Lawrence Berkeley National Laboratory
Recent Work

Title
NUCLEAR STUDIES USING SEMICONDUCTOR DETECTORS

Permalink
https://escholarship.org/uc/item/3pv101jc

Author
Frankel, Richard Barry.

Publication Date
1965-01-05
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.
UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

NUCLEAR STUDIES USING SEMICONDUCTOR DETECTORS

Richard Barry Frankel
(Ph.D. Thesis)

January 5, 1964
B. Nuclear Alignment of Pm$^{144}$ in CMN: A Test of the Proposed Temperature Scale
   1. Introduction ................................................. 78
   2. Experimental ............................................. 78
   3. Results and Discussion ................................. 80

V. The Decay of Ce$^{137g}$ and Ce$^{137m}$ and the Level Structure of La$^{137}$
   1. Introduction ................................................ 91
   2. Gamma-Ray Spectroscopy ................................. 91
   3. Position Branching ...................................... 101
   5. Coincidence Studies ................................. 106
   6. Spin Measurements: Nuclear Orientation and
      Angular Correlation .................................... 109
   7. Comparison with Theory ................................ 120

VI. The Angular Distribution of Alpha Particles from E$^{253}$
     Aligned in NES and the Relative Phases of the L=0 and
     L=4 Waves
   1. Introduction .......................................... 125
   2. Background .............................................. 125
   3. Angular Distribution of Alpha Particles from
      Oriented Nuclei ....................................... 130
   4. Previous Experiments ................................ 132
   5. Experiments on E$^{253}$ .................................. 133
   6. Discussion .............................................. 145

Acknowledgments .................................................. 148
NUCLEAR STUDIES USING SEMICONDUCTOR DETECTORS

Contents

Abstract ................................................................. v

I. Introduction ......................................................... 1

II. Theoretical and Experimental Background
    A. Theory of Nuclear Orientation ......................... 1
    B. Experimental
        1. Detectors .............................................. 9
        2. Nuclear Orientation Apparatus .................... 23
        3. Cryostat ............................................. 26
        4. Temperature Measurements ......................... 29
        5. Experimental Procedure ............................ 36
        6. Coincidence Techniques ............................ 37

III. The Angular Distribution of Conversion Electrons from
     Ce\(^{137m}\) Oriented in NES
     A. Conversion Electron Experiment
        1. Introduction ......................................... 39
        2. Experimental ......................................... 41
        3. Measurements and Results ......................... 42
        4. Discussion ........................................... 44
     B. The Angular Distribution of \(\gamma\)-rays from Ce\(^{137m}\) Oriented
        in NES and the NES Temperature Scale
        1. Introduction ......................................... 53
        2. Experimental ......................................... 53
        3. Results and Discussion ................................ 54

IV. Nuclear Alignment of Ce\(^{137m}\) in CMN and the CMN Temperature Scale
    A. Nuclear Alignment of Ce\(^{137m}\)
        1. Introduction ......................................... 63
        2. Experimental ......................................... 65
        3. Discussion ........................................... 72
NUCLEAR STUDIES USING SEMICONDUCTOR DETECTORS

Richard Barry Frankel
Department of Chemistry and Lawrence Radiation Laboratory
University of California, Berkeley, California
January 5, 1965

ABSTRACT

Germanium surface barrier detectors were used to measure the angular distribution of conversion electrons from the 255 keV isomeric transition in Ce$^{137m}$ for Ce$^{137m}$ nuclei aligned in the neodymium ethylsulfate lattice. Direct comparison with the γ-ray angular distributions gave the particle parameters $\langle b^2 \rangle_k = 1.061(18), \langle b^2 \rangle_L = 1.059(20)$. From the variation of the γ-ray anisotropy with temperature, the hyperfine coupling constant for Ce$^{137m}$ was determined to be $A = 0.0147(7)$ cm$^{-1}$ using the 1957 temperature scale for this salt.

Ce$^{137m}$ was also aligned in cerium magnesium nitrate and the variation of the 255 keV γ-ray anisotropy was studied as a function of temperature. Anomalies were shown to exist in the cerium magnesium nitrate temperature scale derived by Daniels and Robinson. A new temperature scale is proposed which goes down to 0.0019°K. To test the new scale, Pm$^{144}$ was aligned in cerium magnesium nitrate. The anisotropies of the 615 and 695 keV γ-rays were found to be attenuated, but confirmed the Ce$^{137m}$ results.

The decay of Ce$^{137m+g}$ to La$^{137}$ was investigated with Ce(Li) γ-ray detectors, Si(Li) electron detectors, and nuclear alignment. Levels were found at 10, 446, 692, 708, 762, 781, 835, 925, 1004, and 1170 keV. Definite spin and parity assignments were 762 keV (11/2+), 835 keV (9/2+), and 1004 keV (11/2-). A spin of 5/2+ was tentatively assigned to the 446 keV level. The 11/2- state corresponds to a quasi-particle state predicted by Rho.

The distribution of alpha particles from E$^{253}$ oriented in neodymium ethylsulfate was measured using germanium surface barrier detectors. The L=4 wave in the ground state transition was found to be out of phase with the L=0 and L=2 waves. The L=4 wave was found to have...
a larger amplitude and the $L=2$ wave a smaller amplitude than predictions based on the Bohr, Froman and Mottelson coupling relations. The relative amplitudes for the ground state transitions are $a_0 = 1$, $a_2 = 0.11$, and $a_4 = 0.016$. 
I. INTRODUCTION

The introduction of semiconductor crystals as detectors for particles and photons has allowed the investigation of several interesting problems in nuclear physics. With germanium surface barrier detectors, the first reported measurement of the angular distribution of conversion electrons from oriented nuclei was made. The nuclei were Ce$^{137m}$ in neodymium ethylsulfate. This is described in Sec. III. With the same detectors, the relative phase of the $L=0, L=4$ waves in the alpha decay of $^{253}\text{E}_2$ to the ground state of $^{249}\text{Bk}$ was determined by measuring the angular distribution of alpha particles from $^{253}\text{E}_2$ oriented in neodymium ethylsulfate (Sec. VI). High-resolution lithium-drifted germanium and silicon crystals have been used to investigate the decay of Ce$^{137}$ to La$^{137}$ (Sec. V).

