $7/2+$ ground state of Pm^{147} leaves only the assignments $5/2+$ and $7/2+$ as possibilities, and our value of $F_2^2(91) = +0.307 \pm 0.025$ is in good accord with the $5/2+$ assignment if this transition is 1.2 to 2.1% E2. In particular, an excited-state spin assignment of $7/2+$ is absolutely excluded by this measurement for an E2 admixture of 48% or less. Reinterpretation of the Oxford directional and polarization anisotropy data¹⁹ yields essentially the same results. The g factor of $+1.42 \pm 0.20$ recently obtained by Bodenstein et al.² for this state would independently exclude a shell-model $7/2+$ state. A recent experiment by Arya led to an assignment of $7/2+$ for the 91-keV state, but the analysis yielded mixing ratios for the 91- and 319-keV γ ray in disagreement with other work.

Our F_2 and δ above were based on the assumption that the β -decay branch to the 91-keV level is mostly of the type $L=0$, with no net angular momentum carried off by the electron-antineutrino pair. For $L=1$, we obtain $F_2^L(91) = +0.45 \pm 0.04$, requiring that the transition be $5 \pm 1\% E2$. But this is in serious disagreement with the conversion coefficients.^{1,4} Thus our results indicate that this β transition has $L=0$.

The 410-keV Level

In three angular correlation measurements on the 319—91-keV cascade in Pm^{147} , Lindqvist,²⁴ Bodenstedt et al.,² and Arya³ obtained, respectively values of -0.097 ± 0.007 , -0.87 ± 0.008 , and -0.1030 ± 0.0298 for the A_2 coefficient, and $+0.023 \pm 0.018$, -0.001 ± 0.003 , and $+0.0107 \pm 0.0099$ for the A_4 coefficient. Because the predominantly $M1$ multipolarity^{1,4} of the two γ rays disagrees with the two larger A_4 values, we shall use the results of Bodenstedt et al. in our analysis. Combining our $F_2^L(91)$ with $A_2 = -0.087 \pm 0.008$, we obtain $F_2^F(319) = -0.290 \pm 0.036$. Here, as in the rest of our discussion, the error given for derived quantities is the standard deviation. In Table II are listed the values of $F_2^L(319)$ and δ_{319}^L obtained by combining this value of $F_2^F(319)$ with each of the possible spins $3/2$, $5/2$, and $7/2$ for the 410-keV state. We conclude that the $5/2+$ assignment is ruled out, while the $3/2+$ and $7/2+$ assignments are equally acceptable thus far.

The 531-keV Level

From the β decay and multiplicities of the 276-keV and 531-keV γ rays, only spin and parity assignments of $3/2+$, $5/2+$, or $7/2+$ are possible for the levels at 531 and 686 keV. This experiment absolutely excludes the $3/2+$ possibility for either of these levels. If either level had spin and parity $3/2+$, the direct transition to the ground state would be pure $E2$ and would have $F_2^L = -0.143$. The experimental F_2^L values in Table I are nearly four

times this large. Thus only assignments of $5/2+$ and $7/2+$ are left. The $5/2+$ assignment for the 531-kev state may be established in two completely independent ways. The first involves comparison of the Oxford polarization data on the 531-kev γ ray with conversion data.^{1,4} A predominantly M1 γ ray is consistent with a large negative polarization only if the spin of the 531-kev state is $5/2+$. The second method involves comparison of $A_2(440-91) = +0.065 \pm 0.010$ ² with our $F_2^L(91) = +0.307 \pm 0.025$ to obtain $F_2^F(440) = +0.212 \pm 0.037$. The assignments $5/2+$ and $7/2+$ for the 531-kev level then yield $F_2^L(440) = -0.71 \pm 0.01$ and $+0.41 \pm 0.05$, respectively. Our experimental value of $F_2^L(440) = -0.78 \pm 0.20$ leaves only the $5/2+$ assignment. Using this, we find $F_2^L(531) = -0.486 \pm 0.030$ for $L=0$ β decay to this state, and $F_2^L(531) = -0.74 \pm 0.04$ for $L=1$. The lowest possible value of F_2^L for the sequence $5/2 (D, Q) 7/2$ is -0.48 , and we conclude that the β transition is predominantly of the $L=0$ type. Values of δ^L for the 531- and 440-kev γ rays derived from this experiment are given in Table I.

