UCRL -9747

UNIVERSITY OF CALIFORNIA

Ernest O. Lawrence Radiation Laboratory

TWO-WEEK LOAN COPY

This is a Library Circulating Copy which may be borrowed for two weeks. For a personal retention copy, call Tech. Info. Division, Ext. 5545

BERKELEY, CALIFORNIA

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

UCRL-9747

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

NUCLEAR ORIENTATION OF SOME RARE-EARTH ISOTOPES

Carolyn Ann Lovejoy
(Ph.D. Thesis)

June 1961

Printed for the U. S. Atomic Energy Commission

Printed in USA. Price $1.25. Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D.C.

NUCLEAR ORIENTATION OF SOME RARE-EARTH ISOTOPES

Contents

Abstract 3
I. Introduction 4
II. Theoretical Background. 4
III. Experimental Method
A. Chemical Procedure. 6
B. Apparatus 7
IV. Corrections
A. Counting Corrections. 14
B. Temperature Scale 16
V. Isotopes Showing no Gamma-Ray Anisotropies at Low Temperatures
A. Introduction. 18
B. Terbium-161 18
C. Holmium-160 19
D. Samarium-153. 20
VI. Promethium-143
A. Introduction. 21
B. Results 21
C. Discussion. 25
VII. Terbium-156
A. Introduction. 33
B. Results 33
C. Discussion. 36
1. The Spin of the 2042-kev Level. 36
2. The Spin of the 1620-kev Level. 38
3. Existence of the 1935-kev Transition. 39
4. The Other Unknown Quantities 39
5. Nuclear Moments 43
Acknowledgements 50
References 51

NUCLEAR ORIENTATION OF SOME RARE-EARTH ISOTOPES

Carolyn Ann Lovejoy
(Ph D Thesis)

Lawrence Radiation Laboratory
University of California
Berkeley, California

June 1961

ABSTRACT

Nuclear orientation experiments were performed on five isotopes, Pm^{143}, Tb^{156}, Tb^{161}, Ho^{160}, and Sm^{153}, in crystals of $Nd(C_2H_5SO_4)_3 \cdot 9H_2O$.

The angular distribution of the 740-kev γ ray of Nd^{143} was found to be $W(\theta) = 1 - (0.060 \pm 0.006)\, P_2(\cos\theta)$ at $0.02^\circ K$. Values for the mixing ratio, δ, of the 740-kev γ ray of Nd^{143} were obtained as a function of the magnetic moment of the ground state of Pm^{143} for ground state spins of 5/2 and 7/2. The spin of the excited state of Nd^{143} was assigned as 9/2-. An absolute lower limit of $|\mu| > 1.0$ was set on the magnetic moment of Pm^{143}. The mixing ratio of the 740-kev γ ray was found to lie in the range $0.23 \leq (E2/M1) \leq 0.35$ if the ground state spin is 7/2 and in the range $0.255 \geq (E2/M1) \geq 0.217$ if the ground state spin is 5/2.

The spin of the 2042-kev level in Gd^{156} populated by the decay of Tb^{156} was assigned as 4 and the spin of the 1620-kev level was assigned as 5+. The electron capture decay to both the 2042-kev level and the 1931-kev level was found to be mainly L=1. If the ground-state spin of Tb^{156} is 3 the nuclear magnetic moment was found to be $\mu = 1.45 \pm 0.06$ and the nuclear quadrupole moment, $Q = 1.40 \pm 0.16$. Mixing ratios of the γ rays were determined. The angular distribution of the 1417-kev γ ray was found to be $W(\theta) = 1 - (0.206 \pm 0.016)\, P_2(\cos\theta)$ at $1/t = 58.5$.

Explanations are advanced for the failure of Tb^{161}, Ho^{160} and Sm^{153} to show a noticeable anisotropy at temperatures of around $0.02^\circ K$.

I. INTRODUCTION

A knowledge of nuclear moments and spins is essential in order to understand nuclear structure. One method of obtaining these is by nuclear orientation. The theoretical expression for the amount of orientation involves the nuclear moments and ground-state spin of the parent nucleus as well as the spins of one or more levels of the daughter nucleus.

The nuclear orientation method has been applied here to the study of several isotopes of the rare earths. Both Pm^{143} and Tb^{156} were successfully oriented, and some conclusions have been drawn about the spins and angular momenta of the transitions involved in these decay schemes. Attempts were made to orient Tb^{161}, Ho^{160}, and Sm^{153} but without noticeable effects. Some possible explanations are advanced for the lack of positive results.

II. THEORETICAL BACKGROUND

A nucleus is said to be oriented if its spin has a preferred direction with respect to some axis in space. If the populations $W(I_z)$ of the magnetic substates I_z of the nuclear spin are such that $\sum_{I_z} I_z W(I_z) \neq 0$, then the nucleus is said to be polarized. If they are such that $\sum_{I_z} \left(3I_z^2 - I(I+1)\right) W(I_z) \neq 0$, then the nucleus is said to be aligned.

Although nuclear orientation may be achieved by dynamic methods, studies of nuclear structure have generally utilized static ones. These latter methods depend on making the energy separation between nuclear-magnetic substates of the order of kT.

Five ways of getting static orientation have been proposed.[1,1a] In the first method the nuclei are polarized by applying a large external magnetic field that acts directly on the nuclear magnetic moment. In the second method an external magnetic field is used to line up the atomic magnetic fields caused by the unpaired electrons of a paramagnetic ion; these atomic fields then act on the magnetic moment of their nuclei to produce a polarization. The third method proposes the use of crystalline electric fields to orient the atomic magnetic fields. Since electric fields do not split the $\pm m_j$ levels of the electrons, only an alignment is produced. In the fourth method the strong electric field gradients

set up by covalent bonds are used to line up the nuclear quadrupole moments. In the fifth method, an alloy of iron with a diamagnetic element is magnetized to saturation by a small external magnetic field;[1a] the strong internal magnetic field set up in the alloy then polarizes the nuclei of the diamagnetic element.

Although the first method is applicable to the most nuclei, experimentally it is very difficult to get it to work. The second and third methods are particularly useful when working with the rare earths. The Hamiltonian for paramagnetic rare-earth ions in a rare-earth ethylsulfate with the applied field along the z axis is

$$\mathcal{H} = g_{\parallel}\beta H_z S_z + A S_z I_z + \Delta_x S_x + \Delta_y S_y + P[I_z^2 - 1/3\, I(I+1)] + c\, S_z(S_{1z} + S_{2z}).$$

The c term refers to the interaction between the ion and its nearest neighbor rare-earth ions. It is applicable in neodymium ethylsulfate but not in yttrium ethylsulfate. The P term is negligible for Pm but not for Tb. The S is the effective electronic spin of the lowest doublet.

Although the spatial distribution of radiation from a single magnetic substate of a radioactive nucleus may be anisotropic, if the populations of all the substates are equal, the total distribution of radiation will be isotropic. If the populations are unequal, the distribution may be anisotropic.

The distribution of radiation can be written as:[1]

$$W(\Theta) = \sum_{\nu} B_{\nu}(T)\, U_{\nu}\, F_{\nu}\, P_{\nu}\, (\cos\Theta).$$

Here Θ is the angle between the z axis and the direction of observation; the B_{ν} are orientation parameters; U_{ν} is a function of the multipolarities of the radiations and the spins involved in the unobserved preceding transitions. F_{ν} depends on the multipolarities of the observed radiation and the initial and final spins of the observed transition. P_{ν} is the Legendre polynomial of degree ν. Odd-ν terms drop out for γ radiation. Since we have $\nu \leq$ the smaller of $2I_{initial}$ or $2L_{observed}$, it is usually limited to 2 or 4.

III. EXPERIMENTAL METHOD

A. Chemical Procedure

The radioactive rare earths used in these experiments were produced by bombardment of stable rare-earth oxides. After the bombardment the rare-earth oxides were dissolved in a small amount of aqua regia. Ammonium hydroxide was added till the rare-earth hydroxides formed.[2] These were centrifuged and washed several times, then dissolved in HCl. The acidity of the solution was adjusted to pH 1 and it was placed on a heated Dowex-50 100-200 mesh ion-exchange column. The rare earths were eluted from the column by using 0.4 M α-hydroxy-isobutyric acid buffered to about pH 4.[3] The α-hydroxy solution containing the activity was acidified to pH 1 and was run through a small unheated Dowex-50 ion-exchange column. The rare earth remained on the column. After thorough washing with 0.1 M HCl, the activity was stripped off with 8 M HCl. This solution was evaporated to dryness. The residue was then dissolved in a drop or two of rare-earth ethylsulfate solution. A seed crystal of the appropriate rare-earth ethylsulfate was added, and more solution was added whenever needed to keep the crystal covered.

Two types of crystals were used in the experiments, neodymium ethylsulfate and yttrium ethylsulfate. The rare-earth ethylsulfates were obtained by adding the desired rare-earth sulfate to barium ethylsulfate. After barium sulfate was filtered off the solution was allowed to evaporate. The rare-earth ethylsulfate was purified by several recrystallizations. Since the rare-earth ethylsulfates form an isomorphous series of crystals, it is possible to grow small amounts of any rare earth into the ethylsulfate of any other rare earth.

Yttrium ethylsulfate was used in these experiments because yttrium is not paramagnetic. In neodymium ethylsulfate the radioactive rare-earth ion experiences a magnetic field due to the magnetic moments of its two nearest neighbor neodymium ions. This field may be opposed or parallel to its own magnetic moment. In yttrium ethylsulfate this splitting of the ground state in the absence of an external field does not occur. Since yttrium ethylsulfate does not cool by adiabatic demagnetization, it has to be cooled by contact with neodymium ethylsulfate, grown on the outside of an yttrium ethylsulfate crystal. The activity was grown into a small

yttrium ethylsulfate crystal; then an inactive layer of the yttrium ethylsulfate was grown on the outside, so that none of the activity could get into the neodymium ethylsulfate. The crystal was afterward placed in a mixed solution of neodymium and yttrium ethylsulfate, and the percentage of neodymium ethylsulfate in the solution covering it was gradually increased. Finally, the crystal was placed in pure neodymium ethylsulfate and a layer of this was grown on the crystal. The gradual transition from yttrium ethylsulfate to neodymium ethylsulfate was necessary for good thermal contact between the inner yttrium ethylsulfate crystal and the outer shell of neodymium ethylsulfate.

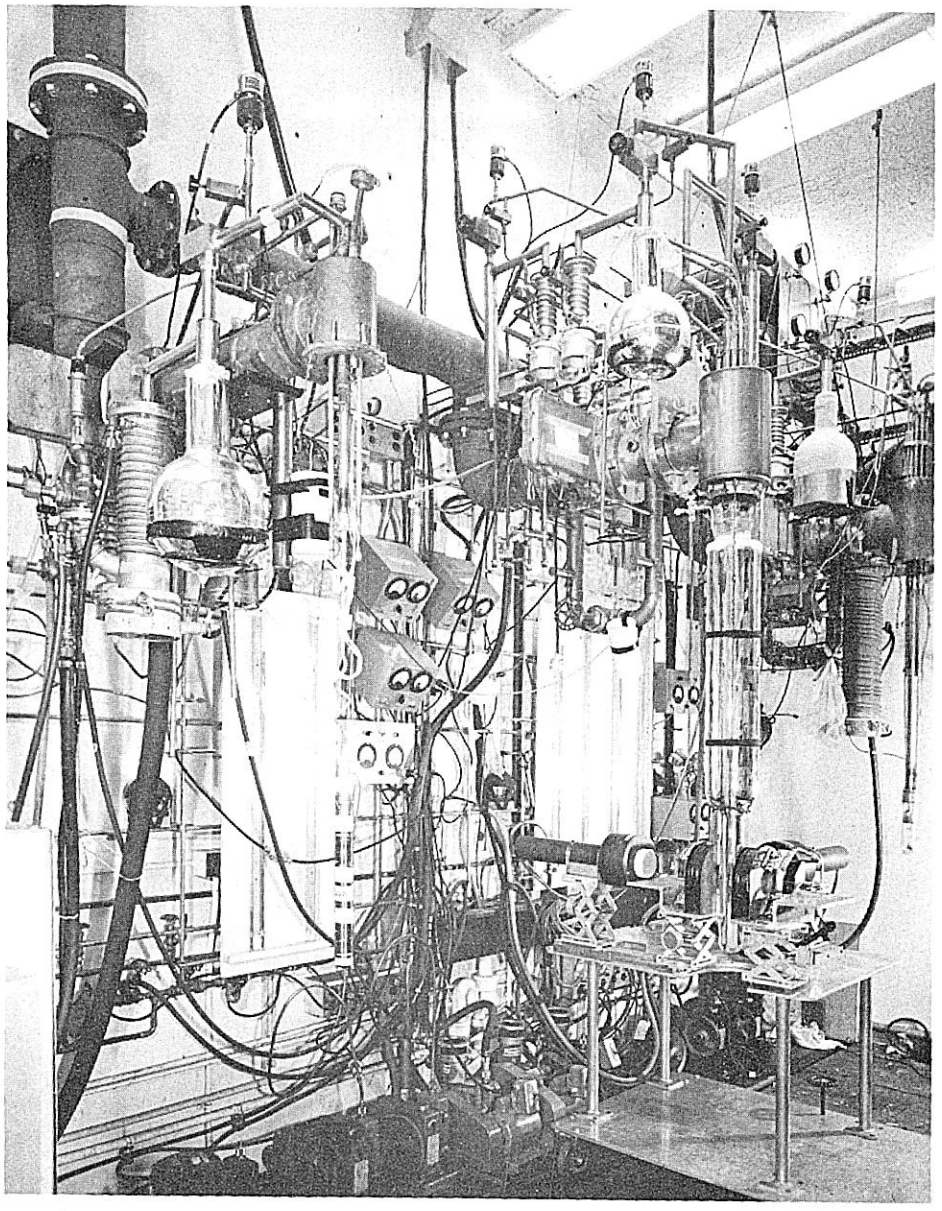
B. Apparatus

Two demagnetization cryostats were used in these experiments. They are quite similar, and the following description applies to both.[4] A large outer Dewar vessel (Fig. 1) contains liquid nitrogen at 77°K, open to the air. Inside this is a Dewar containing liquid helium. By pumping on the helium its temperature is lowered to about 1°K. Inside this Dewar is the vacuum jacket containing the crystal (Fig. 2). The vacuum jacket is soft-soldered to a brass cap. This cap is attached to and supported by the tubing through which helium gas is pumped. The gas is used for a thermal contact between the crystal and the liquid helium. The crystal is mounted in a glass holder (Fig. 3) sealed to a tungsten wire that is hard-soldered to the brass cap.

The magnet (Fig. 4) used for the adiabatic demagnetizations rolls along tracks in the floor. It can be raised and lowered and its pole gap can be changed. The iron-free polarization magnet (Fig. 5) is on a wheeled table that can be rolled into position after demagnetization and locked there.