During the course of these experiments, anomalies in the temperature scales for neodymium ethylsulfate and cerium magnesium nitrate were found. The temperature scale for cerium magnesium nitrate was investigated by nuclear alignment (Sec. VI).

The first section contains an introduction to nuclear orientation and describes the experimental apparatus.

II. THEORETICAL AND EXPERIMENTAL BACKGROUND

A. Theory of Nuclear Orientation

Nuclear orientation is a very powerful technique for the study of the angular momentum properties of nuclear states and of the radiation emitted in transitions between nuclear states.\textsuperscript{1} Since all methods of nuclear orientation involve an interaction between the nucleus and its environment, these interactions and the nuclear electric or magnetic moments involved may also be studied.

Nuclear orientation results when the $(2I+1)$-fold spatial degeneracy of a nucleus with total angular momentum (spin) $I$ is resolved and certain of the nuclear magnetic substates are preferentially populated. The substates may be characterized by the projections $I_z$ of the spin $I$. 
on an axis, called the orientation axis. If \( I_+ \) and \( I_- \) have equal population, the system is referred to as aligned. If \( I_+ \) and \( I_- \) have unequal populations, the system is referred to as polarized. The methods of nuclear orientation may be classified as equilibrium or static, and nonequilibrium or dynamic. In addition, angular correlations of radiations may be said to involve a nuclear orientation in the intermediate state. The dynamic methods involve resonance techniques and will not be considered here, as the static methods are more generally used for the study of nuclear properties.

Since the equilibrium populations of the nuclear magnetic substates depend on the ratio of their energies to \( kT \) as \( \exp[-E(I_+)/kT] \), and since these energy differences are usually small (\(< 1^\circ K\) ), the static methods require low temperature techniques. The static methods may be summarized as follows:

a) "Brute Force" polarization. The nuclei being oriented are cooled by contact with an adiabatically demagnetized paramagnetic salt. A large field is applied, which couples directly to the nuclear magnetic moment. For reasonable polarizations (\(~20\%) \) fields of the order of 50,000 gauss at 0.01\(^\circ\)K are needed. This is difficult experimentally, as application of the field would tend to warm up the salt. The technique has been applied to \( \text{In}^{115} \) in In metal.

b) Polarization in ferromagnets (anti-ferro magnets). The nuclei are dissolved or diffused into a ferromagnetic material, as iron, which is placed in contact with a paramagnetic salt, usually through copper fins or wires. The salt is demagnetized and a small magnetic field is applied to magnetize the sample. Core polarization or conduction electron polarization results in large fields at the impurity nucleus, which at 0.01\(^\circ\)K results in large polarizations. An example is the polarization of \( \text{Au}^{197} \) in iron.

c) Magnetic hyperfine structure polarization. This technique can be applied to nuclei in atoms with unpaired \( d \) or \( f \) electrons. In certain ionic salts, the unpaired electrons give rise to large (\(~10^6 \) gauss) fields at the nucleus. In most cases, the nuclei are embedded in the
paramagnetic salt used for cooling. Upon demagnetization, a small field (200-800 gauss) is applied to polarize the electron spins. This results immediately in nuclear polarization. For example, Tb\(^{160}\) nuclei were polarized in neodymium ethylsulfate.\(^6\)

\[\text{d) Magnetic hyperfine structure alignments. For some ions in certain paramagnetic salts, the hyperfine structure interaction is anisotropic with respect to the crystalline axis. Merely cooling the ions leads automatically to alignment. This is true for many rare earth ions in crystals of neodymium ethylsulfate and cerium magnesium nitrate. These salts can be demagnetized to low temperatures—the former to 0.02°K and the latter to \(\sim 0.002°K\). This is the major method used in this work.}\]

\[\text{e) Electric hfs alignment. Large electric field gradients with a definite orientation with respect to the crystalline axis exist at the nuclei in certain salts, such as at }\text{U as }\text{UO}_2^{+2}\text{ in uranyl rubidium nitrate. These field gradients can couple to the nuclear quadrupole moment to produce alignment. }\text{U}^{233}\text{ and }\text{I}^{131}\text{ were aligned by this technique.}\(^7,^8\)

\[\text{f) Polarization of closed shells. The polarization of closed shells by crystal fields can give rise to large field gradients in rare earth nuclei. Lu}^{177}\text{ has been aligned in neodymium ethylsulfate by this method.}\(^9\)

The Spin Hamiltonian

The various static methods of alignment may be conveniently summarized by the introduction of a spin Hamiltonian.\(^10\) This formulation may be applied to ions in paramagnetic crystals with axial symmetry and to nuclei in external fields. Neglecting collective effects, the electronic and nuclear interactions may be represented by:

\[\mathcal{H} = g_{||}H_zS_z + g_{\perp}(H_xS_x + H_yS_y) + D(S_z^2 - \frac{1}{2}S(S+1)) + E(S_x^2 - S_y^2) + AS_zI_z + B(S_xI_x + S_yI_y) + P(I_z^2 - \frac{1}{2}I(I+1)) - (\frac{\mu}{I}) \cdot \vec{H} \cdot \vec{I}.\]
$S$ is a fictitious spin, obtained by setting the multiplicity of the levels populated at low temperature to $2S+1$. For Kramer's ions (ions with an odd number of unpaired electrons) $S$ generally equals 1/2 for the rare earths (not for the iron group). In the first two terms, $\beta$ is the Bohr magneton and $g_\parallel$ and $g_\perp$ are the ionic $g$ factors measured along and perpendicular to the $z$ axis. The $D$ and $E$ terms represent the splitting of the $2S+1$ electronic levels by an axial distortion (the $D$ term) and a nonaxial distortion (the $E$ term). For $S = 1/2$, these terms vanish. The $A$ and $B$ terms represent the magnetic hyperfine interaction between the electronic spin and the nucleus; the $P$ term represents the electric hyperfine interaction between a field gradient at the nucleus and the nuclear quadrupole moment. The last term represents the direct coupling of the nuclear spin with an externally applied magnetic field. For individual cases, certain terms in the Hamiltonian dominate and the others can be neglected. The term in $H \cdot I$ corresponds to orientation in external fields or ferro- and anti-ferromagnets, with $H$ the external field or the internal field, respectively. Magnetic hfs polarization results from combinations of the first six terms and a small (500 gauss) external field; magnetic hfs alignment occurs if the $D$, $E$, $A$, and $B$ terms predominate, with $A \neq B$. The $P$ term leads to electric hfs alignment. It is often possible to predict which terms will apply to a given experiment, but one includes as many terms as are necessary to fit the data.