The 686-kev Level

As discussed above, only spin and parity assignments of $5/2+$ and $7/2+$ are possible for this level. Both Bodenstein et al.² and Arya³ have measured the angular correlation of the 319-kev-276-kev cascade, and their values for A_2 of $+0.0117 \pm 0.0025$ and $+0.0710 \pm 0.0162$, respectively, are in very poor agreement. Only the former result agrees with our experiment, and this agrees only for the sequence $5/2+ (D, Q) 7/2+$ for the 276-kev transition. A value of $F_2^L(276) = +0.21 \pm 0.10$ was obtained from this experiment, in fair agreement with $F_2^L(276) = +0.14 \pm 0.02$ obtained by comparison of $A_2 = +0.0117 \pm 0.0025$ with $F_2^L(319) = -0.30 \pm 0.05$ to yield $\delta_{276}^L = +0.14 \pm 0.02$. All other combinations of spins give $F_2^L(276) < 0$.

In view of the poor agreement of the angular correlation measurements, it is difficult to have as much confidence in the spin assignments for the states at 410 kev and 686 kev as in the other two excited-state spin assignments. The really crucial datum on which the respective assignments of $3/2+$ and $5/2+$ depend, however, is the sign of $F_2^L(276)$ in our experiment. Our least accurate data were those obtained on this γ ray. We have plotted the ratio of warm to cold counting rates along the crystal axis in the 276-kev region in Fig. 4. The peak-to-background ratio is about 1:1 for this peak, and the background is anisotropic, with $(K_c/I_w) < 1$. The dip at 276 implies that (I_c/I_w) for this peak is considerably greater than for the background radiation. A quantitative evaluation yields $(I_c/I_w) > 1$, which requires $F_2^L(276) > 1$. We note that the next best possible assignments are $5/2+$ for the 686-kev state and $7/2+$ for the 410-kev state. Thus an accurate determination of the multipolarity of the 410-kev transition to the ground state would be especially valuable. In fact, the very low intensity of this transition provides indirect evidence against any spin other than $3/2+$ for the 410-kev state, inasmuch as the probabilities for E2 transitions are somewhat less than those for M1 transitions for several other γ rays in Pm^{147} . The decay scheme that fits all the foregoing data is shown in Fig. 5.

Promethium-147 is very near the region $150 < A < 190$, where nuclear properties may be described by the collective model.²⁵ The partial E2 transition probability is of interest in this connection, as the deviation of this parameter from the single-particle value should provide a rough measure of the extent to which collective behavior may be expected. In Fig. 6 we have plotted the comparative lifetimes for E2 transitions in this region against neutron number, in the manner of Goldhaber and Sunyer.²⁶ By combining the E2/M1 mixing ratio of the 91-kev γ ray determined in this experiment with

with the lifetime of the 91-kev state of Pm^{147} ,² the partial E2 transition probability for this transition may be obtained. The position of this point on the curve, (a) suggests that Pm^{147} is fairly well up out of the collective region, and (b) provides further evidence that the onset of collective behavior is rather sharp at $A \approx 150$, whereas this behavior changes more slowly with neutron number near the higher end of this region, for $A \approx 190$.²⁶

An unusual correlation, which is available here, is illustrated in Fig. 7, where we have plotted $|\delta(E2/M1)|$ against energy for six γ rays. From Moszkowski's Table²⁷ we find the ratio of E2 to M1 transition probabilities on a single-particle model is given by

$$\frac{T(E2)}{T(M1)} = 5.7 \times 10^{-6} A^{4/3} E_{\gamma}^2 S_{E2}/S_{M1}.$$

Using the definition of δ , one obtains, for $A = 147$,

$$|\delta| \sqrt{S_{M1}/S_{E2}} = 0.066 E_{\gamma}. \quad (5)$$

The statistical factors have been omitted in Fig. 7 because they cannot be calculated without a knowledge of the orbital angular momentum of each single-particle state. For at least one set of ℓ assignments the linearity of the plot is made worse by inclusion of the statistical factors. With this qualification, we observe that Fig. 7 is in fair agreement with the form of Eq. (5) for most of the transitions, but with a slope of 1.4, rather than $0.066 (\text{Mev})^{-1}$. Thus the M1 transition probabilities are hindered by a factor of about 450 relative to the E2 transition probabilities in Pm^{147} .