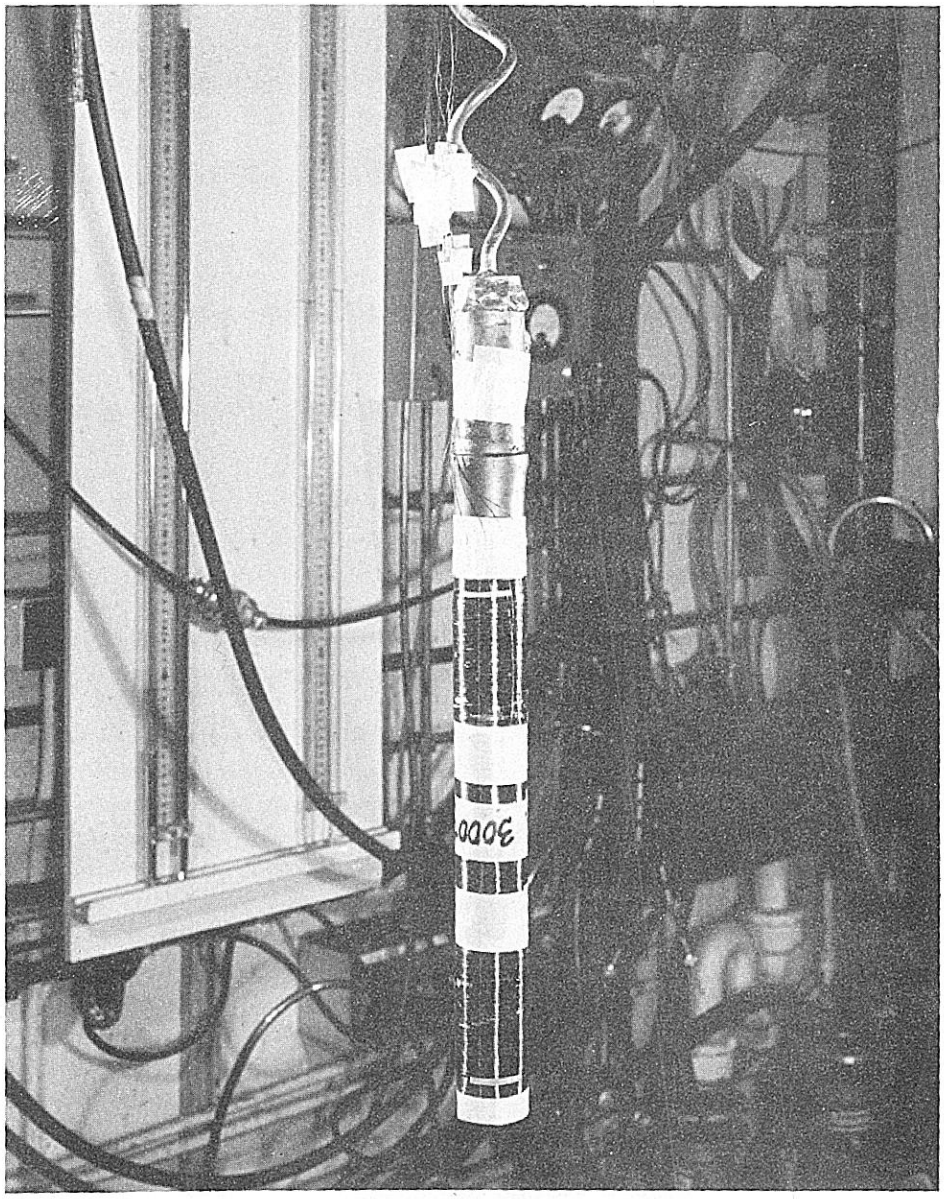
The magnetic temperature of the crystal was measured by an ac mutual-inductance bridge (Fig. 6). Meyer's data were used to convert magnetic temperature to absolute temperature.[5]

In order to get the temperature dependence of the anisotropy, demagnetizations were made from fields of different initial strengths. This method ensured that all parts of the crystal were at the same temperature.



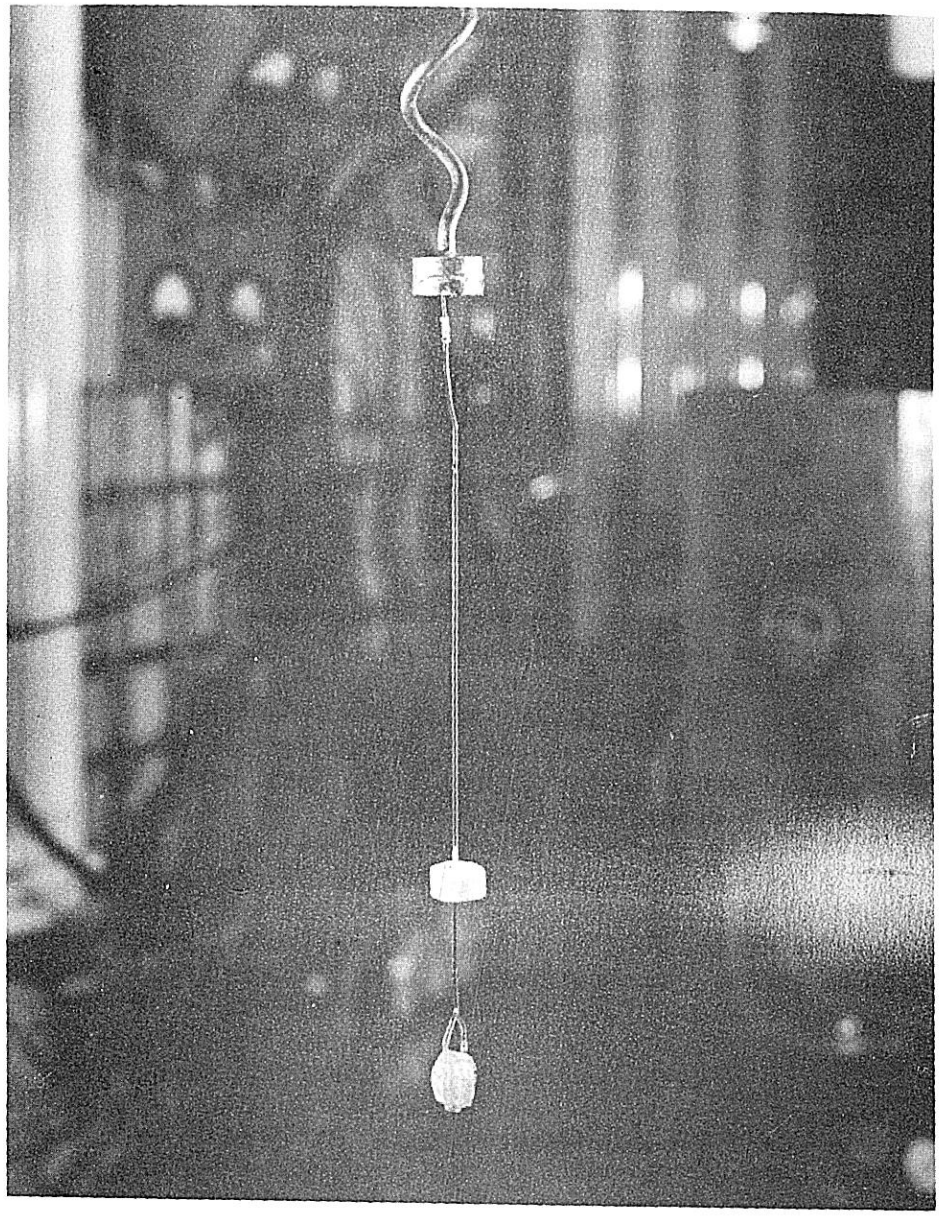
ZN-2799

Fig. 1. Cryostat with counters and polarization magnet around it. In the foreground is a vacuum jacket with mutual-inductance coils on it.



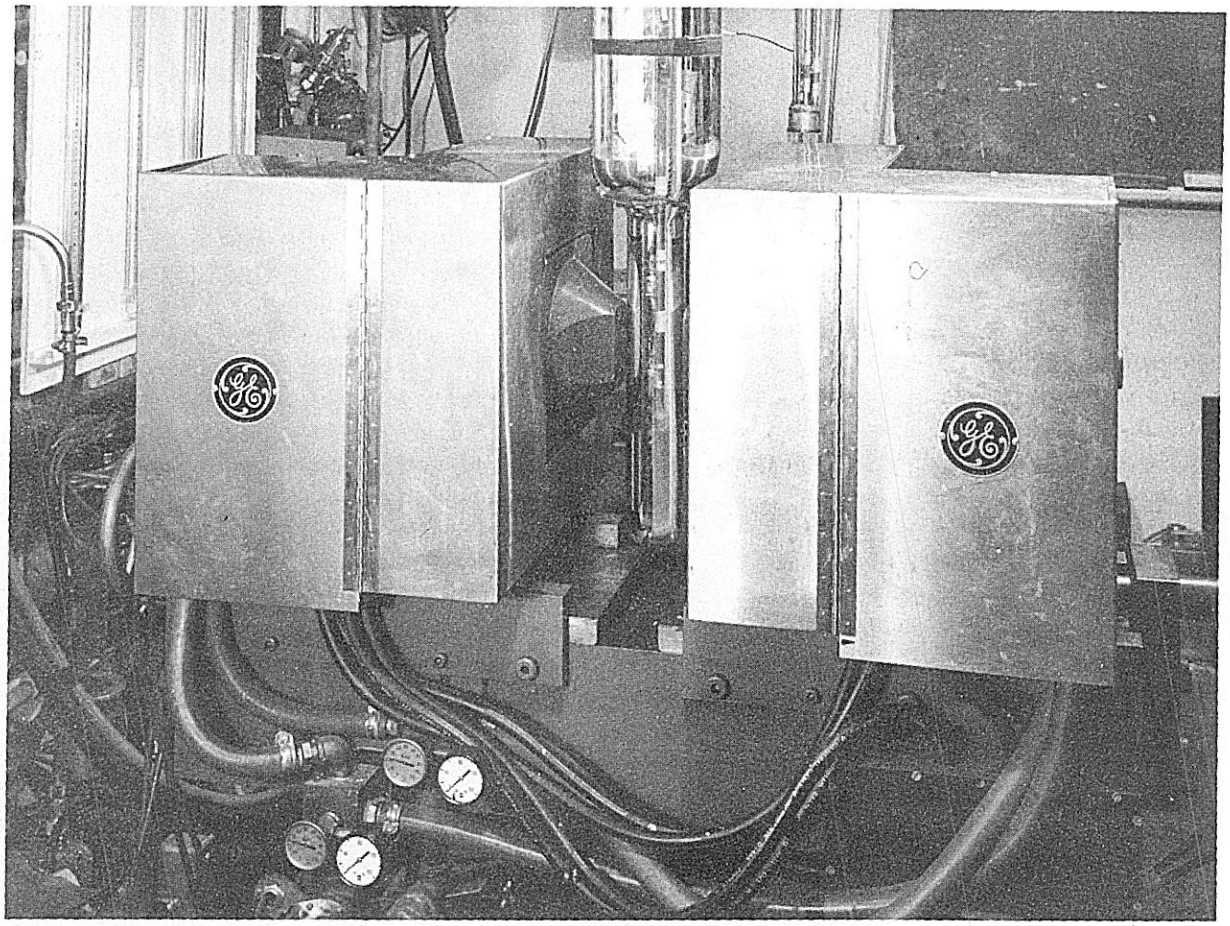
ZN-2800

Fig. 2. Vacuum jacket with coils for measuring mutual inductance on it.



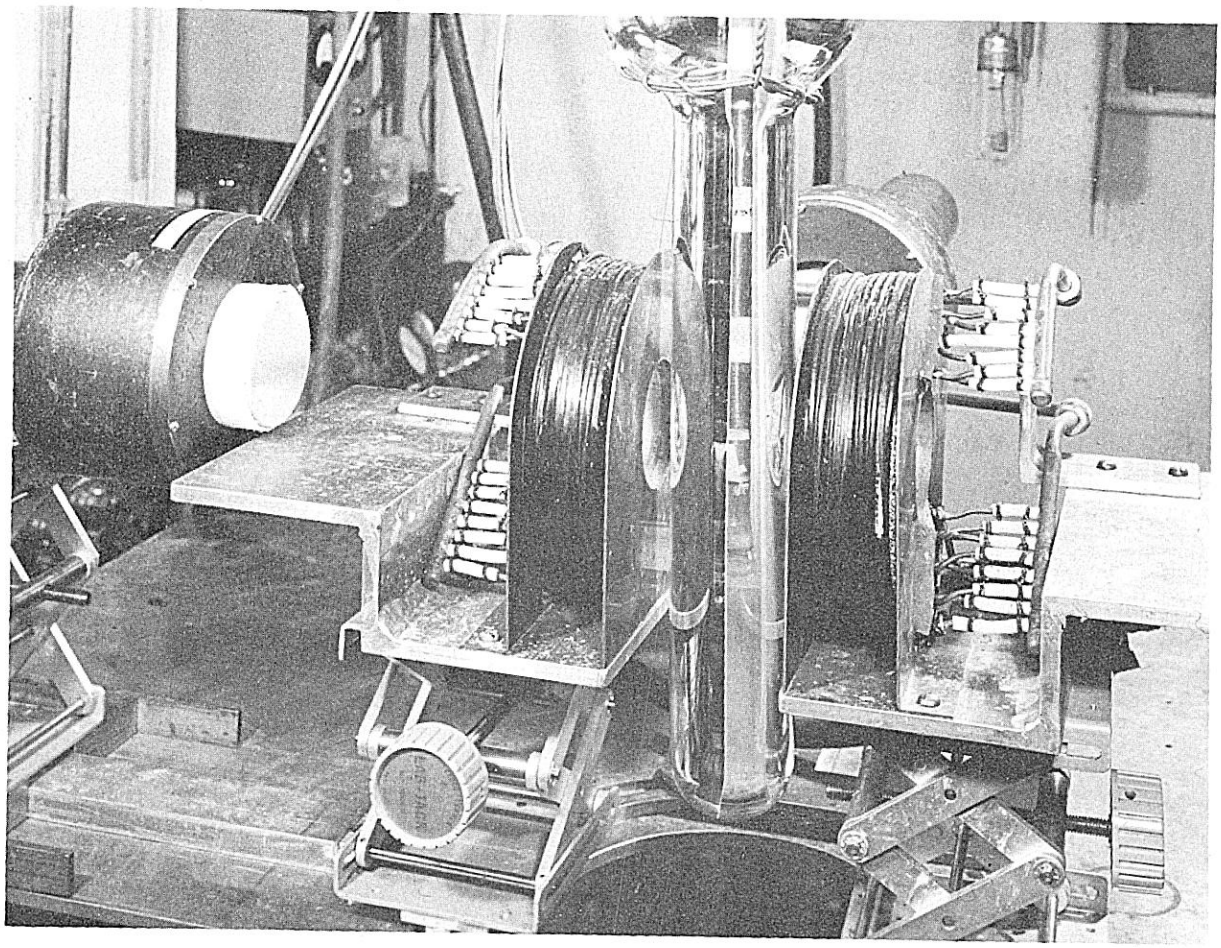
ZN-2801

Fig. 3. $Nd(C_2H_5SO_4)_3 \cdot 9H_2O$ incorporating a radioactive rare-earth isotope, mounted in a glass holder with a manganous ammonium sulfate pill mounted above it.