Having selected the proper Hamiltonian for a given case, one solves the equation

$$H \psi_n = E_n \psi_n$$

for the energy eigenvalues $E$, where the $\psi_n$ are states of the form $|S_z, I_z\rangle$. The populations are obtained by applying Boltzmann statistics. In certain cases, resonance experiments give values of $g_\parallel$, $g_\perp$, $A$, $B$, etc. Since the nuclear orientation depends on these constants and on $1/T$, by varying $1/T$, and observing the degree of orientation, one can determine
their values. Using the crystal field theory of Elliott and Stevens, it is possible to obtain values for the nuclear moments from the hyperfine interaction constants. There is some uncertainty in the derived moments since the expressions include a factor of \( \frac{1}{r^3} \) which must be calculated.

The Orientation Parameters

For systems with axial symmetry, specification of the occupation probabilities of the nuclear magnetic substates completely defines the nuclear orientation. In discussing the geometrical aspects of oriented nuclei (such as the angular distribution of radiation) it is more convenient to introduce parameters which can be derived from a "multipole" expansion of the density matrix. These are defined as:

\[
B_k(I, \mu, T) = \sum_{Iz} (2I+1)\langle I\cdot I\cdot I_z - I_z | k \cdot 0 \rangle W(I_z), \quad (II.2)
\]

where \( I \) is the nuclear spin, \( I_z \) is projection of \( I \) on the z axis, \( \langle \cdot \rangle \) is a Clebsch Gordan coefficient and \( W(I_z) \) is the occupation probability for the substate with \( I_z \). If \( B_1(I) \neq 0 \), there is nuclear polarization; if \( B_1(I) = 0 \) but \( B_2(I) \neq 0 \), there is nuclear alignment.

Radiation from Oriented Nuclei

If radioactive nuclei are oriented, their decay products may show a spatial anisotropy. The angular distribution of the radiation depends on the degree of initial orientation and on the angular momenta involved in the decay. For electromagnetic radiation, the angular distribution may be expanded as:

\[
W(\theta) = 1 + \sum_k B_k U_k F_k P_k (\cos \theta), \quad (II.3)
\]

where the \( B_k \) are the orientation parameters described above, \( P_k \) are Legendre polynomials of order \( k \), \( \theta \) is the angle between the direction of emission and the axis of orientation, \( U_k \) are coefficients which account for realignment due to any (unobserved) preceding transitions, and \( F_k \) are coefficients related to the multipolarity of the observed transition...
and to the spins of the states it connects. If detectors are used which subtend a finite solid angle at the source, solid angle correction coefficients $g_k$ must be included in Eq. (II.3). These have been calculated for NaI detectors.\(^{15}\) The upper limit on $k$ is the smaller of $2I$ (initial) or $2L$. Terms in the expansion for $k > 4$ are generally omitted because $B_6$ is usually small with respect to $B_2$ and $B_4$. The odd Legendre polynomials do not appear for the angular distribution of $\gamma$-radiation because parity conservation requires that the distribution be symmetric about $\theta = \pi/2$. The same condition applies to all radiation from parity conserving interactions, for example, alpha and conversion-electron emission.

As one might expect, the circular polarization of emitted $\gamma$-rays depends on the polarization of the parent nuclei; therefore, observation of circular polarization requires retention of the odd polynomials. The odd terms are also included for beta particles as the beta decay interaction violates conservation of parity.

If the sequence of transitions is $I_A \rightarrow I_B \rightarrow I_C$, where the parent nuclei with spin $I_A$ are aligned and radiation of multipolarity $L_2$ is observed,

$$F_k(L_1, I_B) = (-1)^{I_B} \left( \frac{1}{2} \right)^{L+1/2} \langle L_2 L_2 L_2 | k 0 \rangle W(L_1 I_B L_2 k L_2),$$

\[(II.4)\]

and

$$U_k = (-1)^{I_A + I_B} \left[ (2I_A + 1)(2I_B + 1) \right]^{1/2} W(I_A I_B \Lambda B_k L_1).$$

\[(II.5)\]

If there are several unobserved transitions, $U_k = \sum_i U_k(i)$, where $U_k(i)$ refers to the $i$th unobserved transition. Radiation of multipolarity $L+1$ often competes with radiation of multipolarity $L$. For

$$F'_k = \frac{F_k(L_2 I_B C) + \delta^2 F_k(L_2 I_B C) + 2F_k(L_2 I_B I_B C)}{1 + \delta^2},$$

\[(II.6)\]
where $\delta^2$ is the intensity of the $L_2$ component. $\delta$ can be positive or negative. The corresponding expression for $U_k$ does not contain the interference term (term in $\delta$) and

$$U'_k = \frac{U_k(L_k) + \delta^2 U_k(L'_k)}{1 + \delta^2}. \quad (II.7)$$

Note that $F_k$ and $U_k$ vanish for $k > 2L$. Radiation from states with spin 0 or 1/2 or of multipolarity $L=0$ will be isotropic.

In addition to reorientation effects due to unobserved transitions between the original state in which the nuclei are oriented and the state from which the observed radiations are emitted, reorientation may occur in the intermediate states. These effects are due to interactions of the nuclear states with extranuclear fields. They may be included in the theory by introducing the coefficients $Q_k$, which multiply the $k$th term in the expansion of $W(\theta)$. For $n$ intermediate states, $Q_k = \prod_{i=1}^{n} Q_k(i)$, where $Q_k(i)$ is the coefficient for the $i$th state. Although a theory is available, it is not in practice possible to calculate $Q_k(i)$ exactly. In general, $Q_k(1)$ for lifetimes much shorter than 1 nsec. Substantial attenuations have been observed for lifetimes of the order of a few nsec.\textsuperscript{17,18,19}

The Angular Distribution of Particles

The $F_k$'s have been tabulated by Ferentz and Rosenzweig.\textsuperscript{20} It is therefore convenient to consider the expression for the gamma ray angular distribution as a reference and to define coefficients which allow the calculation of the angular distribution for particles in terms of the tabulated coefficients for the angular distribution for gamma rays. This has been done by Biedenharn and Rose,\textsuperscript{21} who introduce particle parameters $b_k$ so that

$$W(\theta)_{\text{particle}} = 1 + \sum_k B_k b_k U_k F_k (\cos \theta). \quad (II.8)$$