The β decay in this system is not easily understood and gives evidence of an unfamiliar selection rule. Our experiment suggests that the $L = 1$ components of first-forbidden β decay to the five levels in question are retarded or absent. This in turn may explain the absence of β decay to the $7/2+$ ground state, the possible absence of decay to the 410-kev spin

$3/2+$ state, and the $L=0$ character of the β branches to the other three states. These $L=0$ assignments are obtained indirectly and we cannot place very much confidence in them. A different experiment to check this point would be very desirable. Among such experiments are (a) direct measurement of the β -particle anisotropy, and (b) measurement of the γ -ray anisotropies at a temperature low enough to produce saturation of the alignment.

APPENDIX

After this paper had been written we received a communication from G. T. Ewan et al.²⁸ They had made spin and parity assignments for the excited states of Pm^{147} identical to ours, except that they had not eliminated the $3/2+$ possibility for the 686-kev state. Their multipolarity assignments for the mixed γ rays were in good agreement with ours. Such agreement between independent measurements is very encouraging, and we conclude that these experimental aspects of the decay of Nd^{147} are well understood. A theoretical explanation of the β decay would be very interesting.

End of Appendix

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Table I. Values of several quantities characterizing the decay of Nd^{147} oriented in neodymium ethylsulfate as determined by this experiment.

Energy level	E_γ	$B_2 U_2 F_2$ at $T=0.025^\circ\text{K}$	F_2^L	$\delta_{E_\gamma}^L$	$\%E2$	Type of β decay to level	Spin of level
91						$L = 0$	$5/2+$
	91	$+0.0906 \pm 0.005$	$+0.307 \pm 0.025$	$+0.13 \pm 0.02$	1.6 ± 0.4		
410							$3/2+$
	319	-0.058 ± 0.01	-0.30 ± 0.05	-0.36 ± 0.02	11.5 ± 1.1		
489							$(7/2+)$
	398	< 0	< 0		$2 \pm 1^{(a)}$		
531						$L = 0$	$5/2+$
	121				$2.8^{(b)}$		
	440	-0.24 ± 0.04	-0.78 ± 0.20	$+0.82 \pm 0.65$	33 ± 30		
	531	-0.149 ± 0.006	-0.486 ± 0.030	-0.95 ± 0.30	45 ± 15		
686						$L = 0$	$5/2+$
	276	$+0.064 \pm 0.03$	$+0.21 \pm 0.10$	$+0.14 \pm 0.02$	2.0 ± 0.6		
	686	-0.163 ± 0.003	-0.532 ± 0.05	-0.95 ± 0.30	45 ± 15		

(a) Assuming spin $7/2$ for the 489-kev level

(b) Obtained from Reference 4.

Table II. Parameters F_2^L and δ^L for the 319-kev γ ray of Pm^{147} with various spins assumed for the 410-kev level.

Spin of 410-kev level	3/2+	5/2+	7/2+
$F_2^L(319)$ from 319-91 angular correlation	-0.24 ± 0.02	-0.55 ± 0.03	-0.33 ± 0.07
$F_2^L(319)$ from nuclear alignment	-0.30 ± 0.05	-0.35 ± 0.06	-0.26 ± 0.04
$\delta^L(319)$ from 319-91 angular correlation	-0.36 ± 0.02	$+0.13 \pm 0.03$	$+0.37 \pm 0.05$
% E2 from correlation data	11.5 ± 1.1	1.8 ± 0.8	12.2 ± 2.8

Figure Captions

Fig. 1. Normalized angular distribution of three γ rays at 0.01°K . Normalized theoretical curves are fitted to the data.

Fig. 2. Temperature dependence of the angular distribution of three γ rays of Pm^{147} following the decay of Nd^{147} oriented in neodymium ethylsulfate. Intensities are measured along the crystal c axis. Data for the 531-keV γ ray include points taken in external magnetic fields of 0, 100, 200, and 400 gauss. Data are shown with theoretical curves calculated by using Eqs. (1), (2), and (3). Curves are normalized to the data at $1/T = 40$.