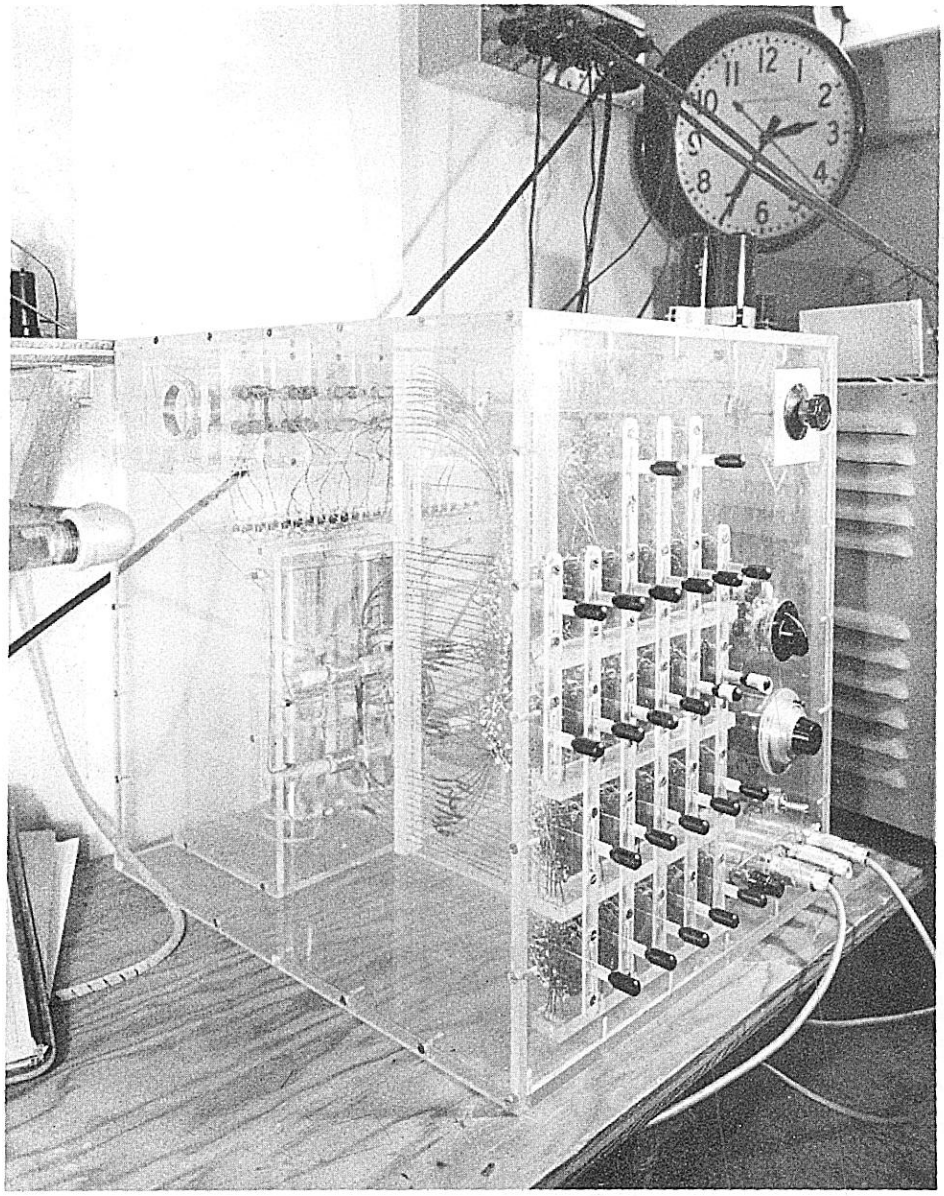
ZN-2803

Fig. 4. The 18-kg magnet in place about the cryostat.



ZN-2802

Fig. 5. Air-core polarization magnet and NaI(Tl) scintillation crystals in place around the cryostat. The crystals have magnetic shielding around them.



ZN-2797

Fig. 6. The ac mutual-inductance bridge.

Counting (Fig. 7) was done by 3x3-in. cylindrical NaI(Tl) crystals. The signals from these were fed into 100-channel differential pulse-height analyzers. The crystals, with their photomultipliers and preamplifiers, were mounted in soft-iron jackets with mu-metal shields. These assemblies were placed on the wheeled table and rolled into position after each demagnetization.

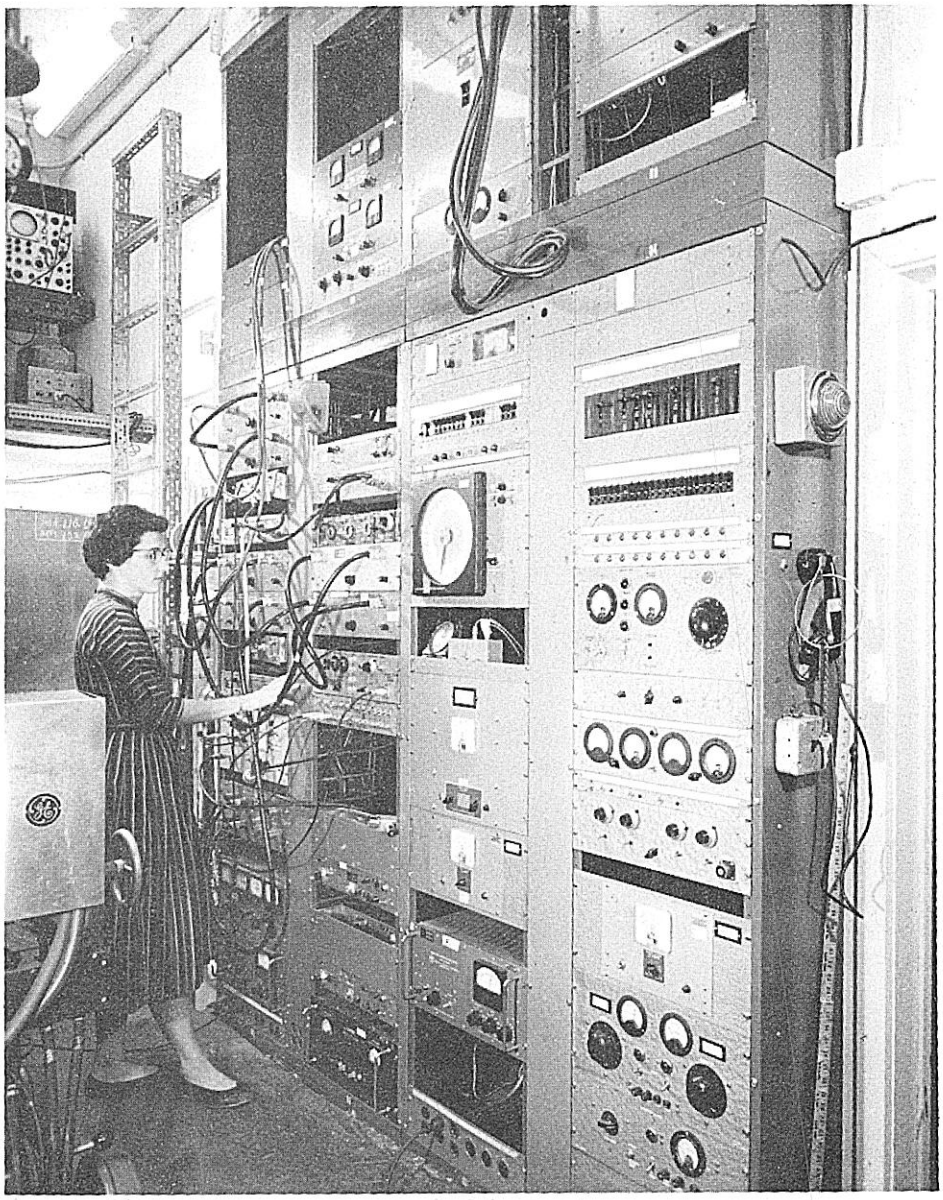
IV. CORRECTIONS

A. Counting Corrections

In order to get $B_2U_2F_2$ for a γ ray in a particular experiment, several corrections must be made to the observed ratio of cold counting rate to warm counting rate (c/w).

In some cases, the counters' gains drift between the cold and warm counts. The total area under the peaks is still the same as it would have been if the drift had not occurred, but the number of counts within a given number of channels is different. Since the c/w ratio is obtained by using the same few channels near the peak in both spectra, this effect gives a false c/w. Even if a peak were really isotropic, it would appear to have more counts in the spectrum in which it was centered in a lower channel because the spectrum would be compressed along the abcissa. To correct for this effect, one takes the product of two factors. The first is the ratio of the apparent shift of the peak to the energy of the peak. Usually the shift is no more than one-quarter channel. The second factor is the ratio of the number of counts in the channel immediately above or below the channels included in the peak to the average number of counts per channel in these peak channels. Since one tries to choose the peak channels so that the first and the last ones have approximately equal numbers of counts, the two adjacent channels--one above, the other below the peak channels--should also have approximately equal numbers of counts in them. Therefore, either could be used in the correction. This product of the two factors is added to unity to give the correction factor. The correction factor multiplies the counting rate in the spectrum in which the peak appears in a higher channel:

$$\left(\frac{\Delta E}{E}\right)\left(\frac{\text{counts (adj)}}{\text{counts (av)}}\right) + 1.$$



ZN-2798

Fig. 7. Electronic racks for counters, pumps and magnets.

The change in dead time between cold and warm counts makes the difference in counting rate appear less than it is. To obtain a first-order correction for this effect, one finds the change in dead time with counting rate, and then multiplies this by the change in counting rate between the entire cold and warm spectra. The correction factor for this effect is the resultant number plus one:

$$\left[\left(\frac{\Delta\ D\ T}{\Delta\ C\ R}\right)\left|CR_w - CR_c\right|\right] + 1.$$

The finite-detector-size correction depends on the detector's size and its distance from the source. Graphs of these corrections have been made.[6] This correction acts to increase the size of the effect.

The most important correction is that for background. In a complex spectrum it cannot be assumed that the Compton background from other peaks is isotropic, so this background must be subtracted separately for cold and warm counts. This complete resolution of the spectra needs to be done only once because, under the same experimental conditions and when only the B_2 term is important in the orientation, the anisotropy of the background is related to the anisotropy of the peak by a constant factor.

B. Temperature Scale

The magnetic temperature of the paramagnetic salt, neodymium ethylsulfate, was determined by magnetic-susceptibility measurements using an ac mutual inductance bridge. The susceptibility χ is related to the magnetic temperature T^* by the equation $\chi = C/(T^* + K)$. The term K is small and may be ignored at temperatures above 1^oK. The C is determined by taking readings of the susceptibility while cooling the sample from 4^oK to 1^oK by pumping on the helium bath. The K is determined by setting $T^*(H_i/T_i) + K = T^*_{HM}(H_i/T_i)$. $T^*(H_i/T_i)$ is the magnetic temperature reached experimentally when demagnetizing from the initial value, (H_i/T_i), of magnetic field over absolute temperature. $T^*_{HM}(H_i/T_i)$ is the magnetic temperature which Meyer[5] showed a single spherical crystal of neodymium ethylsulfate would reach when demagnetizing from (H_i/T_i). To get highest accuracy, the data from the lowest temperature were used to determine K.

The average value of χ during the counting period was used to compute $1/T^*$. Then $1/T^*_{HM}$ was calculated by using K. Meyer's data were used to convert $1/T^*_{HM}$ to $1/T$ (here T is the absolute temperature).

If the measurement of susceptibility is made in a field, as is done in polarization experiments, it is necessary to convert $1/T^*(H)$ to $1/T^*(H=0)$ before making the correction for K. The correction is derived as follows: where H is the polarizing field, h is the measuring field, $g_{\parallel}$ is the atomic g factor parallel to the polarizing field along the z axis, $g_{\perp}$ is the atomic g factor perpendicular to the polarizing field and parallel to the measuring field along the x axis, β is the Bohr magneton, S_z and S_x are the components of the effective electron spin along the z and x axes respectively, W is the energy of the atomic levels, and N is the total number of atoms; then the Hamiltonian is

$$\mathcal{H} = g_{\parallel}\beta HS_z + g_{\perp}\beta hS_x .$$

Then we have

$$W = \pm (1/2)\left((g_{\parallel}\beta H)^2 + (g_{\perp}\beta h)^2\right)^{1/2},$$

the partition function is

$$Z = \Sigma\, e^{-W/kT} = 2 \cosh (W/kT),$$

and the free energy is $F = -NkT \ln Z$. The perpendicular magnetization is expressed by

$$M_{\perp} = -(\partial F/\partial h)_T = (NkT/Z)\ (\partial Z/\partial h)_T \approx \left[(g_{\perp}{}^2\beta^2 h)/(2kTg_{\parallel}\beta H)\right]$$
$$\times\ NkT \sinh (|W|/kT)/\cosh (|W|/kT) = \left[(Ng_{\perp}{}^2\beta^2 h)/(4kT)\right]$$
$$\times\ \left(\tanh(W=kT)\right)/(W=kT).$$

The susceptibility χ is expressed as

$$\chi = \lim_{h\to 0} M_{\perp}/h = (\tanh x)/x\ (Ng_{\perp}{}^2\beta^2/4kT),$$

where $x = g_{\parallel}\beta H/2kT$.

If there is no applied field, we take $\mathcal{H} = g_\perp \beta h S_x$; so $W = \pm(1/2)\, g_\perp \beta h$, and $Z = 2 \cosh (g_\perp \beta h/2kT)$. Then we have

$$M_\perp = (NkTg_\perp \beta/2kT) \tanh (g_\perp \beta h/2kT),$$

and if we obtain $g_\perp \beta h/2kT \ll 1$, then $M \approx (Ng_\perp{}^2\beta^2 h)/4kT$, so $\chi_{H=0} = (Ng_\perp{}^2\beta^2/4kT)$. Then

$$\chi_H/\chi_{H=0} = (\tanh x)/x = (1/T^*_H)/(1/T^*_{H=0}) = (T^*_{H=0})/(T^*_H).$$

IV. ISOTOPES SHOWING NO GAMMA-RAY ANISOTROPIES AT LOW TEMPERATURES

A. Introduction

Nuclear orientation experiments were performed on several isotopes that proved not to have statistically significant γ-ray anisotropies. That is, any anisotropies seen were small enough to fall within the statistical limits of error. At least two experiments were done with each isotope, and during each experiment several demagnetizations were made. These experiments were all done in neodymium ethylsulfate at temperatures down to 0.02°K. Although the anisotropies cannot definitely be said to be zero, some upper limit can be placed on them.

B. Terbium-161

Tb^{161} was produced by irradiating Gd^{160} with thermal neutrons. The 3-min Gd^{161} decays to 7-day Tb^{161}. Neither the 74.5-kev transition nor the 106-kev transition showed any anisotropy. An upper limit of 2% may be set on both $B_2U_2F_2$'s. The 74.5-kev transition goes from a 3/2- state to a 5/2- state. The 106-kev transition goes to a 5/2- state from a state that is probably 5/2-. The ground state spin of Tb^{161} is 3/2+ by analogy with Tb^{159}.