The particle parameters for alpha decay and conversion electron emission are discussed in further detail in Secs. (III) and (VI) respectively.
Angular Correlation

Angular correlation can also be used to determine nuclear spins and multipolarities of radiations in nuclear transitions. Consider two successive gamma rays of multipolarity $L_1, L'_1$ and $L_2, L'_2$ emitted in a nuclear cascade, $I_A \rightarrow I_B \rightarrow I_C$. If coincidences between $\gamma_1$ and $\gamma_2$ are measured in two detectors, the coincidence rate may be found to be a function of the angle $\theta$ between the detectors. The theoretical correlation function is similar to that for the distribution of radiation from oriented nuclei, and angular correlation may be thought of as a special case of nuclear alignment. In this case, an alignment of $I_B$ is obtained by choosing as the axis of orientation the direction of emission of $\gamma_1$. Then $\gamma_2$ may show spatial anisotropy with respect to the axis. The theory has been reviewed by Frauenfelder and others, and the theoretical expression is:

$$W(\theta) = 1 + \sum_{k_{even}} A_k F_k(\cos \theta), \quad (\text{II}.9)$$

where $A_k$ is a product of $F$ coefficients (Eq. II.4), one of which involves $\gamma_1$ only and the other of which involves $\gamma_2$ only. Thus,

$$A_k = F'_k(L_1 L'_1 I_{AB}) F'_k(L_2 L'_2 I_{CB}).$$

Note that the coefficients are defined as if both $\gamma$-rays originated at $I_B$. If $\gamma_1$ is observed from oriented nuclei, the coefficient $F'_k(L_1 L'_1 I_{AB})$ is used, while for $\gamma_1$ correlated to $\gamma_2$, $F'_k(L_1 L'_1 I_{AB})$ is used, because $I_B$ is the aligned state in angular correlation. In addition, the sign of $\delta$ changes. If $\delta$ is measured positive for nuclear alignment, it will be negative for angular correlation. This does not apply to $\gamma_2$, because the same $F_k$ is used for both nuclear alignment and angular correlation.
B. **Experimental**

1. **Germanium Surface-Barrier Detectors**

Measurements of the angular distribution of particles from oriented nuclei were made using germanium surface-barrier detectors, similar to those used by Roberts and Dalbs and Navarro for alpha particle detection and by Westenbarger for electron detection. Their characteristics were first investigated in detail by Walter et al.

The detectors' most important advantages are their ability to count at 10 K, and that they may be placed inside the cryostat.

**Preparation.** Several methods of fabrication were tried. The most successful is given in detail below.

1. Germanium wafers were cut from a bar of 10 ohm-cm n-type Ge. The wafers were lapped with No. 1900 silicon carbide lap compound and etched in a CP-4 bath (2 parts HNO₃, 1 part HF, 1 part acetic acid).

2. A thick coating of gold was evaporated onto one (back) side of the wafer and was alloyed with the Ge by heating to above 325°C.

3. A thin sheet of gold (500-1000 Å) was evaporated onto the other (front) side of the wafer. In both evaporation, care was taken to keep the gold away from the sides of the wafer. A thin gold wire was attached to the gold sheet on the front surface by thermal-compression bonding.

4. The wafer was attached to a molybdenum tab using thermal-setting epoxy resin, made conducting with gold (from E. I. DuPont and Co.). The thin gold lead was attached to a B.S. No. 30 manganin wire with the same epoxy resin. The assembled detector was placed in a furnace at 160°C for a few hours to cure the resin.

5. Before use, the detector was washed with deionized water, methyl alcohol, and trichloroethylene, and dried in dry N₂ gas.

The detectors made in this way were good diodes at temperatures below 77 K and capacitance measurements showed a linear dependence on the square root of the applied bias. This is consistent with surface-barrier behavior, in which increasing bias results in an increased depletion region. These relationships are shown in Figs. II.1 and II.2. An alpha-
Fig. II.1. Dependence of capacitance and pulse height on bias for a germanium surface-barrier detector at 4.2°K.
Fig. II.2. Current as a function of applied bias for a germanium surface-barrier detector at 77°K and 4.2°K.
particle spectrum taken at 1°K is shown in Fig. II.3. The source was Am$^{241}$ and Cm$^{244}$. The variation of the pulse height due to Am$^{241}$ alphas as a function of applied bias is shown in Fig. II.1. The detector response to electrons at 1°K is illustrated by the spectrum shown in Fig. II.4. The source was Ce$^{137}$ on neodymium ethylsulfate, and was one of the sources used in the conversion electron experiment described in Sec. III.A. In addition, a detector made 1-cm thick was found to detect the 123 and 137 keV γ-rays from Co$^{57}$ (in a source placed outside the cryogenic system) with 12 keV resolution at 77°K. The peaks did not appear at 1°K.

The detectors showed anomalous behavior at certain times. The optimum bias setting (for the best resolution) was from -5 to -30 V and would change from day to day for the same detector. Often, particles would be detected with no bias at all, indicating a polarization phenomenon. Under experimental conditions, they sometimes exhibited infuriating vagaries. The pulse height was sometimes affected by introducing He gas into the cryostat. Or, the pulse rate might effect the pulse height. The most insidious change, however, was a change in resolution at constant pulse height as was sometimes observed in the conversion-electron experiments. This made analysis very difficult and subject to errors. Experimental runs showing changes in pulse height of resolution were generally discarded.

Other types of detectors were also made and tested. Germanium detectors with aluminum front and back surfaces or detectors with one gold and one aluminum surface were tried but were unsuccessful. Pulses were observed, but resolution was poor. Silicon detectors were also tried. A commercially available lithium-drifted detector showed good resolution at 1°K, but the pulse height drifted with time, indicating trapping of change in the depletion region, and thus lowering the electric field. This effect is illustrated in Fig. II.5. Detectors made of 10,000 ohm-cm silicon according to the germanium detector recipe also showed good resolution at 1°K, but had poor stability. They are, however, very promising because they can withstand much higher applied bias than Ge (up to 900 V) at 1°K.
Fig. II.3. Alpha particle spectrum of a mixed Am$^{241}$-Cm$^{244}$ source taken with a germanium surface-barrier detector at 1$^\circ$K.
Fig. II.4. Conversion electron spectrum of the 255 keV transition in Ce$^{137m}$ taken with a germanium surface-barrier detector at 10K.
Fig. II.5. Cs$^{137}$ conversion electron spectra taken with a TMC lithium-drifted silicon detector at 1 K, showing polarization phenomenon.
The pulses from the detectors were amplified with special
UCRL 5V1012 charge-sensitive preamplifiers and UCRL 5V1034 (model V
modified) linear amplifiers with R-C pulse shaping. The amplified
pulses were then fed into a TMC model 213-multi-channel pulse height
analyzer or into a RIDL 400-channel analyzer. These analyzers auto­
cationally correct for analyzer dead time. In later experiments, Goulding­
Landis 11 × 2950 P-1 pre-amplifiers and 11 × 198 OP-1 linear amplifiers
were used.