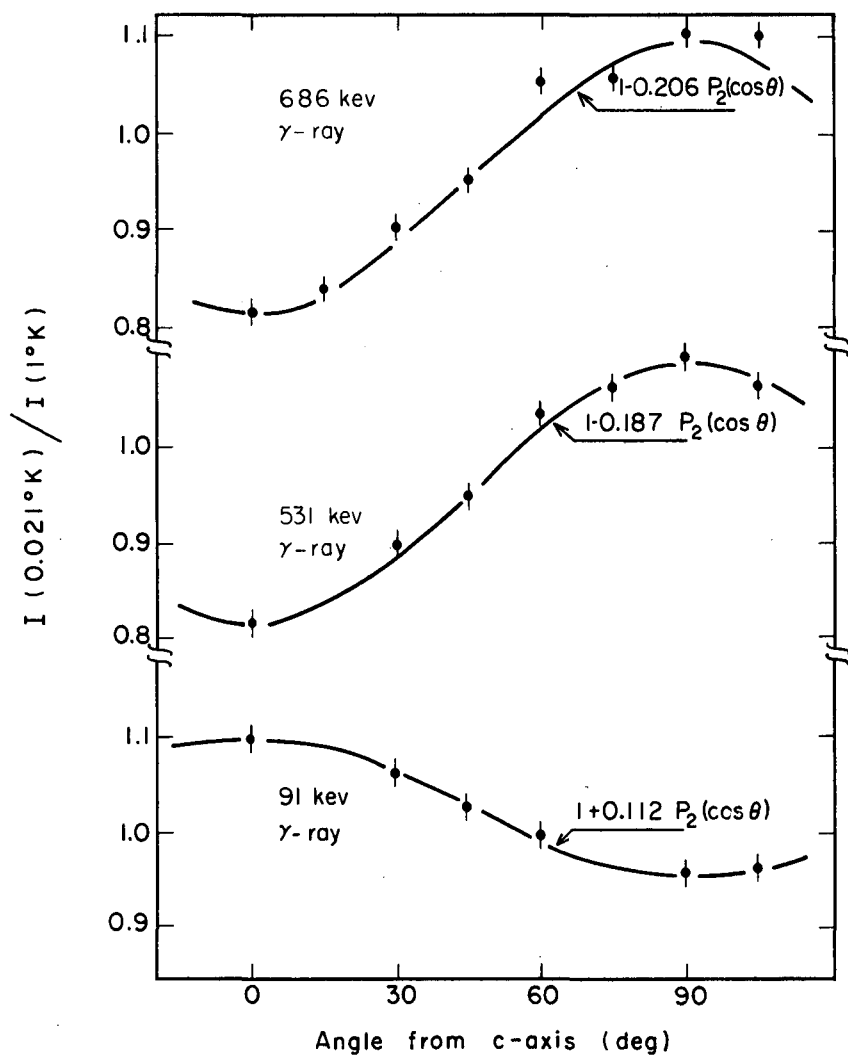
Fig. 3. Energy-level diagram showing the effects of several terms on the hyperfine splitting of the ground-state electronic doublet of Nd^{147} in neodymium ethylsulfate. (From Eq. 3.)

Fig. 4. Relative decrease in counting rate along the crystal axis in the 276-keV region of the energy spectrum obtained from decay of Nd^{147} . Ordinate is ratio of the isotropic counting rate to the counting rate at $T = 0.021^\circ\text{K}$.

Fig. 5. Decay scheme of Nd^{147} , including spin assignments for the energy levels in Pm^{147} as determined by this experiment.

Fig. 6. Comparative E2 lifetimes plotted vs neutron number for several isotopes. The symbol $\langle \rangle$ denotes a mixed gamma transition. Points are calculated from data obtained by a search of the literature to February 1961.

Fig. 7. Absolute magnitude of the mixing ratio, $\delta(\text{E2/M1})$, for several γ rays of Pm^{147} .



MU-22769

Fig. 1.

