Nuclear Orientation and Reorientation Studies with Nd^{147}*

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The results of new nuclear orientation measurements on Nd^{147} and reanalysis of old data in light of the revised temperature scale for neodymium ethylsulfate (NES) are presented. Previously assigned spins in Pm^{147} are confirmed, while several γ-ray mixing ratios are altered somewhat, and the previously reported $L=0$ character of several β branches is found to be incorrect. Auxiliary experiments in cerium magnesium nitrate (CMN) give further support to the new NES temperature scale. The attenuation factors of the 91-keV level in Pm^{147} were determined for the two lattices to be $G_2(91)_{NES}=0.89\pm0.11$ and $G_2(91)_{CMN}=0.42\pm0.12$. The latter is in poor agreement with Daniels and Misra's theoretical values. Available data on reorientation following decay of oriented nuclei in paramagnetic salts are summarized, and are interpreted as indicating dependences on both evironment and intermediate-state lifetime.

I. INTRODUCTION

SEVERAL nuclear-orientation studies[1–4] of the decay of Nd^{147} have been reported, the most recent and extensive being that of Westenbarger and Shirley, hereafter referred to as I. Spin assignments were made in I for several states in Pm^{147} and mixing ratios were derived for several γ rays. In addition three β branches were found to be largely of $L=0$ character (i.e., with the leptons carrying away no net angular momentum).

Studies with other experimental methods[5,6] have supported these spin assignments, but it was subsequently discovered[7,8] that the temperature scale for neodymium ethylsulfate (NES), upon which some of the multipolarity results reported in I were based, was in error. In Secs. II and III, we report the results of a reanalysis of the data in I along with additional data from nuclear orientation experiments on Nd^{147} in both the NES and the cerium magnesium nitrate (CMN) lattices. The new data provide another test of the new NES temperature scale and yield the attenuation factor G_2 for the 91-keV level of Pm^{147} in both lattices.

II. EXPERIMENTAL

The nuclear orientation apparatus has been described elsewhere.[9] The new measurements are more extensive than those reported in I in that CMN was used as well as NES and in that both NaI(Tl) and Ge(Li) detectors were employed. To take advantage of the new temperature scales for the two paramagnetic salts,[8,10] the temperatures attained on demagnetization were determined from the initial magnetizing fields and temperatures preceding demagnetization. Irreversible effects during demagnetization were found to be negligible. We compensated for irreversible effects *after* demagnetization by extrapolation backward in time to the moment of demagnetization. Counting was done at 0° and 90° from the crystalline *c* axes, mostly on the 531-keV γ ray. The temperature dependence of the normalized intensity function $W(0°)-W(90°)$ for this γ ray is shown in Fig. 1 for CMN and in Fig. 2 for NES. The relative anisotropies of the 91- and 531-keV γ rays were studied in both CMN and NES, and results are shown in Table I.

III. DISCUSSION

A. Spins, Multipolarities, and β-Decay Parameters

The interpretation of the nuclear orientation data in I was based on the observation that the angular distributions of all the γ rays studied could be represented by the well-known[11] expression $W(\theta)=1+B_2U_2F_2P_2(\cos\theta)$, where θ is the angle from the crystalline *c* axis. In I the orientation tensor B_2 was calculated from the spin Hamiltonian of Nd^{3+} in the ethyl-sulfate lattice, using the known (but erroneous) temperature scale for NES: U_2F_2 for each γ ray could then be obtained by comparing $W(\theta)$ with B_2. We could reinterpret the old results simply by using the new temperature scale,[8] but we have instead chosen a way that does not require a detailed knowledge either of the spin Hamiltonian or of the absolute temperature. By using CMN as a lattice for orientations, we were able to achieve almost complete saturation of the nuclear orientation (Fig. 1). This

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TABLE I. Spins and γ-ray mixing ratios in Pm^{147}.

E (keV)	$I\pi$	E_γ (keV)	$G_2U_2F_2$[a]	F_2^L	$\delta(E_\gamma^I)$	% $E2$
91.1	$\frac{5}{2}+$	91.1	+0.202(14)[b]	≥0.202, +0.259(7)[c]	+0.089(5)[d]	0.8(1)
410.5	$\frac{3}{2}+$	319.4	−0.12(2)	−0.21(7)	+0.55(5)	23(3)
488.2	($\frac{7}{2}+$)	397	<0	<0	···	2(1)
531.0	$\frac{5}{2}+$	120.5	···	···	···	~2
		439.8	−0.485(80)	−0.63(23)[e], −0.70(2)[f]	+0.70(9)	33(6)
		531.0	−0.300(12)	−0.45(8), −0.42(5)	−0.69(32)	31(19)
685.8	$\frac{5}{2}+$	275.4	+0.13(6)	+0.18(8), +0.157	−0.112(6)	1.2(1)
		685.8	−0.329(6)	−0.42(9)	−1.05(65)	44(30)