**Gamma Ray Detectors**

Measurements of the angular distributions of γ-rays from oriented
nuclei and spectroscopy measurements were made with NaI(Tl) crystals and
with the newly developed lithium-drifted germanium crystals.

The NaI(Tl) scintillation crystals were cylindrical 3 × 3-in.
or 2 × 2-in. and were mounted on photomultiplier tubes. They were obtained
from the Harshaw Chemical Company. The voltage pulse from the photo­
multipliers went into cathode followers and then into linear amplifiers
with double delay line pulse shaping. The amplifier output went to a
pulse height-analyzer. The best crystal and photomultiplier had 7.8%
resolution at 600 keV. For angular correlations and conversion electron
studies, the Goulding-Landis amplifier was used.

The introduction of lithium-drifted germanium\(^{30}\) as a detector
for gamma rays has constituted a major step forward in gamma-ray spec­
troscopy. They were first made at Atomic Energy of Canada, Ltd. labora­
tories at Chalk River, Ontario, but a considerable development effort
has been made by the Lawrence Radiation Laboratory.\(^{31,32,33}\)

Lithium is evaporated onto the surface of 30 ohm-cm p-type
germanium and is diffused into a depth of several microns. The lithium
is a interstitial donor is germanium. Under the influence of an applied
electric field, and at temperatures of the order of 120°C, the Li drifts
into the p-type material and compensates the acceptors in the region.
This results in an intrinsic zone where the acceptors equal the donors.
The depth of the zone can be controlled by the drifting field and the
ambient temperature. At the present state of the art, depletion zones
of one centimeter depth are obtainable. These require 10 days of drift­
ing at 1000 volts and 100°C.
The zone will support potentials of 150 to 900 volts. Gamma rays produce photo- or Compton-electrons in this region which in turn create electron-hole pairs. These pairs are swept out of the depleted region by the electric field, giving a voltage pulse proportional to the original energy of the γ-ray. Because the average energy needed to create an electron-hole pair is only 2.8 keV, the statistical uncertainty in a 100 keV γ-ray is

\[ \frac{\sqrt{105/2.8}}{10^5/2.8} \approx 0.5\%, \text{ or } 0.5 \text{ keV}. \]

This may be compared with NaI(Tl) for which the average excitation energy for electrons into the conduction band is 10 eV. The statistical fluctuation in pairs created for a 100 keV γ-ray is now \[ \frac{\sqrt{105/10}}{10^5/10} \approx 1\%. \] In a scintillation counter however, this is not the only statistical consideration. The signal is broadened by statistical fluctuations in photon production, photon collection at the photo-cathode, electron emission by the photo-cathode and electron emission at each dynode. Non-uniformity of the photo-cathode also limits resolution.

Since the primary electron-hole pairs are collected directly in the semiconductor detector, the resolution limiting statistical fluctuations found in NaI + photo-multiplier are absent. The present limit to resolution is primarily noise in the preamplifier.

The detector resolution varies inversely with the detector capacity because for a given charge collected \( Q \), \( V = \frac{Q}{C} \), and this limits the surface areas. An unfortunate aspect of the Li-Ge system is the tendency for the Li to precipitate out of solid solution. To avoid this, it is necessary to keep the detectors at reduced temperatures (generally of the order of 77°K for operation). In addition, the detectors are very sensitive to surface contamination, presumably because of surface leakage current. Holders which keep the detectors at 77°K and in vacuum have been devised. They are also designed to minimize stray capacitance between the preamplifier input and ground. A schematic diagram of the holder is shown in Fig. II.6.
Fig. II.6. Holder for Ge(Li) detectors.
The preamplifier used with these detectors is an 11 x 2950 P-l with an Amperex EC 1000 tube as the first stage. A schematic diagram is shown in Fig. II.7. The linear amplifier is a 11 x 198 OP-l. It can be used with positive or negative input pulses, has provisions for delay line, double delay line and RC clipping, and has a biased amplifier in the same chassis. A single channel analyzer and a slow and fast coincidence system with a variable delay can also be added to the chassis. The best resolution obtained with a germanium detector and these electronics is 2 keV at 100 keV FWHM. This is also the FWHM of a pulser signal passed into the preamp through a known capacitance and of a size to give the charge equivalent of a 100 keV pulse in germanium. Numerous spectra are shown in Sec. V. The resolution decreases to 4 keV at 1 MeV.

Because of the small volumes the efficiencies of these detectors are fairly low. The relative efficiency as a function of energy has been measured by Easterday, Haverfield and Hollander, and their curve is shown in Fig. II.8. The efficiency follows the photoelectric cross section in Ge to a minimum at about 1 MeV. At higher energies, the pair-production cross section increases and the efficiency increases as well. An illustration of pair production with single and double escape is shown in the spectrum in Fig. II.9. Calculations of the efficiency have been made by Goulding. Because of the reduced efficiency at high energy and the low intensity of some of the transitions studied, counting runs of up to 30 hours were required. These were possible due to the remarkable stability of the electronics.