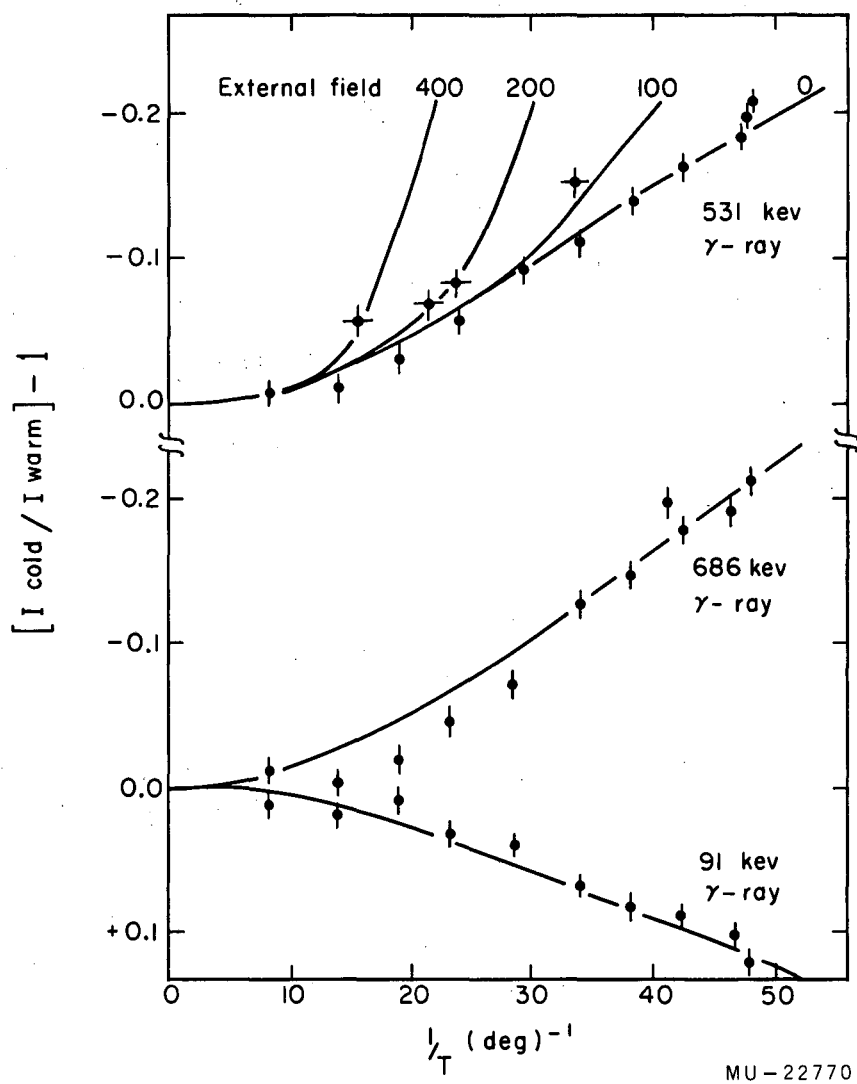


Fig. 2.