Nuclear systematics indicate that the magnetic moment of Tb^{161} should be about equal to that of Tb^{159}. A magnetic moment of this size would give a B_2 of about 0.42, at a temperature of 0.02°K. Therefore U_2F_2 would have to be less than 0.048.

The U_2F_2 of the 106-kev γ ray would be less than this value if the β decay to this level carried off one unit of angular momentum, and if the 106-kev transition were between 9% and 17% quadrupole or between 95% and 99% quadrupole.

Conversion coefficient data indicate that the 84.5-kev γ ray is primarily dipole. If no angular momentum were carried away in the β decay, the U_2F_2 of a pure dipole transition would be 0.10. If one unit of angular momentum were carried away in the β decay, then U_2F_2 of a pure dipole transition would be 0.02. In the first case, there would have to be less than 1.5% of quadrupole admixture if U_2F_2 were to be less than 0.048. In the second case, there would have to be less than 11% of quadrupole admixture.

The half life of the 74.5-kev state is 2.3×10^{-9} sec. This is long enough for an appreciable fraction of the nuclei to reorient, causing an attenuation of the expected anisotropy. The half life of the 132-kev state, the initial state for the 106-kev transition, is not known.

C. Holmium-160

The ${}^{93}_{67}Ho^{160}$ was produced by the reaction $Tb^{159}(\alpha,3n)Ho^{160}$. There is a 5-hr isomer of Ho^{160} that makes a 60-kev transition to the 28-min ground state. Both states decay to excited states of Dy^{160} by EC and $\beta+$ emission. No anisotropy was found in the γ rays from the excited states. An upper limit of 1% may be set on $B_2U_2F_2$ for all the γ rays investigated. Since a large effect has been seen in some of these γ rays in the decay of Tb^{160}, the U_2's and the F_2's cannot all be small, so either B_2 must be small or there must be attenuation effects.

For at least two reasons, the B_2 could be small or zero: first, the spin of the parent nucleus could be zero; secondly, the nuclear moments are small.

It appears that the spin is not zero. The decays populate states with spins of 3 and 4. The neutron is probably in a 3/2- state. The spins of ${}^{93}_{64}Gd^{157}$, ${}^{95}_{66}Dy^{161}$, and ${}^{91}_{64}Gd^{155}$ are 3/2-. The proton is probably in a 7/2- state, since the spin of ${}^{98}_{67}Ho^{165}$ is 7/2-. The proton and neutron can couple to resultant spins of 5 or 2. The former seems more likely because the decay is to high spin states.

Using the wave functions for a deformed nucleus and the formula for magnetic moments given by Nilsson,[7] one obtains $\mu = 3.9$ n.m. This moment is large enough to produce a noticeable alignment.

After electron capture and Auger processes the resultant atom may be multiply charged, and the electric and magnetic fields at the nucleus may be changing rapidly. If most of the decays went through a high-energy long-lived (10^{-8} sec) state, a large percentage of the nuclei would lose their orientation. Recent work[8] indicates that a large enough percentage of the decays go directly to the 966.4-kev and the 1048-kev states (which are known to be fast) to invalidate this hypothesis. That is, an effect should be seen just from direct decays to these levels.

D. Samarium-153

An upper limit of 1% may be set on the $B_2U_2F_2$ of the 103-kev γ ray of Sm^{153}. The level scheme has been worked out by Sund and Wiedenbeck.[9] The spin of Sm^{153} is 3/2-.[9a] Of the decays, 26% go to the 172-kev level in Eu^{153} and 53% go to the 103-kev level. The 172-kev level (spin 5/2) decays to the 103-kev level (spin 3/2) by a mixed M1 and E2 transition. Several measurements have been made of the conversion coefficients of the 70-kev and the 103-kev transitions. Sund and Wiedenbeck did an angular correlation measurement on the 70-kev— 103-kev cascade. Their results were in agreement with the conversion coefficient data. The multipolarities of 98.2% M1 and 1.8% E2 for the 70-kev γ ray and 98.4% M1 and 1.6% E2 for the 103-kev γ ray were used. The F_2 for the 103-kev γ ray on the basis of this multipolarity is 0.044. About a third of the population of the 103-kev level comes through the 172-kev level. The U_2 for this third is 0.559 if one unit of angular momentum is carried off in the β decay to the 172-kev level, and the U_2 for the 2/3 coming directly from Sm^{153} is 0.20 if the angular momentum carried off in the decay is one, and is 1.0 if no angular momentum is carried off. The total U_2 is then 0.320 in the first case and 0.853 in the second. This gives $U_2F_2(103)$ as 0.014 in the first case and 0.0375 in the second. To be consistent with the present results, in the first case $|B_2(1/T=50)|$ would have to be less than 0.82, and in the second case less than 0.26. The ratio of the B term to the A term in the Hamiltonian has been measured[9b]

in lanthanum ethylsulfate and found to be 4.16. At $1/T = 50$ this would give $B_2 \approx -0.14$. Although this is an approximate calculation, it would seem to explain the absence of an effect in both cases.

VI. PROMETHIUM-143

A. Introduction

Several authors have described the decay of Pm^{143}. The most detailed work was done by Ofer.[10] This investigation included γ-ray spectra, γ-ray— x-ray coincidence spectra, and internal-conversion coefficients. Ofer observed only one γ ray, which had an energy of 740 kev, and he determined that 45% of electron capture went to the excited level, and 55% directly to the spin-7/2- ground state of Nd^{143}.

Recent experiments have shown that promethium nuclei can be oriented in the ethylsulfate lattice.[11] Low-temperature nuclear orientation experiments to study the decay of Pm^{143} have been carried out.

B. Results

The angular distribution of the 740-kev γ ray was found to obey the equation $W(\theta) = 1 - (0.060 \pm 0.006)P_2(\cos\theta)$ at $0.02^\circ K$ (Fig. 8), where θ is the angle between the direction of propagation and the crystalline c axis. The ratio of cold to warm counting rate at $\theta = 0$ deg as a function of reciprocal temperature is shown in Fig. 9 and Table I.

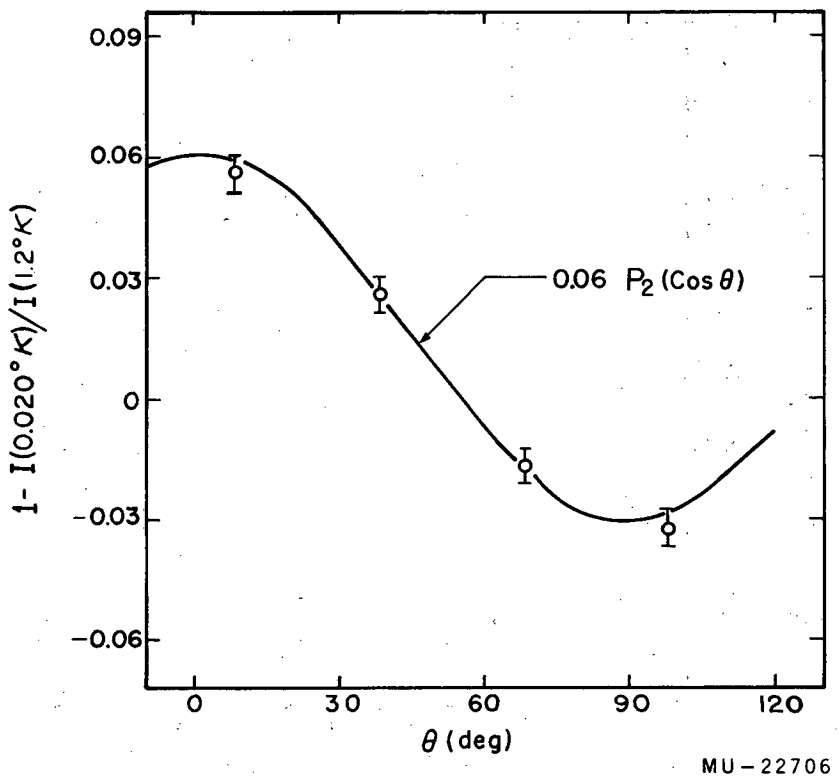


Fig. 8. Dependence of $1 - \left(I(0.020^{\circ}K)/I(1.2^{\circ}K)\right)$ for the 740-kev γ ray of Pm^{143} on angle θ between detector and crystalline axis. The curve is $0.06\ P_2(\cos\theta)$.

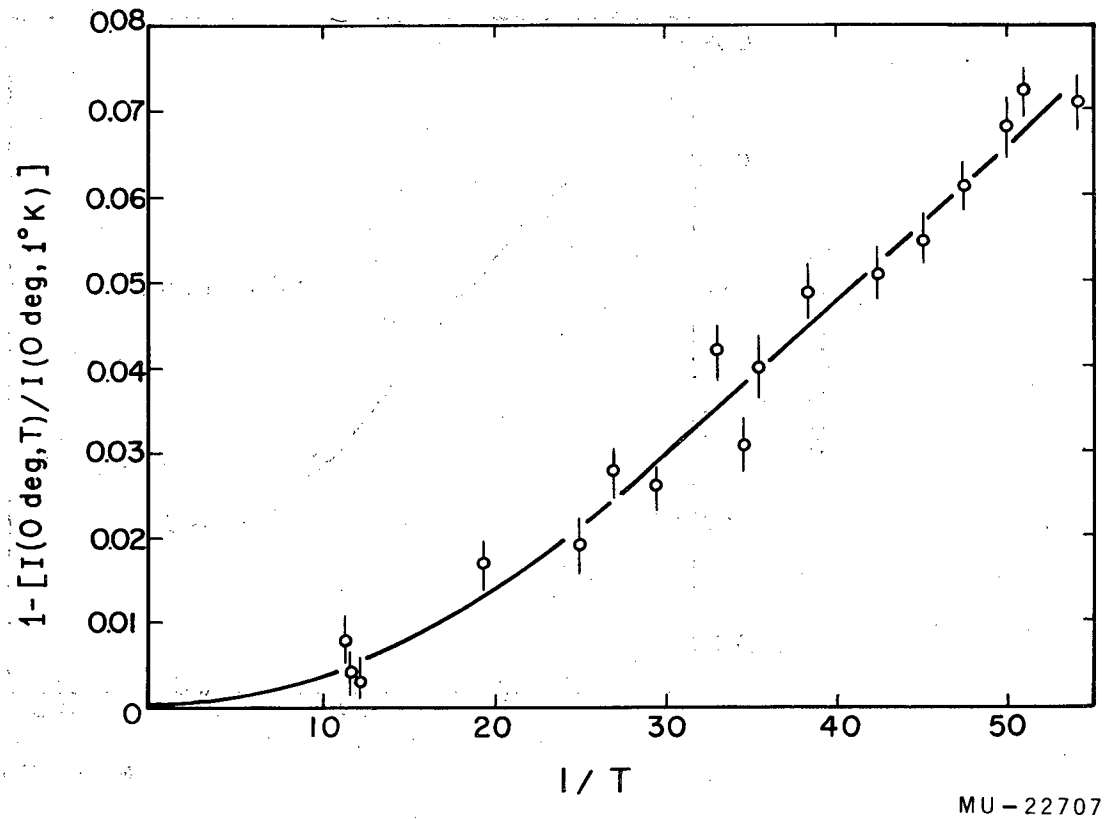


Fig. 9. Observed variation with 1/T of 1-(I(0 deg., T)/I(0 deg., 1°K)) for the 740-kev γ ray of Pm^{143}.

Table I

Temperature and magnetic-field dependence of the anisotropy

H (gauss)	1/T	$1 - \frac{I(0\ deg, T)}{I(0\ deg, 1^{o}K)}$
0	11.4	0.008
0	11.9	0.004
0	12.3	0.003
0	19.4	0.017
0	24.7	0.019
0	27.0	0.028
0	29.6	0.026
0	31.8	0.042
0	34.4	0.031
0	35.6	0.040
0	38.6	0.049
0	42.6	0.051
0	44.8	0.055
0	47.6	0.061
0	50.0	0.068
0	51.0	0.072
0	54.4	0.071

Table I (continued)

Temperature and magnetic-field dependence of the anisotropy

H (gauss)	1/T	$1 - \frac{I(0\ deg,\ T)}{I(0\ deg,\ 1^{o}K)}$
103	20.0	0.014
103	26.4	0.024
103	29.2	0.033
103	32.6	0.046
103	32.6	0.047
200	20.5	0.018
200	23.3	0.015
200	24.6	0.027
200	24.6	0.028
200	25.4	0.029
360	18.9	0.028
370	18.0	0.027
390	19.0	0.020

C. Discussion

The interpretation of nuclear orientation experiments is greatly facilitated by paramagnetic resonance data on atomic energy levels and hyperfine structure. In the absence of such work for promethium, interpolation of crystal-field parameters from neighboring rare-earth elements was used.

The energy levels of the tripositive Pm^{143} in neodymium ethylsulfate may be calculated by using the Hamiltonian:

$$\mathcal{H} = g_{\parallel}\beta H_z S_z + AS_z I_z + \Delta_x S_x + \Delta_y S_y + P[I_z^2 - \tfrac{1}{3} I(I+1)] + cS_z(S_{1z} + S_{2z}).$$

The last term represents dipole-dipole interactions with the two nearest-neighbor Nd^{+3} ions,[12] and the other terms have their usual significance. The term in P can be shown to be negligible in this case for purposes of nuclear alignment.[4] A value of 0.0039 cm^{-1} was used for c, and a value of 0.014 cm^{-1} was used for Δ, where $\Delta^2 = \Delta_x^2 + \Delta_y^2$.

Tripositive promethium is a non-Kramer's ion with the configuration $4f^4$. By Hund's rule, the ground term of the free ion is $^5I_4$, and calculations indicate that in the ethylsulfate lattice the lowest level is a doublet composed of admixtures of the states $|J_z = \pm 4>$ and $|J_z = \mp 2>$.[4] The magnetic hyperfine-structure constant A was calculated to be $(0.019 \pm 0.002)(\mu/I)cm^{-1}$ by use of crystal-field theory.[4]

Experimentally, Ofer found the 740-kev γ ray to be predominantly M1,[10] but experimental uncertainty precludes an accurate determination of how much E2 admixture may be present. The K-conversion coefficient reported is $(6.5\pm1) \times 10^{-3}$. The theoretical value interpolated from the tables of Sliv and Band[13] is 5.5×10^{-3} for an M1 transition and 3.4×10^{-3} for an E2 transition. Thus, the spin and parity of the excited state of Nd^{143} may be 5/2-, 7/2-, or 9/2-.

The spins of Pm^{143} and of the excited state may be inferred from the following evidence: James and Bingham have found M4 isomerism in 81-neutron Sm^{143},[14] which strongly suggests that the ground state of Sm^{143} is $d_{3/2}$ like its isotones, Nd^{141}, Ce^{139}, Ba^{137}, Xe^{135}, and Te^{133}. The log ft ~ 5 for positron decay of Sm^{143} to Pm^{143} indicates allowed decay and precludes a spin change of greater than one. Ofer gives an

estimate of log ft = 8.8 for the decay of Pm^{143} to the 7/2- ground state of Nd^{143}, and log ft = 8.5 for decay to the excited state, indicating first-forbidden decay.