The Li-drifted Si Detectors

Measurements of relative conversion electron intensities were made using lithium-drifted silicon detectors at 77°K. The same electronics as used for the lithium-drifted germanium detectors were used here. Conversion coefficient measurements were made using a lithium-drifted germanium γ-ray detector and a lithium-drifted silicon electron detector placed in an evacuated chamber to simultaneously measure the γ-rays and conversion electrons for a given transition. The system was calibrated using sources with transitions for which the conversion coefficients were known. This is described in detail in a report by Easterday, Haverfield, and Hollander.
Fig. II.7. Schematic for preamp used with semiconductor detectors.
Fig. II.8. Relative efficiency curve for Ge(Li).
Fig. II.9. Na$^{24}$ 2754 keV γ-ray showing single and double escape from a Ge(Li) detector.
2. The Nuclear Orientation Apparatus

The apparatus used for the nuclear alignment experiments is similar to those described elsewhere, but was modified to allow particle detection inside the cryostat. The experimental chamber was an adiabatic demagnetization cryostat of Pyrex glass attached to a copper-Pyrex seal. The copper seal was soft-soldered to a brass skirt at the end of a 1.5-in. stainless steel tube with a 0.020-in. wall. This seal was broken when access to the experimental chamber was desired. The stainless steel tube was suspended from the top of a brass support and was connected by a welded side arm at the top to an MCF 60 oil-diffusion pump and forepump capable of pumping the cryostat to $10^{-6}$ mm Hg. A valve in the line allowed the introduction of He exchange gas into the cryostat. The top of the stainless steel tube was capped with a plate containing kovar steel to glass seals, to which were attached the B.S. 36 magnum wire signal leads from the detectors in the cryostat. The wires ran up the center of the tube, which was an wide and as short as possible to minimize stray capacitance. The brass skirt at the bottom of the tube included a radiation shield with feed-throughs for the signal leads and a half-inch bypass pumping tube. The cryostat was inserted into a double glass walled helium dewar which in turn was inside a double glass-walled liquid nitrogen dewar. The dewars were silvered except for half-inch windows and were tailed down to fit into the electromagnet. The helium dewar was connected to an 8-in. diameter pumping line, which terminated at a KMBV 1250 Kinney vacuum pump, capable of pumping the helium bath to 0.97°K. The gas pressure above the bath was monitored with a mercury manometer, dibutyl phthalate manometer, and a McLeod gauge. Pictures of the apparatus are shown in Figs. II.10 and II.11.

The adiabatic demagnetizations were carried out using an iron core electromagnet capable of 22 kilogauss across a 2.626-in. gap. The magnet was mounted on wheels which fitted tracks in the floor, and had a yoke which permitted horizontal access to the gap. It was rolled up around the dewars for magnetization and rolled back upon demagnetization.
Fig. II.10. Schematic of Nuclear Alignment apparatus, used for particle anisotropy studies.
Fig. II.11. Picture of Nuclear Alignment apparatus, showing Ge(Li) detector holder and associated electronics.
3. The Cryostat

A schematic representation of the experimental chamber used for particle angular distribution studies is shown in Fig. II.12. A picture is shown in Fig. II.13. A brass cage inside the cryostat was used to support the detectors. It was attached by screws to the radiation shield inside the stainless steel pumping tube. The paramagnetic crystal was suspended by a glass rod from the top of the cage. The glass rod was anchored at the top by a tungsten-glass joint and was tied to the cage half-way down with nylon threads to damp out vibrations. Guard salts of chrome alum-glycerin slurry and manganous ammonium sulfate were placed about the crystal to minimize heat flow down the rod and to trap any gas that might come down from the pumping tube. The detectors and collimators were mounted at 0° and 90° to c axis of the crystal, with axes normal to the center of their counting areas intersecting at the position of the radioactive spot (see Sec. III.A for details of the sample preparations). The cage provided a grounding point for the detectors. To eliminate ground loops, the brass head of the apparatus was isolated electrically from the main pumping line. After the experiment was assembled, the cryostat was soldered to the pumping tube. It was then painted with colloidal graphite (Aqua-dag) to reduce the thermal radiation falling on the crystal. The magnetic susceptibility coils were then put on and placed so that one primary-secondary pair was around the crystal and the other pair was around an empty (of paramagnetic salt) portion of the cryostat. Care was taken to center the guard salts between the two pairs of coils.

For experiments in which the angular distribution of γ-radiation only was measured, a similar arrangement was used, only the detectors and their leads were omitted.
Manganous ammonium sulfate pill (0.14°K)

He 1°K

Chrome alum-glycerin slurry (0.01°K)

Ce$^{137m}$ or E$^{253}$ activity

e- or α detectors and collimators (1°K)

NES crystal (0.02 to 0.1°K)

Glass cryostat

Brass cage

Fig. II.12. Cryostat arrangement for conversion electron and alpha particle anisotropy studies. NaI counters are mounted outside the dewar system at 0° and 90° to the crystalline c axis.
Fig. II.13. Cage assembly, cryostat, and mutual inductance coils used in particle anisotropy studies. Detectors are shown at right.
4. Temperature Measurements

The temperature of the paramagnetic crystals used in the nuclear alignment experiments was determined by measuring the magnetic susceptibility \( X \) and using Curie's Law,

\[
X = \frac{C}{T}
\]

where \( C \) is the Curie constant. This relation is valid only for temperatures such that \( kT > U_{\text{co}} \), where \( U_{\text{co}} \) is the energy due to cooperative effects such as magnetic dipole or exchange interactions between the ions in the crystal. \( U_{\text{co}} \) also determines the lowest temperature to which a paramagnetic crystal may be demagnetized, and therefore Curie's Law may be expected to be approximately valid over most of the temperature range from 1° to the lowest obtainable temperature. Because of the low temperature deviations of Curie's Law, it is necessary to correlate the temperature derived from susceptibility measurements, designated \( T^* \), with the absolute temperature, \( T \). Thus we define:

\[
T^* = \frac{C}{X}.
\]

Applying magnetic field \( H \) to a paramagnetic substance results in a net magnetization \( M \), where

\[
X \lim_{H \to 0} \frac{M}{H}
\]

\( M \) may be calculated from the partition function \( Z \),

\[
M = [\partial kT \ln Z / \partial H]_H
\]

with \( H \) defined as the field acting on the ions in the sample. It is necessary to consider the relation between the externally applied field, \( H_{\text{ext}} \), and the field at the ions, \( H \).
H_{\text{ext}} may be defined as the field that results when a current is passed through an empty solenoid. If a paramagnetic substance is placed in the solenoid, the field in the substance, H_{\text{int}}', will be homogeneous only if the sample shape is ellipsoidal.\textsuperscript{37} For an ellipsoidal sample,

\[ H_{\text{int}} = H_{\text{ext}} - \frac{NM}{V} \]  