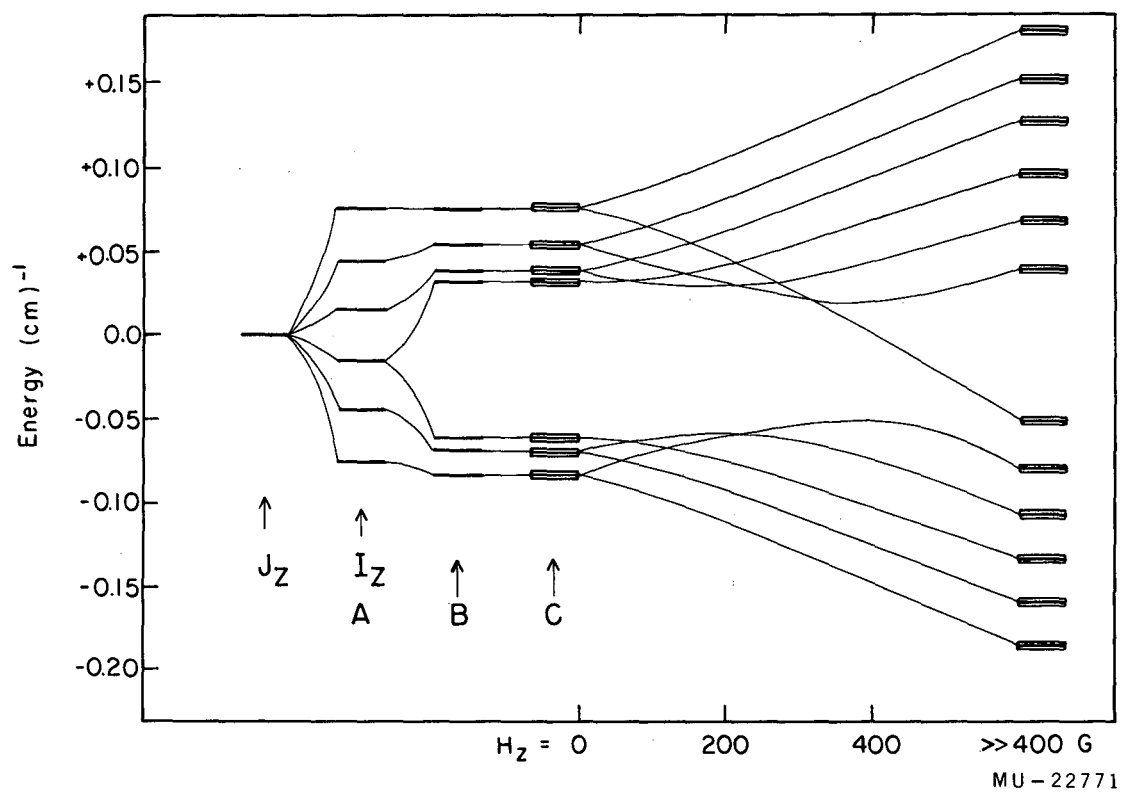
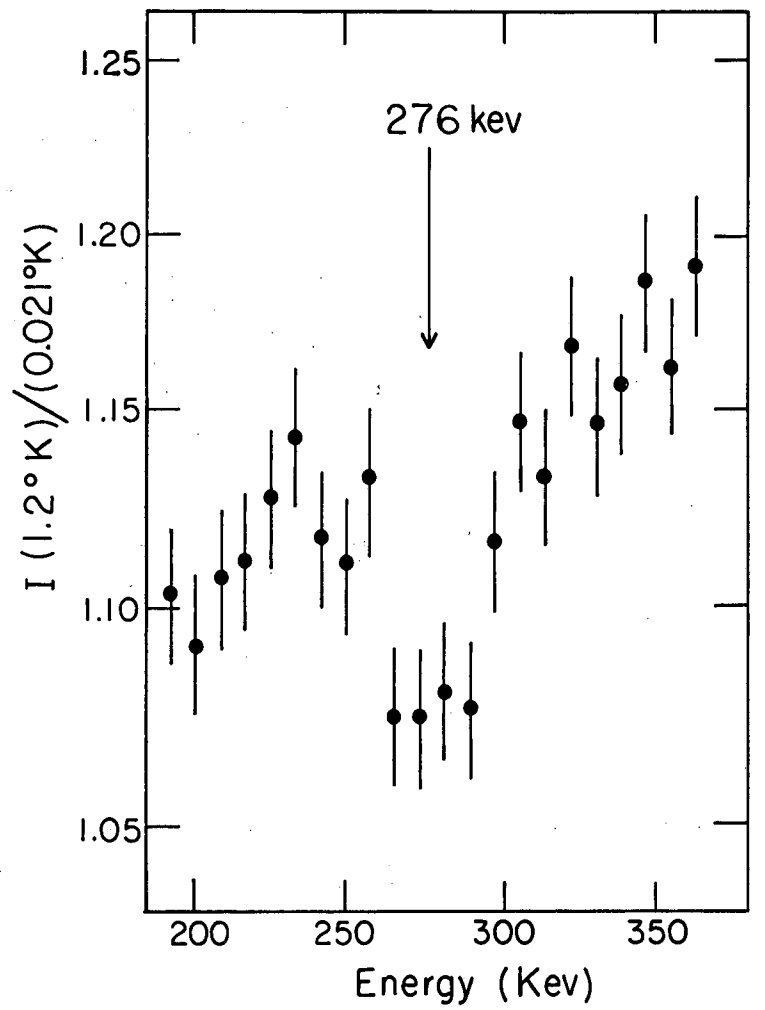