[a] Obtained from the saturation value for the 531-KeV γ ray in CMN.
[b] Errors in last place are given parenthetically.
[c] Calculated from the known spins and the mixing ratio from Ref. 5. The sign is from nuclear orientation data, which give $F_2=+0.255(70)$.
[d] Error taken to correspond to 0.1% error in E2 admixture, which we have assumed.
[e] In extracting F_2's from U_2F_2's we have assumed that the B_{ij} matrix element contributes negligibly to the β-decay *intensity*.
[f] Where a second, more accurate value of F_2 is given, it was determined by combining nuclear orientation and other data.

provides an unambiguous determination of B_2 and thus of U_2F_2. For the 531-keV γ ray we found $U_2F_2=-0.300\pm0.012$. Normalization of the results for other γ rays from I to this figure gave the U_2F_2 values set out in Table I. Also given are spin and multipolarity values that are consistent with a reanalysis of these data together with angular correlation and conversion data. With this new analysis it is not possible to set useful limits on the ratios of $L=0$ to $L=1$ character in the β branches. Thus the "unfamiliar selection rule" (i.e., no $L=1$ character), which the authors of I found puzzling, does not exist.

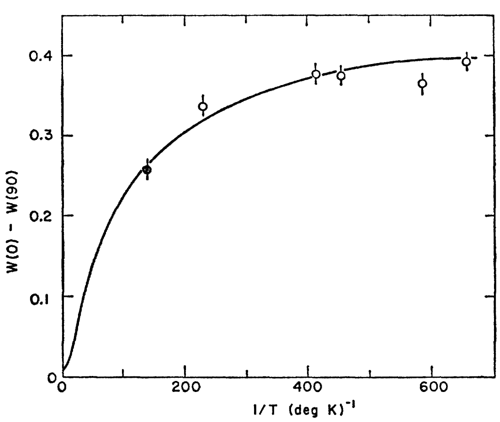

FIG. 1. Temperature dependence of the function $W(0)-W(90)$ for the 531-keV γ ray following the decay of Nd^{147} oriented in CMN, and theoretical curve based on $U_2F_2=0.300$. Solid-angle correction has not been made. To make it, both data and curve should be divided by 0.930.

B. NES Temperature Scale

Comparison of the $W(\theta)$ data for the 531-keV γ ray from NES with $U_2F_2(531)$ yielded B_2(NES), which is shown in Fig. 2. A theoretical curve based on the known spin Hamiltonian (discussed at length in I) is also shown. The over-all satisfactory agreement and experiments on Sm,[12] Eu,[13] Tb,[14] and Lu [8] in this lattice

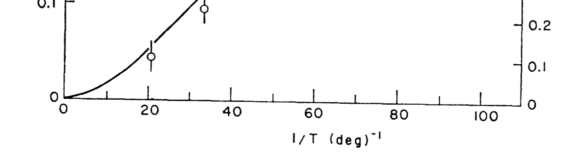

FIG. 2. Temperature dependence of $W(90)-W(0)$ for the 531-keV γ ray from NES. The statistical tensor B_2 (right ordinate scale) is derived from the data using $U_2F_2=0.300$ and the solid-angle correction of $g_2=0.933$. Calculated curve based on the new temperature scale is shown.

[12] M. Kaplan, J. Blok, and D. A. Shirley, preceding paper, Phys. Rev. **184**, 1177 (1969).
[13] J. Blok and D. A. Shirley, Phys. Rev. **143**, 278 (1966); **143**, 911 (1966).
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support the new temperature scale. The systematic deviations at the lowest temperatures may arise in part from errors in the temperature scale itself, but are almost certainly also partly due to (ferromagnetic) collective effects in NES. These effects would tend to diagonalize the spin Hamiltonian and increase B_2.

C. Attenuation in the 91-keV State of Pm^{147}

A portion of the decay scheme of Nd^{147} is shown in Fig. 3. In earlier work[1,2] on Nd^{147} in CMN, Ambler *et al.* found anisotropy in the angular distribution of the 531-keV γ ray, but none for the 91-keV γ ray, outside their sensitivity of 5%. Some attenuation is expected in the 91-keV, $t_{1/2}=2.50$-nsec state[15] from which this γ ray proceeds. The Oxford group, however, using the NES lattice, found a large anisotropy in the 91-keV γ ray as well. Our measurements confirm the results of both groups and provide a value for the anisotropy of the 91-keV γ ray in CMN.