(II.12)

where M/V is the magnetic moment per unit volume and N is the demagnetization factor. N, as a function of the ellipsoidal shape, is given by Kurti and Simon.\textsuperscript{38} For a sphere, N = 4π/3. The field at a given ion will also depend on the magnetic interaction with neighboring ions. Lorentz\textsuperscript{39} calculated the field due to the interaction to be proportional to the magnetization,

\[ H = H_{\text{int}} + \frac{4}{3} \pi \frac{M}{V} \]  

(II.13)

and using Eq. (II.12),

\[ H = H_{\text{ext}} + \frac{4}{3} \pi \frac{M}{V} - N \frac{M}{V} \]  

(II.14)

Onsager\textsuperscript{40} tried to include the effect of the ion polarizing the surrounding medium. Van Vleck\textsuperscript{41} used the actual magnetic interactions between ions and developed a series expansion which converged slowly at low temperature. These calculations are all unsatisfactory at low values of T. At higher values of T, all three treatments converge to the same result. Thus, H_{\text{int}} can be related exactly to H_{\text{ext}}, but there is an uncertainty in going to H (at the ions). In defining T* then, we use H_{\text{ext}}' so that T* = C/X_{\text{ext}} where X_{\text{ext}}' is defined as \( \lim_{H_{\text{ext}} \to 0} \frac{M}{H_{\text{ext}}} \). This choice means that T* (at a given T) will depend on the sample shape. To facilitate the use of the T*-T correlations for a given substance, Kurti and Simon\textsuperscript{38} introduced T\textsuperscript{®}, the T* for a spherical sample. The relation between T\textsuperscript{®} and T* may be derived as follows:
Let

\[ T^* = \frac{C}{X} \]  \hspace{1cm} (II.15)

where \( X \) is the susceptibility using the field at the ions \( H \):

\[ H = H_{\text{int}} + H_m \]

with \( H_m \) the field due to the other dipoles. Then

\[ H = H_{\text{ext}} - \frac{NM}{V} + H_m \]  \hspace{1cm} (II.16)

\[ X = \frac{M}{H} \left( \frac{M}{H_{\text{ext}} - \frac{NM}{V} + H_m} \right) \]

\[ = \frac{X_{\text{ext}}}{1 - \frac{N}{V} X_{\text{ext}} H_m/H_{\text{ext}}} \]

Using Eq. (II.15),

\[ T^* = \frac{C}{X} = \frac{C}{X_{\text{ext}}} \left[ 1 - \frac{N}{V} X_{\text{ext}} + \frac{H_m}{H_{\text{ext}}} \right] \]  \hspace{1cm} (II.17)

\[ = T^* + \left[ \frac{C H_m}{M} - \frac{N C}{V} \right] \]

If there are two samples of different shape, they will have a different \( N \), but \( T^* \) would be the same for each (by definition). Then we find

\[ T^*_2 = T^*_1 + \frac{C}{V} \left[ \left( \frac{H_m}{M} \right)_1 - \left( \frac{H_m}{M} \right)_2 \right] + (N_2 - N_1) C/V \]  \hspace{1cm} (II.18)

The effect of dipoles far away from a given dipole is included in \( N \). \( H_m \) may be considered as arising from nearby dipoles, and \( H_m/M \) will be independent of sample shape. Therefore,

\[ T^*_2 = T^*_1 + (N_2 - N_1) C/V \]  \hspace{1cm} (II.19)
If sample 2 is a sphere, \( T^*_{2} = T_{\infty} \) and \( T_{\infty} = T_{1}^{*} + \left( \frac{4\pi/3 - 0}{C/V} \right) \). To estimate the magnitude of the effect of shape, let \( g = 2 \), \( J = 1/2 \), and \( V \approx 250 \) c.c. (volume per mole). The \( C/V \approx 0.002 \). For a flat, thin, disk-shaped sample with the measuring field along the plane of the disk, \( N = 0 \).

\[
T_{\infty} = T_{*} + \left( \frac{4\pi/3 - 0}{0.002} \right) = T_{*} + 0.008
\]

The susceptibility in these experiments was measured using mutual inductance coils and a Hartshorn bridge. Two primary-secondary pairs were used, connected in opposition. One pair was located around the paramagnetic crystal and the other was placed where the cryostat contained no paramagnetic material. If \( \Phi \) is the flux in the secondary, the voltage induced is \( \frac{d\Phi}{dt} \). For the coil around the crystal, \( \Phi = (H + 4\pi M) nA \) where \( A \) is the area of the secondary and \( n \) is the number of turns. For the other coil, \( \Phi = HnA \).

The difference is \( (4\pi M) nA \). The primary voltage is modulated with a 20 c/s audio oscillator. Changes in the difference voltage are directly proportional to changes in the susceptibility.

The difference voltage is amplified and measured using a variable inductance bridge, with an oscilloscope as a null point indicator. The bridge also balances the voltage induced in phase with the primary voltage due to the imaginary part of the susceptibility. A circuit is shown in Fig. II.14.

The bridge and coils were calibrated for each sample in the temperature region \( 4^\circ K \) to \( 1^\circ K \), using the vapor pressure of \( \text{He}^2 \) gas as a measure of the absolute temperature. An example of such a calibration
Fig. II.14. Circuit for mutual inductance measurements.
curve is shown in Fig. II.15. Upon demagnetization, the bridge was balanced and periodically rebalanced as the crystal warmed up (due to stray heat input). Extrapolation of the calibration allowed $T^*$ to be calculated from the bridge reading.

$T^\Theta - T$ correlations are based on the second law of thermodynamics. From

$$ds = dQ/T,$$

$$T = \left(\partial Q/\partial T^\Theta\right) \left(\partial s/\partial T^\Theta\right).$$

$\partial Q/\partial T^\Theta$ may be measured by using a constant heat input (e.g., $\gamma$-ray heating) and measuring the change in $T^\Theta$. $\partial s/\partial T^\Theta$ may be measured by a series of isentropic demagnetizations of a spherical sample from $1^0K$ and a known field, $H$ (initial). $S(H/T)$ may be calculated or measured from the heat output of the crystal upon magnetization. If the magnetic properties of the crystal are anisotropic, the correlation will depend on the direction of the magnetizing field and of the measuring field. The most difficult problem is obtaining a reliable correlation at the lowest values of $T$ obtainable by demagnetizing the sample. The largest deviations from Curie's Law occur at these temperatures. For collective transitions in paramagnets, the susceptibility shows a maximum in the collective region. Thus, $\partial s/\partial T^\Theta$ becomes very large, and large uncertainties result in $T$. The techniques and methods of these determinations are further explicated in the reviews by DeKlerk and by Ambler and Hudson.