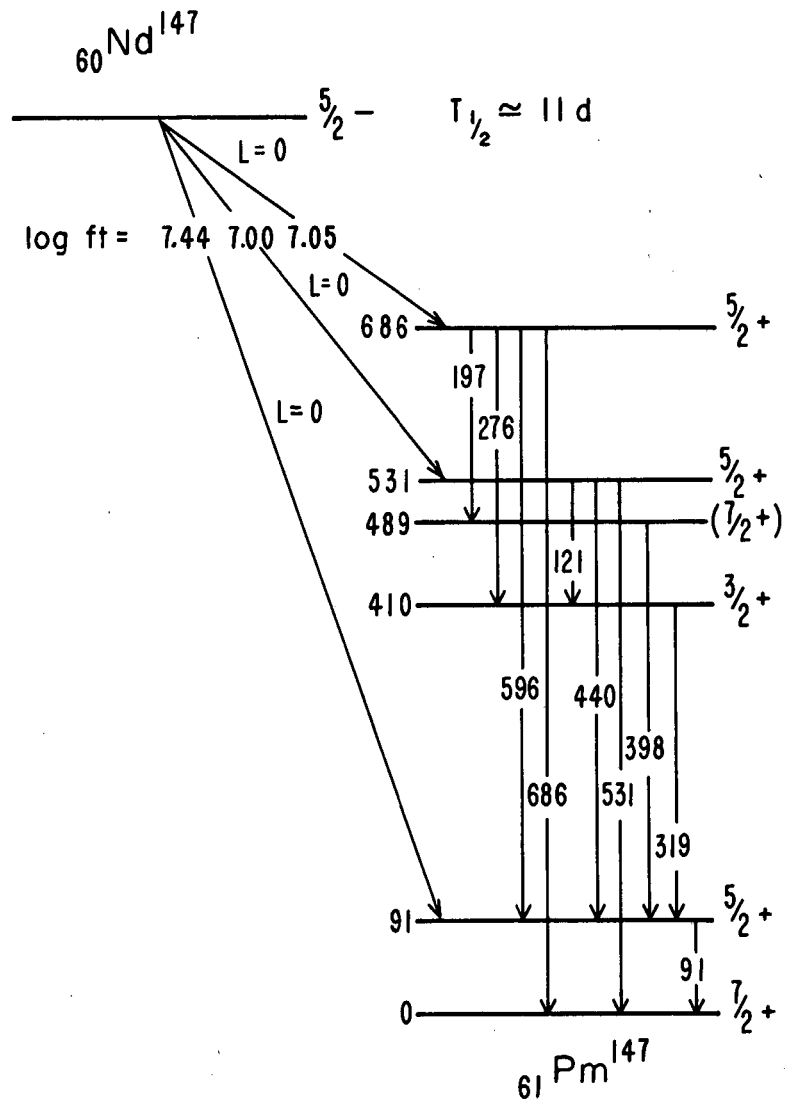