The results summarized in Tables I and II clarify the experimental aspects of this problem. Writing the angular distribution functions for the 91-keV γ ray $W(\theta)=1+B_2U_2G_2F_2P_2(\cos\theta)$, to account for intermediate-state reorientation, and assuming no reorientation in the 531-keV state of Pm^{147} (because it is too short-lived), we may take ratios to find $G_2(91)_{\text{CMN}}/G_2(91)_{\text{NES}}=0.36$. From the $M1$-$E2$ mixing ratio of the 91-keV transition, determined from L-subshell ratios,[5] we may calculate $F_2(91)=+0.259$. Comparison with $G_2U_2F_2=0.202$ gives $G_2U_2=0.78$. Since $U_2\leq 1$, we find $G_2(91)_{\text{NES}}=0.89\pm0.11$, and therefore $G_2(91)_{\text{CMN}}=0.42\pm0.12$.

D. Survey of Other Attenuation Results

The theoretical situation regarding intermediate-state attenuation is far from clear. We shall not discuss it in detail because this has recently been done in a survey article by Daniels and Misra.[16] The basic problem is that no one knows what really happens to the atomic configuration in the intermediate state. Daniels and Misra made

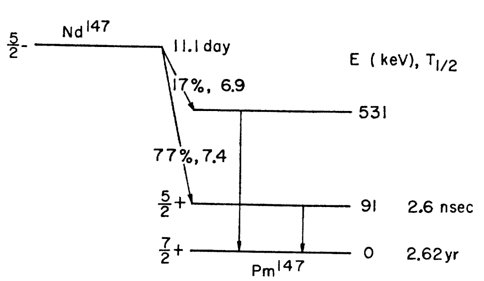

FIG. 3. Portion of the decay $Nd^{147}\rightarrow Pm^{147}$ relevant to this work.

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[16] J. M. Daniels and S. K. Misra, Can. J. Phys. **44**, 1965 (1966).

TABLE II. Attenuation factors for the 91-keV γ ray in CMN.

Run No.	$W(0)_{91}$[a]	$W(0)_{531}$	$R=\frac{1-W(0)_{91}}{W(0)_{531}-1}$	R (corr.)
1	0.964(8)	1.164(14)	0.22(5)	0.24(5)
2	0.976(8)	1.145(14)	$0.16_5(6)$	0.18(6)
3	0.972(8)	1.213(14)	$0.22_5(4)$	0.24(4)
4	0.960(8)	1.213(14)	0.19(14)	0.20(4)

Av. ratio $G_2U_2F_2(91)/G_2U_2F_2(531)=-0.21_5(3)$

Av. ratio from Ge(Li) detector runs $=-0.22(3)$

Final ratio for CMN $=-0.22(3)$

Ratio for NES $=-0.61(4)$

Ratio of ratios, $G_2(91)_{\text{CMN}}/G_2(91)_{\text{NES}}=0.36(5)$

[a] Errors in last place are given parenthetically. Only the results of runs on the CMN lattice with a NaI(Tl) detector are presented in detail, to illustrate consistency. Runs under other conditions were similar.

several calculations of G_2 based on the assumption that daughter Pm has the same (time-independent) electronic crystal-field state during the lifetime of the 91-keV state as did parent Nd^{3+}. They found $G_2(91)_{\text{CMN}}\geq 0.79$ and $G_2(91)_{\text{NES}}\geq 0.87$. The former value is, as Daniels and Misra pointed out, in strong disagreement with earlier experimental results.[1-4] Our new quantitative values for $G_2(91)$ strengthen this conclusion. It now appears, on the basis of these results and others, that there is strong attenuation in intermediate nuclear states with lifetimes of a few nanoseconds, following both β^- and electron-capture (EC) decay. This was found to be the case for the 61-keV, 2.6-nsec state of Pm^{145} following the decay of Sm^{145} oriented in CMN,[12] and for the 396-keV, 3.3-nsec state of Lu^{175} following the decay of Yb^{175} oriented in CMN.[16,17] Following the decay of Eu^{155} oriented in NES, the 86.5-keV, 6.7-nsec state showed attenuation, while the 105.3-keV, 1.0-nsec state showed little, if any.[18] Strohm and Sapp[19] found no appreciable attenuation in the 145-keV, 1.9-nsec state of Pr^{141} following the decay of Ce^{141} oriented in CMN, but did report attenuation in the 136-keV, 8.9-nsec state of Fe^{57} following the decay of Co^{57} oriented in CMN. Stone has found substantial attenuation for this state in the fluorosilicate lattice,[20] but not in the iron lattice. In

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TABLE III. Summary of reorientation results for oriented nuclei.