No nuclear γ rays have been observed in the decay of Pr^{143}, and Starfelt and Cederlund set an upper limit of 10^{-3}% on γ rays in the inner bremsstrahlung spectrum ($E_\gamma < 600$ kev).[15] A similar limit may be set on the 740-kev γ ray in this decay,[16] and a lower limit of log ft $\geqslant$ 10.5 may thus be obtained for β branching to the 740-kev state of Nd^{143}. This β decay therefore probably involves a spin change of at least 2. The most likely ground-state assignment of the odd proton in Pr^{143} is 5/2+. Then only spin and parity 9/2- seem admissible for the excited state of Nd^{143}, involving the odd neutron in an $h_{9/2}$ orbit.

Way et al. have suggested that there is a low-lying 5/2+ level in Pm^{143}.[17] This would account very well for the fast β decay from Sm^{143}, with subsequent γ-ray de-excitation to a 7/2+ ground state. These two states are energetically close in other odd promethium isotopes.[17] The only level scheme compatible with all the data is shown in Fig. 10, and one analysis of the results will be based on this scheme. An analysis will also be based on the theoretical work of Kisslinger and Sorensen, which predicts that the ground state of Pm^{143} is 5/2+, with the 7/2+ level an excited state.[18]

On shell-model grounds it seems unlikely that the 740-kev state in 83-neutron Nd^{143} would be other than 9/2-, corresponding to the $h_{9/2}$ orbital. It is not possible to be quite so confident of the β-decay ft-value arguments that the ground state of Pm^{143} is 7/2+ and not 5/2+. The log ft of 8.5 for decay of Pm^{143} to the excited state is high enough that $\Delta I = 2$, yes character is not precluded.

The analysis to follow is based on the assumption that the Pm^{143} spin is 7/2+.

The anisotropy of the radiation as a function of temperature was fitted to the theoretical function $W(\theta) = 1 + B_2(T)U_2F_2P_2(\cos\theta)$.

At this point the interpretation of this work becomes somewhat tentative, because the anisotropy of the 740-kev γ ray depends on the mixing ratio δ(E2/M1) and on the magnetic moment of the ground state of Pm^{143}. In Fig. 11 is shown the functional relationship between δ and $|\mu|$ as derived from the anisotropy data (Table II). Lower limits

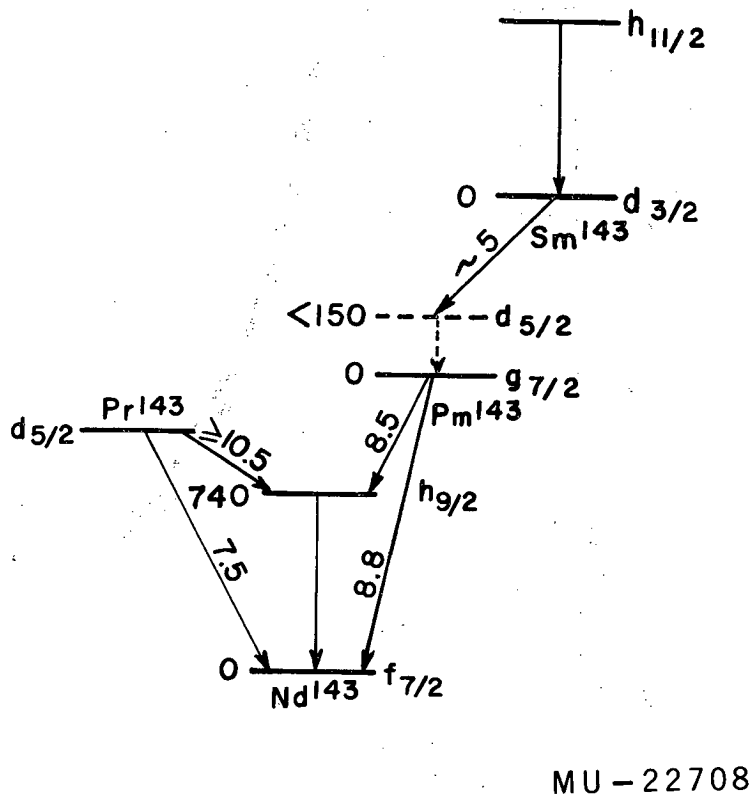


Fig. 10. Proposed level schemes for several nuclei with A=143. Numbers on arrows denote log-ft values. Only indirect evidence is available for the excited state of Pm^{143}.

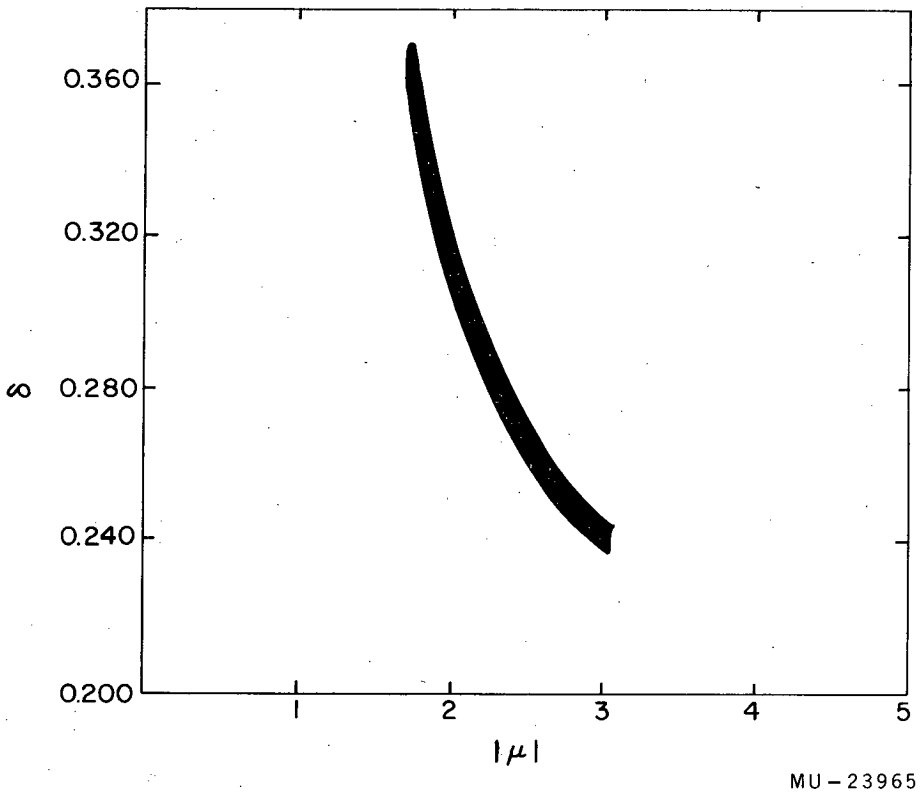


Fig. 11. Functional relationship between the magnetic moment of Pm^{143} and E2/M1 mixing ratio of the 740-kev γ ray of Nd^{143}, as determined by this experiment. Width of line includes experimental error. It is assumed that I = 7/2+.

Table II

Theoretical and experimental $B_2(T)U_2F_2$, assuming a spin of 7/2 +

	Exptl	A	.01350	.01400	.01500	.01700	.01900	.02100	.02400
		μ	1.72	1.79	1.91	2.16	2.42	2.68	3.06
		U_2F_2	.31160	.29270	.26040	.21205	.17813	.15343	.12728
		F_2	-.337	-.316	-.282	-.229	-.193	-.166	-.138
		δ	.364	.350	.328	.294	.272	.256	.241
1/T (H=0)									
5	0.0005		.00081	.00082	.00084	.00087	.00092	.00096	.00104
10	0.0031		.00322	.00325	.00332	.00346	.00362	.00380	.00409
15	0.0070		.00715	.00722	.00735	.00765	.00798	.00834	.00893
20	0.0140		.01249	.01260	.01281	.01327	.01378	.01434	.01524
25	0.0214		.01910	.01923	.01951	.02012	.02079	.02150	.02265
30	0.0295		.02680	.02695	.02728	.02798	.02873	.02952	.03078
35	0.0385		.03541	.03557	.03590	.03660	.03734	.03812	.03933
40	0.0475		.04476	.04489	.04517	.04577	.04639	.04703	.04801
45	0.0567		.05467	.05476	.05493	.05529	.05566	.05604	.05661
50	0.0650		.06500	.06500	.06500	.06500	.06500	.06500	.06500
1/T (H=200)									
5	0.003		.00081	.00082	.00084	.00087	.00092	.00096	.00104
10	0.007		.00322	.00325	.00332	.00346	.00362	.00380	.00409
15	0.012		.00715	.00722	.00735	.00765	.00798	.00834	.00893
20	0.019		.01249	.01260	.01281	.01327	.01378	.01434	.01524
25	0.027		.01910	.01923	.01952	.02013	.02079	.02150	.02265
1/T (H=400)									
5	0.004		.00081	.00082	.00084	.00087	.00092	.00096	.00104
10	0.011		.00322	.00325	.00332	.00346	.00362	.00380	.00409
15	0.018		.00715	.00722	.00735	.00765	.00798	.00834	.00893
20	0.026		.01249	.01260	.01281	.01327	.01378	.01434	.01524
25	0.036		.01910	.01924	.01952	.02013	.02079	.02151	.02265

of $\delta > 0.23$ and $|\mu| > 1.0$ may be set from this work alone. According to the conversion-coefficient data, this transition is essentially pure M1, with the limits of error just including this multipolarity. The results presented here necessitate an E2 admixture of at least 5%. Indeed, a pure M1 transition would require the anisotropy to have a sign opposite to that observed. Assuming the magnetic moment of Pm^{143} to lie between the Schmidt and Dirac limits of 1.72 and 3.11, respectively, we find $0.23 < \delta(E2/M1) < 0.37$. Thus the transition is 8 ± 3% quadrupole.

If the spin of Pm^{143} were 5/2+, the relationship between μ and δ (Table III) would be that shown in Fig. 12. Assuming that the magnetic moment of Pm^{143} falls between the Schmidt and Dirac limits of 4.79 and 3 respectively, these results give $0.255 \geqslant \delta \geqslant 0.217$. This means that the amount of quadrupole admixed would be less than 6.2%. Unlike the spin 7/2+ case, wherein the theoretical curves of B_2 vs 1/T for large $|\mu|$ fit the data best, for a spin of 5/2+ the curves for the smaller μ were better.

When a 400-gauss polarizing field was applied, tne anisotropy was enhanced. None of the theoretical curves predicted a noticeable enhancement.

Table III

Theoretical and experimental $B_2(T)U_2F_2$, assuming a spin of 5/2+

1/T	Exptl	A	0.03270	0.0349	0.0371	0.0392	0.0414	0.0436	0.0458	0.0480	0.0051	0.522
		μ	3.07	3.28	3.48	3.68	3.89	4.1	4.3	4.51	4.71	4.9
		F_2	0.1594	0.1467	0.1361	0.1277	0.1202	0.1138	0.1084	0.1036	0.0997	0.0962
		δ	0.253	0.245	0.239	0.234	0.229	0.226	0.223	0.220	0.218	0.216
H=0												
5	0.0005		0.00083	0.00112	0.00117	0.00123	0.00129	0.00135	0.00142	0.00149	0.00156	0.00163
10	0.0031		0.00419	0.00438	0.00457	0.00477	0.00499	0.00522	0.00546	0.00571	0.00596	0.00622
15	0.0070		0.00912	0.00950	0.00989	0.01028	0.01071	0.01115	0.01161	0.01208	0.1254	0.01301
20	0.0140		0.01553	0.01609	0.01667	0.01724	0.01786	0.01849	0.01914	0.01980	0.02044	0.02109
25	0.0214		0.02301	0.02371	0.02442	0.02512	0.02586	0.02662	0.02738	0.02815	0.02889	0.02962
30	0.0295		0.03119	0.03194	0.03271	0.03235	0.03422	0.03499	0.03577	0.03654	0.03727	0.03799
35	0.0385		0.03973	0.04043	0.04114	0.04182	0.04252	0.04322	0.04391	0.04458	0.04522	0.04584
40	0.0475		0.04833	0.04889	0.04945	0.04997	0.05051	0.05104	0.05156	0.05206	0.05253	0.05298
45	0.0567		0.05681	0.05713	0.05744	0.05773	0.05803	0.05832	0.05860	0.05887	0.05912	0.05936
50	0.0650		0.06500	0.06500	0.06500	0.06500	0.06500	0.06500	0.06500	0.06500	0.06500	0.06500
H=200												
5	0.003		0.00083	0.00112	0.00117	0.00123	0.00129	0.00135	0.00142	0.00149	0.00156	0.00163
10	0.007		0.00419	0.00438	0.00457	0.00477	0.00499	0.00522	0.00546	0.00571	0.00596	0.00622
15	0.012		0.00912	0.00950	0.00989	0.01028	0.01071	0.01115	0.01161	0.01208	0.01254	0.01301
20	0.019		0.01553	0.01609	0.01667	0.01724	0.01786	0.01849	0.01914	0.01980	0.02044	0.02110
25	0.027		0.02302	0.02371	0.02442	0.02512	0.02586	0.02662	0.02738	0.02815	0.02889	0.02962

Table III (continued)

Theoretical and experimental $B_2(T)U_2F_2$, assuming a spin of 5/2+

H=400											
5	0.004	0.00083	0.00112	0.00117	0.00123	0.00129	0.00135	0.00142	0.00149	0.00156	0.00163
10	0.011	0.00419	0.00438	0.00457	0.00477	0.00499	0.00522	0.00546	0.00571	0.00596	0.00622
15	0.018	0.00912	0.00950	0.00989	0.01028	0.01071	0.01115	0.01161	0.01208	0.01254	0.01301
20	0.026	0.01553	0.01609	0.01667	0.01724	0.01786	0.01849	0.01914	0.01980	0.02044	0.02170
25	0.036	0.02302	0.02371	0.02443	0.02512	0.02587	0.02662	0.02739	0.02815	0.02889	0.02963

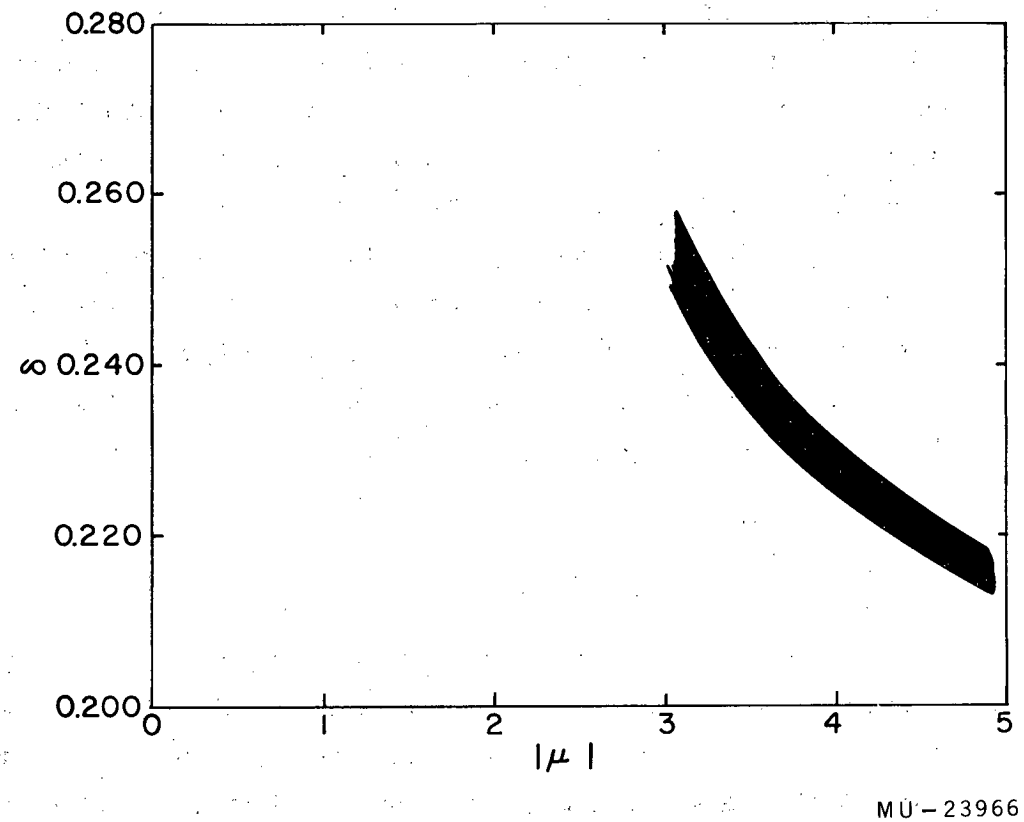


Fig. 12. Functional relationship between the magnetic moment of Pm^{143} and E2/M1 mixing ratio of the 740-kev γ ray of Nd^{143}, as determined by this experiment. Width of line includes experimental error. It is assumed that I = 5/2+.