Fig. 3.



MU-22773

Fig. 4.



MU - 22772

Fig. 5.

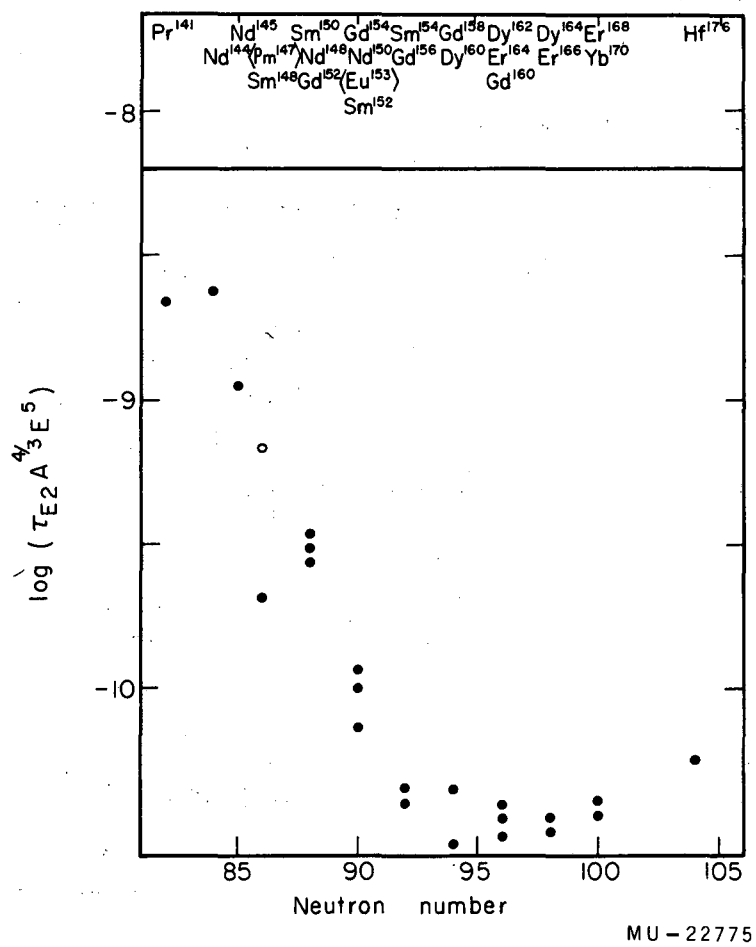


Fig. 6.

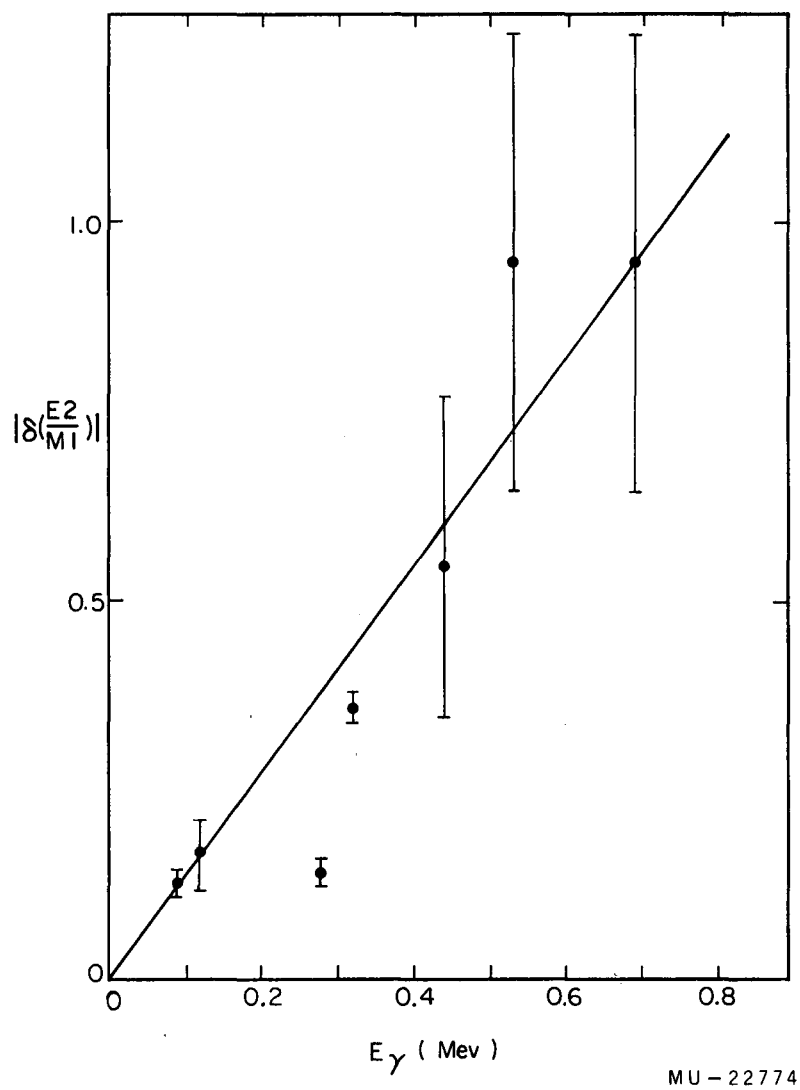


Fig. 7.

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