Parent	Daughter	Lattice	E_γ	T (°K)	Decay	Electron config. of parent	$T_{1/2}$ (nsec)	G_2	Ref.
^{177}Lu	^{177}Hf	NES	many	0.011	β^-	$4f^{14}$	10^9	~ 1	13
^{57}Co	^{57}Fe	Fe	14.4	0.05	EC	···	100	~ 1	21
^{57}Co	^{57}Fe	Fe	136.4	0.02	EC	···	8.9	~ 1	20
^{57}Co	^{57}Fe	CMN	136.4	0.007	EC	$3d^7$	8.9	0.81(10)	19
^{57}Co	^{57}Fe	FS[a]	136.4	0.02	EC	$3d^7$	8.9	0.32(2)	20
^{155}Eu	^{155}Gd	NES	86.5	0.011	β^-	$4f^6$	6.7	0.26(8)	18
^{155}Eu	^{155}Gd	NES	105.3	0.011	β^-	$4f^6$	1.0	0.80(9)	18
^{154}Sm	^{145}Pm	NES	61	0.011	EC	$4f^5$	2.6	B[b]	12
^{145}Sm	^{145}Pm	CMN	61	0.003	EC	$4f^5$	2.6	$0.44(10)B$	12
^{175}Yb	^{175}Lu	NES	396	0.02	β^-	$4f^{13}$	3.3	C	22
^{175}Yb	^{175}Lu	CMN	396	0.003	β^-	$4f^{13}$	3.3	$<\frac{1}{6}C$[b]	16, 18
^{147}Nd	^{147}Pm	NES	91	0.011	β^-	$4f^3$	2.5	0.89(11)	this work
^{147}Nd	^{147}Pm	CMN	91	0.002	β^-	$4f^3$	2.5	0.42(12)	this work
^{141}Ce	^{141}Pr	CMN	142	0.007	β^-	$4f^1$	1.9	1.08(8)	19

[a] Dilute nickel fluorosilicate.
[b] Only ratios of $G_2(CMN)/G_2(NES)$ obtained.

fact, there is no evidence for attenuation even for the 14.4-keV, 100-nsec state of Fe^{57} following the decay of Co^{57} oriented in iron.[21] These latter observations are expected to reflect the situation in all ferromagnetic metals, where the conduction electrons provide a very good reducing medium and the electronic state comes to equilibrium in very short times. For these cases spin memory should be lost only by spin-lattice relaxation, i.e., in times of the order of seconds. Another result that appears at first to be contradictory is the observation by Blok and Shirley[13] of essentially complete retention of spin memory in a 1-sec isomer of Hf^{177} following the decay of oriented Lu^{177}. This case was exceptional, however, in that both Lu^{3+} and Hf^{4+} have no open electronic shells, making relaxation by magnetic hyperfine interaction unlikely. The available data on G_2 are set out in Table III.[22]

The experimental information regarding reorientation following the decay of oriented nuclei is sparse, but consistent. The picture that is emerging may tentatively be summarized as follows.

(a) In ferromagnetic metals and for closed-shell ions in dielectrics reorientation times are very long, of the order of seconds at 10^{-2} °K. However, no experimental results are available for the latter case with EC decay.

(b) For states with $T_{1/2}=1$ nsec or less, there is no evidence for reorientation even in paramagnetic ions in dielectrics.

(c) For states with $T_{1/2}>1$ nsec, G_2 may or may not be significantly less than unity. Both lifetime (as in the Gd^{155} states) and environment (NES versus CMN for Pm^{145}, Pm^{147}, and Lu^{175}) appear to affect G_2.

(d) As yet there is no demonstrated qualitative difference between the β^- and EC cases.

In summary, the experimental situation is better characterized now than it was when Daniels and Misra made their survey, and more theoretical work on this problem seems justified. Time-dependent processes in the nanosecond region are strongly, but indirectly, suggested by (b) and (c) above. There is still no conclusive evidence that G_2 can be reduced below the static hard core value, and time-differential measurements of $G_2(t)$ are highly desirable.

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[22] M. A. Grace, C. E. Johnson, R. G. Scurlock, and R. T. Taylor, Phil. Mag. **3**, 456 (1968).