VII. TERBIUM-156

A. Introduction

Terbium-156 was considered a suitable isotope for a nuclear-orientation experiment for several reasons. First, stable Tb^{159} had been studied in an ethylsulfate lattice by paramagnetic resonance, so the atomic parameters were known. Second, an orientation experiment had been done on Tb^{160} in which a large effect was observed. Third, the decay scheme of Tb^{156} had been thoroughly investigated by Ofer[19] and by Hansen, Nielsen, and Sheline.[20] From their work enough was known about the decay scheme to make it possible to interpret the orientation results. It also showed that the ground-state spin of Tb^{156} was not zero.

The Tb^{+3} ion has eight 4f electrons and the configuration $^7F_6$. In the ethylsulfate lattice a doublet $J_z = \pm 6$ lies lowest. Paramagnetic-resonance experiments[21] give $g_{\parallel} = 17.72 \pm 0.02$, $\Delta = 0.387 \pm 0.001$ cm^{-1}, or 0.5569 ± 0.001 ^oK. It has been calculated that $A/k = 0.234\ \mu/I$ ^oK, $P/k = 0.192\ Q/4I(2I-1)$ ^oK and $c = 0.211$ ^oK.[12]

B. Results

Fifteen photopeaks of Tb^{156} were resolved in the spectrum and all appeared to be anisotropic. It was not possible to find values of the anisotropy for any γ of energy less than 534 kev, because the correction for Compton background was too large to allow any sort of accuracy. Anisotropies were calculated (Table IV) for the 1841-kev, the 1643-kev, the 1417-kev, the 1220-kev, and the 534-kev γ rays. The 534-kev γ ray was the most prominant peak in the spectrum and was used for determining B_2.

It may be noted from the decay scheme (Fig. 13) that both the 1841-kev and 1643-kev transitions originate in the 1931-kev level, whereas the 1417-kev and the 1220-kev transitions originate in the 1507-kev level. Transitions originating in the same level have the same U_2's. Also, it may be seen that about 95% of the population of the 1507-kev level comes through the 534-kev transition. The 2042-kev and 1931-kev levels are populated directly by the electron decay of Tb^{156}.

Table IV

Experimental $B_2U_2F_2$'s at 1/T=56 for Tb^{156} in neodymium ethylsulfate

Gamma energy (kev)	$B_2U_2F_2$
534	-0.317±0.011
1220	0.1335±0.0045
1417	-0.206±0.016
1643	0.095±0.005
1841	0.205±0.025
2002	0.154±0.040

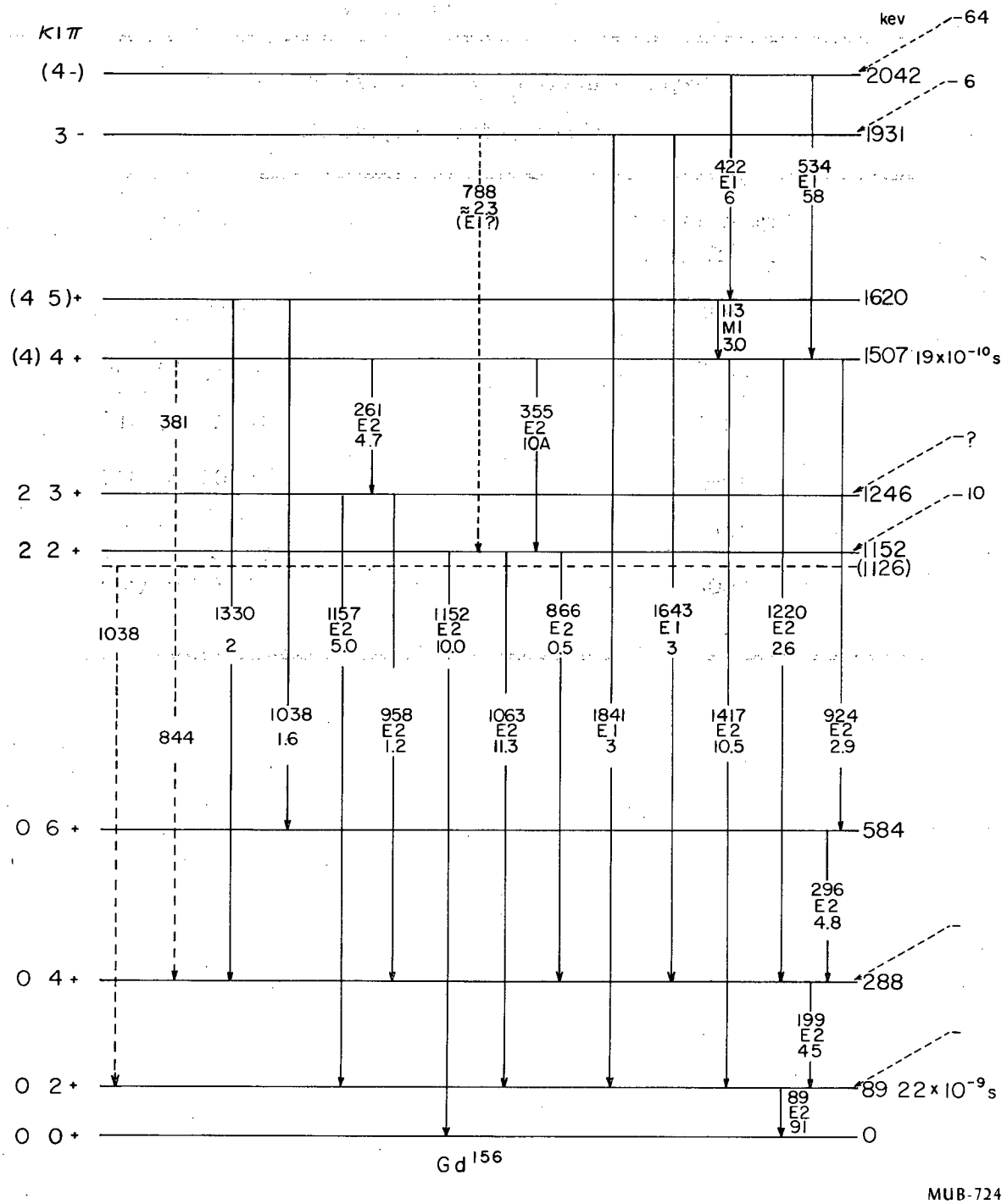


Fig. 13. The decay scheme for $Tb^{156} \rightarrow Gd^{156}$. The transitions are labelled with the energy in kev, the predominant multipole order when known, and the percent intensity of all K-capture decays.

The angular distribution of the 1417-kev γ ray was found to obey the equation $W(\theta) = 1 + X_2P_2(\cos\theta)$, where θ is the angle between the direction of propagation and the crystalline c axis (Fig. 14). At $0.017^{\circ}K$ we find $X_2 = -0.206 \pm 0.016$. Here X_2 is the experimental coefficient of the $P_2(\cos\theta)$ term. If there are no effects tending to attenuate the orientation, it is equal to $B_2U_2F_2$. No evidence for a B_4 term could be found, even at this temperature.

C. Discussion

The decay scheme as worked out by Hansen, Nielsen, and Sheline[20] and Ofer[19] has several unknown quantities: the spin of the 2042-kev level, the spin of the 1620-kev level, the existence of the 1953-kev transition, the ground-state spin of Tb^{156}, the angular momenta carried away in the electron-capture decay of Tb^{156}, and the mixing ratios of the γ rays. The nuclear-orientation work can determine some but not all of these, since they are not independent of one another; that is, in some cases there are just too many unknowns.

Both Ofer and Hansen, Nielsen, and Sheline have determined multipolarities of some of the transitions by conversion coefficients. The results are:

Energy (kev)	Hansen, Nielsen, and Sheline	Ofer
534	E1	E1 ($\delta^2 = 0.032$)
1220	E2	E2 + M1 ($\delta = -2.3^{-2.7}_{+0.8}$)
1417	E2	E2
1643	E1	
1841	E1	
"1953"	E1	

1. The Spin of the 2042-kev Level:

Both Ofer[19] and Hansen, Nielsen, and Sheline[20] tentatively assign a spin of 4- to this level. The 534-kev transition, which is primarily E1, goes from this level to the 4+ level at 1507 kev. The spin of this level could also be 3 or 5. If the 534-kev transition were pure E1, its

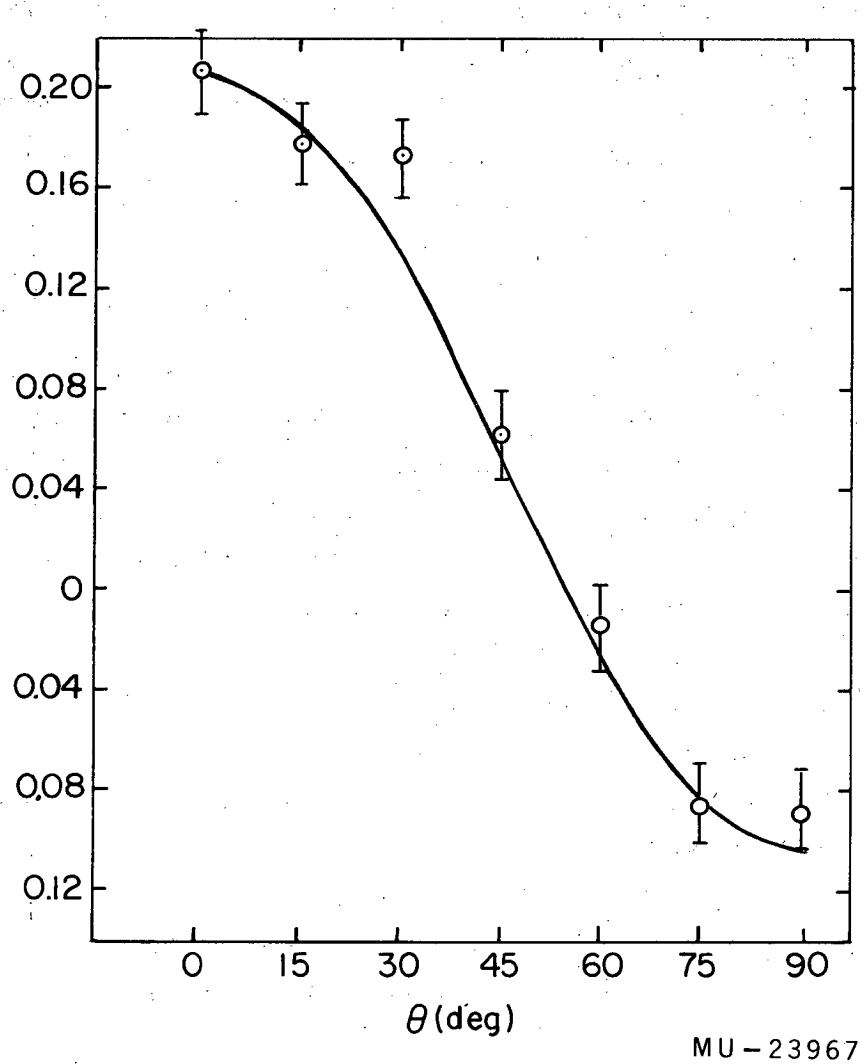


Fig. 14. Dependence of $-X_2P_2$ (cos θ) for the 1417-kev γ ray of Tb^{156} on the angle θ between detector and crystalline axis. The curve is 0.206 P_2(cos θ). 1/T = 58.5.

F_2 would be positive in both these cases. It would require more than 0.9% M2 admixture in the first case, and more than 2.4% M2 admixture in the second case, to make the F_2 negative. If the spin of the 2042-kev level were 3, the spin of Tb^{156} would probably be 3. If the spin of the 2042-kev level were 5, the spin of Tb^{156} would probably be 4. The ratio of the X_2 of the 534-kev γ ray to the X_2 of the 1841-kev γ ray is $(-0.317 \pm 0.011)/(0.205 \pm 0.025) = (-1.55 \pm 0.25)$. Since the B_2's are the same, this is also the ratio of the $U_2F_2(534)$ to the $U_2F_2(1841)$. The U_2's depend on the spin of Tb^{156}, the spin of the level to which they are going and the angular momentum which they carry away. Therefore if the $F_2(1841)$ is known, the $F_2(534)$ can be found as a function of the ground-state spin of Tb^{156}, the spin of the 2042-kev level, and the angular momentum which the two electron-capture decays carry away. If the 1841-kev γ ray is pure E1, then $F_2(1841) = 0.346$. The following extreme cases are possible, as well as those in which the electron-capture transitions are mixed. The L(kev) is the angular momentum carried away in the electron capture to the designated level.

I(2042)	I(Tb^{156})	L(2042)	L(1931)	$F_2(534)$	% M2 (534)
3	3	0	0	- 0.536	impossible
3	3	1	1	- 0.536	impossible
3	3	0	1	- 0.485	34
3	3	1	0	- 0.593	impossible
5	4	1	1	- 0.548	22

All these possibilities contradict the conversion coefficient data, therefore a spin of either 3 or 5 is ruled out for the 2042-kev level.

2. The Spin of the 1620-kev Level:

It is possible to say from the nuclear alignment experiments that the $B_2U_2F_2$ of the 422-kev γ ray is positive, and therefore that $F_2(422)$ is positive. This is consistent with an E1 transition going from a 4- state to a 5+ state— the state at 1620 kev— and is inconsistent with an E1 transition going from a 4- state to another spin 4 state.

Although also consistent with an E1 transition going from a spin 4- to a spin 3+ state, a spin of 3 can be ruled out for this level because an appreciable fraction of its decays go to a spin 6 state.

3. Existence of the 1953-kev Transition:

Ofer includes a transition of 1953 kev in the decay scheme, which goes from the 2042-kev state to the 89-kev state. Hansen, Nielsen, and Sheline find several transitions with energies between 1990 and 2030 kev which seem to be E1 in character, but find no transition with an energy of 1953 kev. In the experiments reported here, there was a peak which appeared to have an energy of about 2002±5 kev. Although no particular effort was made to determine the energy accurately, it is felt that the energy of the apparent peak in the γ spectrum is definitely greater than 1953 kev. It is not possible for us to say whether this is a single or multiple peak. Both Ofer and Hansen, Nielsen, and Sheline find the peak in coincidence with the 89-kev transition only, so it must go to the 2+ state of the ground-state band. The sign of the experimental F_2 is positive, which seems to rule out the possibility that the transition is between a 4- and 2+ state, since at least 24% octupole character would be needed to make F_2 positive.

4. The Other Unknown Quantities:

The multipolarity of the 534-kev γ ray, the multipolarity of the 1417-kev γ ray, the multipolarity of the 1220-kev γ ray, the angular momenta carried away in the electron-capture decay to the 2042-kev and the 1931-kev levels, and the spin of the ground state of Tb^{156} all depend on one another. In particular, the assumptions made about the multipolarities of the 534-kev and 1417-kev γ rays determine the values of the rest of the quantities. There are at least three ways of looking at the data. An assumption common to all three is that the 1841-kev γ ray is pure E1.

a. First Method of Interpretation— Assumptions:

The 1417-kev transition is pure E2. The 534-kev transition may have some M2 mixed with the E1. There is good reason to believe that the 1417-kev transition should be pure E2. The transition is

between a 4+ and a 2+ state. Hansen, Nielsen, and Sheline[20] found $\alpha_k = (9.6 \pm 3) \times 10^{-4}$. The values given by Sliv and Band are α_k (E2)= 1.0×10^{-3} and α_k (M3) = 5.8×10^{-3}.[13] Ofer's conversion-coefficient data allow as much as 3.1% M2 in the 534-kev transition; it is also assumed that there is no reorientation during the lifetime of the 1507-kev level. This level has a measured half life of 1.9×10^{-10} sec.

Results:

With these assumptions, the $F_2(1417)$ would be -0.448. The ratio of $X_2(534)$ to $X_2(1417)$ is $(-0.317 \pm 0.011)/(-0.206 \pm 0.016) = 1.539 \pm 0.187$. This ratio theoretically also equals $F_2(534)/[U_2' \times F_2(1417)]$. Here U_2' describes the reorientation of the nucleus during the 534-kev transition. It is a function of the spins of the 2042-kev and 1507-kev levels, and of the multipolarity of the transition. Both $F_2(534)$ and U_2' are functions of δ, the ratio of the quadrupole to the dipole matrix elements in this transition. When one solves for δ it is found to be $\left(0.32^{+?}_{-0.16}\right)$, which means that the 534-kev transition is $\left(9.3^{+?}_{-6.8}\right)$ % M2. With the assumptions that the 1417-kev transition is pure E2, and that there is no attenuation in the intermediate state, the angular correlation data of Ofer allow between 4 and 7% M2.

The ratio $X_2(534)/X_2(1841)$ is $(-0.317 \pm 0.011)/(0.205 \pm 0.025) = (-1.55 \pm 0.25)$. The B_2's are the same, so if one computes $U_2(534/U_2(1841)$ using the $F_2(534)$ of $\left(-0.568^{+0.043}_{-?}\right)$ calculated above, this ratio is found to be $\left(0.944^{+0.221}_{-?}\right)$.

$I(Tb^{156})$	L(2042)	L(1931)	$U_2(534)/U_2(1841)$
4	0	1	1.104
4	1	1	0.939
3	1	0	0.905
3	1	1	1.208

The experimental ratio of the U_2's is seen to be closest to the theoretical value for the case in which Tb^{156} has a spin of 4, and one unit of angular momentum is carried away in the decay to both the 2042-kev and 1931-kev levels.

The experimental value of $X_2(1643)$ is 0.095 ± 0.005. From the ratio of this quantity to $X_2(1841)$, it is found that $F_2(1643) = 0.160 \pm 0.008$. The ratio $X_2(1220)/X_2(1417) = (0.1335 \pm 0.0045)/(-0.206 \pm 0.016)$ gives $F_2(1220) = 0.290 \pm 0.030$. This implies that the 1220-kev transition is $\left(99.8^{+0.2}_{-0.7}\right)$ % quadrupole.

Although most of the data fit this assumption, the conversion-coefficient data for the 534-kev transition seem contradictory. If this transition were $\left(9.3^{+?}_{-6.8}\right)$ % M2, then α_k would be $\left(0.0093^{+?}_{-0.004}\right)$. The lower limits of this error do fall within the upper limits of Ofer's error, but not within the limits of error of Hansen, Nielsen, and Sheline.

b. Second Method of Interpretation— Assumptions:

The 534-kev transition is pure E1. This is justified on the basis of conversion-coefficient data. Hansen, Nielsen, and Sheline[20] found $\alpha_k = 0.0030 \pm 0.008$ and Ofer[19] found $\alpha_k = 0.0042 \pm 0.0015$. Sliv and Band[13] give theoretical values of $\alpha_k(E1) = 0.0038$ and $\alpha_k(M2) = 0.0632$. The former results indicate that there can be no M2 admixture in this transition, whereas the latter results allow as much as 3.2% M2.

Results:

With these assumptions, the $F_2(534)$ would be -0.439 and the $F_2(1815)$ would be 0.346. We find the ratio $U_2(534)/U_2(1841) = (-1.55 \pm 0.25)(0.346/-0.439) = (1.21 \pm 0.19)$. This experimental value is seen to be closest to the theoretical case in which the spin of Tb^{156} is 3, and one unit of angular momentum is carried away in the decay to both the 2042-kev and the 1931-kev levels.

Since it is assumed that the 534-kev transition is pure E1, U_2' would be 0.850. Using this figure, we determine $F_2(1417) = -0.336 \pm 0.024$ and $F_2(1220) = 0.217 \pm 0.005$. For the 1417-kev transition this would mean $\delta(M3/E2) = -0.11 \pm 0.03$ or that the transition is 1.2 ± 0.5% octupole. For the 1220-kev transition it would mean $\delta\ (E2/M1) = 14.9 \pm 1.6$, or that it is 99.5 ± 0.5% quadrupole.

Although the amount of octupole admixture in the 1417-kev transition is small, it seems rather unlikely that there would be any.

However, Hansen, Nielsen, and Sheline tentatively assign a K quantum number of 4 to the 1507-kev level. The 89-kev level has a K quantum number of 0, which means that there would be $\Delta K = 4$ for this transition. This would tend to hinder it and might allow this much octupole admixture. The single-particle model predicts that the ratio of the half lives would be greater than 10^7.

A spin of 3 for the ground state of Tb seems the more likely from decay-scheme considerations. Hansen, Nielsen, and Sheline have tried to balance intensities, and they find that 64% of the EC goes to the 2042-kev level, which has a spin of 4, 6% goes to the spin 3 level at 1931-kev, and 10% goes to the spin 2 level at 1152 kev. The proton state, as found in other Tb isotopes, is probably 3/2+ (411). The Nilsson diagram shows that there is a 3/2- (521) state available for the 91st neutron. There is also a 5/2- neutron state nearby, but its asymptotic quantum numbers are (523), so that it would couple antiparallel, leading to a spin of 1-. A 5/2+ [642] neutron state is available which would couple to I=4.

c. Third Method of Interpretation— Assumptions:

The 534-kev transition is pure E1. The 1417-kev transition is pure E2. The U_2' reflects the possibility that some reorientation may be taking place during the lifetime of the 1507-kev level.

Results:

The ratio between $U_2(534)$ and $U_2(1841)$ is derived as in the second method above (b), so a spin of 3 is favored for Tb^{156}. The F_2 of the 1220-kev transition is the same as it was in the first method (a).

To give these results, U_2' would have to be 0.637. If the intensities are correct, and 95% of the population of the 1507-kev level comes through the 534-kev transition, the remaining 5% would have to have a U_2 of -3.4. No reasonable assemblage of angular momenta could give this result. Therefore the only explanation for this apparent U_2' is that some reorientation is taking place during the lifetime of this level. The lifetime has been measured as 1.9×10^{-10} sec. Ofer finds that $F_2(1417)$ is 0.55 of the value for a pure transition. In this work, we find that $F_2(1417)$ is 0.75 of the value for a pure transition. Ofer's angular

correlation work was done in a dilute aqueous solution of $Tb(NO_3)_3$, and the nuclear orientation was done in a crystal of neodymium ethylsulfate. It is possible that the amount of reorientation occurring during the lifetime of this state would be different in the two environments. It is somewhat surprising that the attenuation is greater in the aqueous solution, since a solution is generally used to obtain an unattenuated correlation.

5. Nuclear Moments:

The nuclear moments were obtained from the temperature and magnetic-field dependence of $-X_2(534)$ (Fig. 15; Table V). The 534-kev γ ray was used because the statistical accuracy of its X_2 was better than that of any other resolved γ ray, and also because its X_2 had been studied at a larger number of temperatures than had that of any other γ ray.

Since $X_2(T) = B_2(T)U_2F_2$ if there is no attenuation, it is necessary to know U_2 and F_2 in order to obtain $B_2(T)$. The first method of interpreting the data gives one value of U_2F_2, 0.475, and favors a spin of 4 for the ground state of Tb^{156}. The second and third methods give $U_2F_2 = 0.397$ and favor a spin of 3 for Tb^{156}. Thus two different curves of $B_2(T)$ vs $1/T$ are obtained. Theoretical curves were calculated for various values of μ, the magnetic moment, and Q, the quadrupole moment. The relationships between μ and the A term in the Hamiltonian has been calculated[12] to be $Z = 0.234\ \mu/I\ ^{\circ}K$. The relation between Q and the P term in the Hamiltonian has been calculated to be $P = 0.192\ Q/4I(2I-1)$. These curves were fitted to $B_2(1/T=40)$. The alignment data did not determine A and P but gave a relationship between them. For a ground-state spin of 4, we find $A = (11.3 \pm 0.1)\ P + (0.051 \pm 0.004)\ ^{\circ}K$ (Table VI). For a ground-state spin of 3, we find $A = (9.1 \pm 0.1)\ P + (0.07 \pm 0.005)\ ^{\circ}K$ (Table VII). When these sets of values were compared with the alignment data for higher temperatures, and with the data for polarizing fields of 200 gauss and 400 gauss, the best fit was obtained for a spin of 4 for $A = (0.072 \pm 0.010)\ ^{\circ}K$ or $\mu = (1.23 \pm 0.17)$ n.m. and $P = (0.0018 \pm .0007)\ ^{\circ}K$ or $Q = (1.05 \pm 0.41)$ b. The best fit for a spin of 3 was obtained for $A = 0.1134 \pm 0.0047\ ^{\circ}K$ or $\mu = (1.45 \pm 0.06)$ n.m. and $P = (0.0045 \pm 0.0015)\ ^{\circ}K$ or $Q = (1.40 \pm 0.476)$ b.

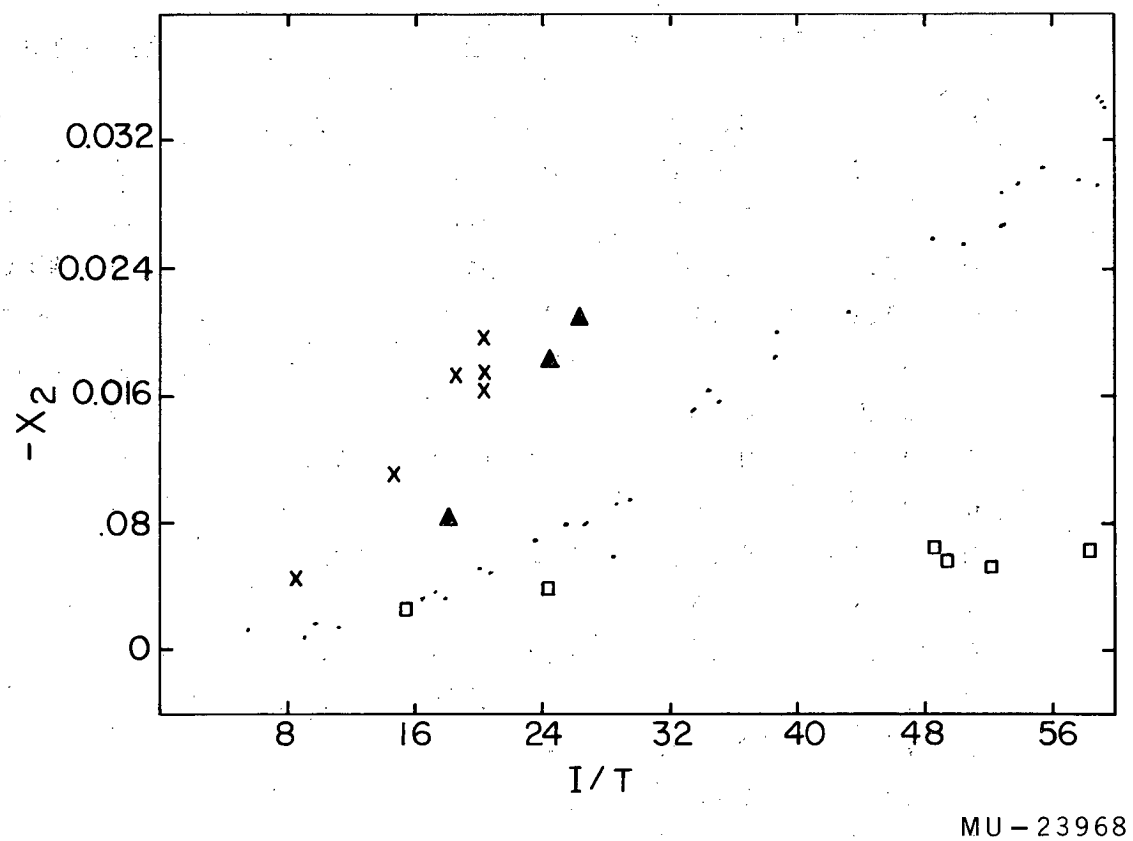


Fig. 15. Observed variation with 1/T and H of $-X_2P_2(\cos 0^\circ)$ of the 534-kev γ ray of Tb^{156}. H_i is the internal crystalline field, which in neodymium ethylsulfate is 180 gauss, in yttrium ethylsulfate 0 gauss.

	H_i (gauss)	H_{ext} (gauss)
■	0	0
•	180	0
▲	180	200
x	180	400

Table V

Experimental $B_2(T)U_2F_2$ for the 534-kev γ ray

Neodymium ethylsulfate						Yttrium ethylsulfate		
H	1/T	$-B_2U_2F_2$	H	1/t	$-B_2U_2F_2$	H	1/T	$-B_2U_2F_2$
0	5.7	0.013	0	57.6	0.295	0	15.5	0.027
0	9.2	0.009	0	58.6	0.292	0	24.5	0.041
0	9.8	0.016	0	58.8	0.347	0	48.6	0.065
0	11.1	0.014	0	59.0	0.345	0	49.4	0.059
0	16.1	0.025	0	59.2	0.342	0	52.1	0.054
0	16.6	0.033				0	58.1	0.065
0	17.3	0.036	200	18.2	0.082			
0	17.8	0.034	200	24.5	0.185			
0	20.2	0.053	200	26.5	0.219			
0	20.8	0.050						
0	23.6	0.069	330	15.0	0.112			
0	25.4	0.079	330	16.7	0.161			
0	26.6	0.079	340	18.5	0.104			
0	28.4	0.060	350	25.4	0.172			
0	28.7	0.092						
0	29.5	0.095	390	8.3	0.047			
0	33.4	0.150	400	14.4	0.115			
0	34.4	0.163	390	14.8	0.114			
0	35.0	0.157	400	18.4	0.172			
0	38.6	0.185	390	20.3	0.163			
0	38.6	0.200	390	20.3	0.171			
0	43.2	0.213	390	20.3	0.194			
0	48.4	0.258						
0	50.4	0.255						
0	52.7	0.266						
0	52.8	0.288						
0	53.8	0.272						
0	57.4	0.304						

Table VI

Calculated and experimental values of B_2 for I = 4 (Tb^{156}) in neodymium ethylsulfate

	B_2 exptl	A	0.0472	0.751	0.8757	0.0996	0.1228	0.1563	0.1891	0.2542
		μ	0.80	1.28	1.50	1.70	2.10	2.68	3.24	4.35
		P	0.000	0.002	0.003	0.004	0.006	0.009	0.012	0.018
		Q	0.00	1.17	1.75	2.34	3.50	5.26	7.00	10.5
1/T (H=0)										
5	0.005		0.0259	0.0056	-0.0026	-0.0087	-0.0167	-0.0209	-0.0181	0.0017
10	0.015		0.0629	0.0338	0.0246	0.0183	0.0125	0.0155	0.0279	0.0695
15	0.050		0.1053	0.0729	0.0643	0.0594	0.0578	0.0688	0.0894	0.1456
20	0.095		0.1556	0.12627	0.1201	0.1180	0.1219	0.1395	0.1648	0.2244
25	0.165		0.2139	0.1918	0.1889	0.1895	0.1971	0.2170	0.2416	0.2939
30	0.250		0.2793	0.2660	0.2655	0.2676	0.2758	0.2930	0.3122	0.3498
35	0.340		0.3502	0.3450	0.3455	0.3473	0.3528	0.3629	0.3734	0.3928
40	0.425		0.4250	0.4252	0.4250	0.4250	0.4250	0.4250	0.4251	0.4250
45	0.500		0.5017	0.5042	0.5017	0.4985	0.4911	0.4793	0.4681	0.4489
50	0.560		0.5784	0.5799	0.5739	0.5668	0.5508	0.5264	0.5039	0.4665
1/T (H=200)										
5	0.01		0.027	0.007	0.001	-0.005	-0.011	-0.013	-0.008	0.014
10	0.03		0.076	0.061	0.059	0.059	0.064	0.079	0.098	0.143
15	0.10		0.145	0.151	0.158	0.166	0.183	0.208	0.232	0.275
20	0.22		0.236	0.268	0.283	0.296	0.318	0.342	0.359	0.383
25	0.40		0.342	0.397	0.415	0.428	0.447	0.460	0.464	0.463
1/T (H=400)										
5	0.04		0.029	0.013	0.008	0.004	0.000	0.002	0.010	0.037
10	0.13		0.096	0.102	0.109	0.116	0.133	0.158	0.182	0.225
15	0.24		0.196	0.239	0.258	0.275	0.302	0.330	0.349	0.375
20	0.39		0.319	0.393	0.417	0.435	0.459	0.476	0.481	0.479
25			0.452	0.540	0.563	0.577	0.591	0.592	0.582	0.552

Table VII

Calculated and experimental B_2 for I = 3 (Tb^{156}) in neodymium ethylsulfate

1/T (H=0)	B_2exptl									
		A	0.0676	0.0891	0.0990	0.1087	0.1181	0.1273	0.1454	0.1803	0.2312
		μ	0.87	1.15	1.27	1.39	1.51	1.63	1.86	2.31	2.96
		P	0.000	0.002	0.003	0.004	0.005	0.006	0.008	0.012	0.018
		Q	0.00	0.63	0.94	1.25	1.56	1.87	2.50	3.75	5.63
5	0.010±0.010		0.0315	0.0188	0.0135	0.0088	0.0046	0.0012	-0.0044	-0.0105	-0.0100
10	0.030		0.0762	0.0584	0.0516	0.0459	0.0413	0.0377	0.0329	0.0314	0.0428
15	0.063		0.1272	0.1073	0.1003	0.0949	0.0909	0.0881	0.0857	0.0901	0.1116
20	0.125		0.1873	0.1692	0.1635	0.1596	0.1572	0.1561	0.1569	0.1672	0.1952
25	0.205		0.2560	0.2422	0.2387	0.2368	0.2361	0.2365	0.2398	0.2531	0.2814
30	0.292		0.3317	0.3234	0.3218	0.3214	0.3219	0.3231	0.3272	0.3394	0.3621
35	0.405		0.4122	0.4088	0.4085	0.4088	0.4095	0.4106	0.4135	0.4208	0.4334
40	0.495		0.4950	0.4950	0.4950	0.4950	0.4950	0.4950	0.4950	0.4950	0.4950
45	0.585		0.5775	0.5789	0.5784	0.5774	0.5759	0.5742	0.5702	0.5613	0.5476
50	0.660		0.6578	0.6588	0.6571	0.6543	0.6508	0.6470	0.6384	0.6201	0.5924
55	0.750		0.7340	0.7334	0.7341	0.7250	0.7194	0.7133	0.7000	0.6722	0.6309
60	0.845		0.8053	0.8020	0.8054	0.7894	0.7816	0.7732	0.7554	0.7185	0.6643
1/T (H=200)											
5	0.02±0.03		0.033	0.021	0.016	0.012	0.008	0.005	0.001	-0.004	-0.001
10	0.06		0.091	0.082	0.078	0.078	0.078	0.079	0.080	0.090	0.112
15	0.16		0.174	0.177	0.180	0.184	0.189	0.193	0.203	0.224	0.254
20	0.27		0.279	0.298	0.307	0.316	0.324	0.331	0.345	0.368	0.392
25	0.47		0.398	0.429	0.442	0.453	0.462	0.470	0.483	0.498	0.509

Table VII (continued)

Calculated and experimental B_2 for I = 3 (Tb^{156}) in neodymium ethylsulfate

1/T (H=400)										
5	0.06±0.03	0.036	0.025	0.022	0.018	0.016	0.014	0.011	0.010	0.017
10	0.16	0.115	0.119	0.122	0.126	0.130	0.134	0.144	0.165	0.195
15	0.30	0.232	0.257	0.269	0.281	0.291	0.301	0.325	0.346	0.374
20	0.45	0.370	0.413	0.431	0.445	0.457	0.468	0.484	0.5045	0.518
25		0.513	0.564	0.582	0.595	0.606	0.614	0.625	0.634	0.629

These values also provided a satisfactory fit for Tb^{156} in yttrium ethylsulfate. Although the data in this salt are not too good, since the temperature is not known very accurately, they do require that P be within the above limits for the two spins (Table VIII).

Table VIII

Calculated and experimental values of $-B_2U_2F_2$ for the 534-kev γ ray of Tb^{156} in yttrium ethylsulfate, at 1/T = 50.

Exptl.	I	3	3	3	3	3	4	4	4	4
	A	0.068	0.089	0.099	0.118	0.127	0.047	0.062	0.075	0.088
	P	0	0.002	0.003	0.005	0.006	0	0.001	0.002	0.003
0.0625 ±0.0250		0.142	0.097	0.068	0.049	0.037	0.142	0.091	0.051	0.021

It is possible to generalize the formula given by Nilsson for the magnetic moment produced by one odd nucleon[7] to the case in which there are both an odd proton and an odd neutron. The formula is

$$\mu = \frac{I}{I+1}\{1/2\,(g_{sp} - g_{\ell p}) \sum_{\ell} (a^2_{\ell,\Omega-1/2} - a^2_{\ell,\Omega+1/2}) + g_{\ell p}\Omega_p + 1/2\, g_{sn} \sum_{\ell} (b^2_{\ell,\Omega-1/2} - b^2_{\ell,\Omega+1/2}) + g_R\},$$

where $g_{sp} = 5.585$, the gyromagnetic ratio due to the intrinsic spin of the proton,

$g_{sn} = -3.826$, the gyromagnetic ratio due to the intrinsic spin of the neutron,

$a_{\ell,\Omega\pm1/2}$ are the coefficients of the basis vectors for the eigenfunction of a proton moving in a deformed nuclear field,

$b_{\ell,\Omega\pm1/2}$ are the coefficients of the basis vectors for the eigenfunction of a neutron moving in a deformed nuclear field,

$g_R \approx Z/A$	is the gyromagnetic ratio of the rotational motion of the nuclear core,
Ω_p	is the total spin of the proton,
I	is the nuclear spin, and
$g_{\ell p}$	is the gyromagnetic ratio due to the orbital motion of the proton.

If the ground-state spin of Tb^{156} is 3, and the deformation is $\eta = 6$, then probably the proton is in a 3/2 + (411) state, and the neutron in a 3/2- (521) state. These states give $\mu = 2.0$ nm. If one uses the g factors given by Rasmussen and Chiao,[22] one finds $\mu = 2.1$ nm. The theoretical value is seen to be in fairly good agreement with the experimental value. This agreement tends to confirm the hypothesis that the two odd particles are not strongly correlated. It is not possible to choose among the several proton and neutron states that could combine to give $I = 4$. Therefore it is not possible to say anything about the agreement between theory and experiment.

ACKNOWLEDGMENTS

I wish to express my gratitude to Professor John O. Rasmussen for his interest in directing this study, and to Professor David A. Shirley who ably and patiently supervised and aided this work.

I wish to thank Dr. Charles E. Johnson for his assistance in the early stages of this thesis.

I am grateful to all the official and unofficial inhabitants of Room 209A throughout the past four years for all the informative and interesting discussions we have had, and for their aid and encouragement. In particular, I would like to thank Dr. Eileen Flamm and Mrs. Mab Tocher for their help and moral support. Special thanks to my two able (addled) minded proofreaders, Lee Hyder and Bruce Wilkins.

I am indebted to the National Science Foundation for a fellowship partially supporting this research.

This work was done under the auspices of the U.S. Atomic Energy Commission.

REFERENCES

1. R. J. Blin-Stoyle and M. A. Grace, Oriented Nuclei, in Handbuch der Physik, Vol. 42 (Springer Verlag, Berlin, 1957), p. 555.

1a. V. N. Samoilov, V. V. Skliarevsky, and E. P. Stepanov, Polarization of Nuclei of Elements Dissolved in Iron, Akad. Nauk S.S.S.R., Institut Atomnoi Energii (1959). Translation: Lawrence Radiation Laboratory Translation UCRL-517.

2. Shirley, Smith, and Rasmussen, Nuclear Phys. 4, 395 (1957).

3. S. G. Thompson, B. G. Harvey, G. R. Choppin, and G. T. Seaborg, J. Am. Chem. Soc. 76, 6229 (1954).

4. J. F. Schooley, Some Low-Temperature Nuclear-Orientation Studies (Ph.D. Thesis), Lawrence Radiation Laboratory Report UCRL-9296, July 1960 (unpublished).

5. H. Meyer, Phil. Mag. (Ser. 8) 2, 521 (1957).

6. A. R. Rutledge, Finite Geometry Corrections to γ-Ray Angular Correlations measured with 5-in. Diameter by 6-in. long NaI(Tl) Crystals and with 3-in. diameter by 3-in. long NaI(Tl) Crystals: Chalk River, Ontario, Report CRP-851, July 1959 (unpublished).

7. S. V. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 29, No. 16 (1955).

8. E. P. Grigor'ev, B. S. Dzhelepov, A. V. Zolotavin, O. E. Kraft, B. Kratzik, and L. K. Peker, Bull. Acad. Sci.USSR, Phys. Ser. 22, No. 2, 99 (1958).

9. R. E. Sund and M. L. Wiedenbeck, Phys. Rev. 120, 1792 (1960).

9a. A. Cabezas, E. Lipworth, R. Marrus, and J. Winocur, Nuclear Spin of Sm^{153}, Lawrence Radiation Laboratory Report UCRL-8826, June 1959 (unpublished).

9b. K. D. Bowers and J. Owen, Paramagnetic Resonance II, in Reports on Progress in Physics, Vol. 18 (Physical Society, London, 1955), p. 304.

10. Shimon Ofer, Phys. Rev. 113, 895 (1959).

11. J. F. Schooley, D. A. Shirley, and J. O. Rasmussen, Nuclear Orientation of Pm^{144}, Lawrence Radiation Laboratory Report UCRL-9232, May 1960; Phys. Rev. 121, 558 (1961).

REFERENCES (con't)

12. C. E. Johnson, J. F. Schooley, and D. A. Shirley, Nuclear Orientation of Tb^{160}, Lawrence Radiation Laboratory Report UCRL-9267, June 1960; Phys. Rev. 120, 2108 (1960).

13. L. A. Sliv and I. M. Band, Coefficients of Internal Conversion of γ Radiation (Academy of Sciences, U.S.S.R., Leningrad Physico-Technical Institute, Leningrad, U.S.S.R., 1956); English translation: Report 57 ICC K1, Physics Department, University of Illinois, Urbana.

14. R. A. James and C. D. Bingham, Phys. Rev. 117, 810 (1960).

15. N. Starfelt and J. Cederlund, Phys. Rev. 105, 241 (1957).

16. R. W. Grant (Lawrence Radiation Laboratory, Berkeley), private communication.

17. Nuclear Data Sheets, NRC 58-11-74 (1958) (National Academy of Sciences National Research Council).

18. L. F. Kisslinger and R. Sorensen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Skrifter 32, No. 9 (1961).

19. S. Ofer, Phys. Rev. 115, 412 (1959).

20. Hansen, Nielsen, and Sheline: Nuclear Phys. 12,389 (1959).

21. J. M. Baker and B. B. Bleaney, Proc. Roy. Soc. (London) A245, 156 (1958).

22. L. W. Chiao and J. O. Rasmussen, in Proceedings of the International Conference on Nuclear Structure, Kingston, Canada, August 29 - September 3, 1960.

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.