The $T^\Theta - T$ correlation for cerium magnesium nitrate has been made by Daniels and Robinson and is discussed in Sec. IV.A. Meyer has made the correlation for neodymium ethylsulfate, which is the subject of Sec. III.B.

In general, the crystals used in this work were single crystals and were not shaped into ellipsoids or spheres for which $N$ could be calculated. The measured $T^*$'s were related to $T^\Theta$ empirically by defining correction $S$, such that, $T^\Theta = T^* + S$. For neodymium ethylsulfate $S$ was
Fig. II.15. Typical calibration curve (neodymium ethylsulfate) for magnetic temperature (\(T^\theta\)) measurements.
determined by comparing $T^*$'s obtained for demagnetizations from various ($H/T$) (initial) with the $T^*$'s obtained by Meyer for the same ($H/T$) (initial). For cerium magnesium nitrate, Daniels and Robinson found $T^* = 312$ for ($H/T$) (initial) $> 18kG/ok$. $T^*$'s for demagnetizations from 18 to 22kG/ok were set equal to 312, giving 5. for this salt.

5. Experimental Procedure

In the most general nuclear alignment experiment, the angular distributions of $\gamma$-radiation and particles were measured and the temperature was monitored with the magnetic susceptibility coils at the same time. The $\gamma$-ray detectors (NaI or Ge(Li), see Sec. II.B.1) were mounted on a counting table which rolled on tracks in the floor. (See Fig. II.11) The detectors were placed at $0^\circ$ and $90^\circ$ with respect to the direction of the crystalline $c$ axis of the cooling crystal. The crystal was magnetized at 10K and in fields up to 23 kilogauss with 0.08mm Hg of He exchange gas in the cryostat. The cryostat was then pumped out to $10^{-6}$ mm Hg. The crystal was demagnetized over a period of about one minute and the magnet was rolled away. The counting table was rolled up and centered with respect to the position of the crystal. Multichannel analysis was started for all four detectors and the magnetic susceptibility was measured using mutual inductance coils. These counts constituted the "cold" counts. After counting for a measured length of time, the counts were recorded and 0.08 mm Hg of He gas was introduced into the cryostat. This allowed thermal contact with the bath and the crystal warmed to 10K where the radiation was isotropic. Normalizing "warm" counts were then taken with all four detectors. The counts were corrected for source decay and background and the ratios (counts cold/counts warm) (=$c/w$) were taken. The temperature measurements were corrected from $T^*$ to $T$ (Sec. II.B.4). This gave $c/w = W(\theta)$ as a function of temperature for $\theta=0^\circ$, 90$^\circ$. These data were then fitted with theoretical curves (Sec. II.A).
6. Coincidence Studies and Angular Correlations

Germanium-germanium, germanium-\(\text{NaI(Tl)}\), and \(\text{NaI(Tl)}-\text{NaI(Tl)}\) coincidence studies were performed using the fast-slow coincidence circuitry in the Goulding-Landis amplifiers. The fast system had a variable delay of up to 450 nsec and a variable resolving time of 10-110 nsec.

The pulses used for the fast coincidence were taken off the crossover in the first amplifying stage. This was done by a circuit in the amplifier. A block diagram of the coincidence circuit is shown in Fig. II.16. In the angular correlations experiments involving \(\text{NaI(Tl)}\) detectors, Cosmic Radiation Company's "Spectrostats" were used as high voltage supplies.

These devices adjusted the high voltage to keep a constant pulse height and this tended to eliminate drifting.
Fig. II.16. Block diagram of coincidence circuit. S, source; D, detector; H.V., high voltage; PA, preamp; LA, linear amp; F.C., fast coincidence; SCA, single channel analyzer; BA, biased amplifier; PHA, pulse height analyzer.
III. THE ANGULAR DISTRIBUTION OF CONVERSION ELECTRONS FROM Ce$^{137m}$ ORIENTED IN NES

A. Conversion Electron Experiments

1. Introduction

The angular distribution of beta particles from oriented nuclei is of interest in the study of both nuclear and solid state phenomena. Parity non-conservation in beta decay was first proved using nuclear orientation.$^{47}$ Studies of the matrix elements in first forbidden beta decay have also used this technique.$^{48}$ Recently, determinations of the signs of magnetic fields at impurities in ferromagnets have been made using beta asymmetries from oriented nuclei.$^{26}$

Previous detection techniques involved plastic scintillators and light pipes. These are unsatisfactory because of poor energy resolution and poor cryogenic characteristics. Development of the Ge surface barrier detectors (Sec. II.B.1) to allow electron counting at 1°K with good resolution increases the feasibility of these experiments.

In order to establish the quantitative accuracy of measurements made with the germanium electron detectors it is advantageous to study a case in which the electron energy spectrum is discrete, rather than continuous, and for which a quantitative theoretical prediction exists. The experiment chosen was the measurement of the angular distribution of conversion electrons from the 255 keV isomeric transition in Ce$^{137m}$. This experiment, the first of its kind, was free from ambiguity in interpretation and allowed a direct determination of a particle parameter for conversion electrons. When aligned in NES, for which the temperature-susceptibility correlation is known,$^{46}$ Ce$^{137m}$ had been shown to give large $\gamma$-ray anisotropies,$^{49}$ and the 255-keV isomeric transition is highly converted ($ek/\gamma \approx 6$).$^{50}$ The existing decay scheme is shown in Fig. II.1.$^{51}$ Many new transitions and several errors have been discovered in this decay scheme. The results are presented and discussed in Sec. V. It should be noted that the changes have no effect on this experiment.
Fig. III.1. Existing decay scheme for $\text{Ce}^{137m}$. 

$\text{Ce}^{137m}$ decay scheme
2. Experimental

A schematic representation of the demagnetization cryostat was shown in Fig. II.12. Electron detectors and collimators are mounted at 0° and at 90° to the crystalline c axis of a neodymium ethylsulfate (NES) crystal containing Ce$^{137m}$ activity. The c axis is the axis of orientation. NaI(Tl) counters were mounted at 0° and 90° outside the cryostat and dewars. The conversion electron spectrum taken at 1°K with an electron detector was shown in Fig. II.4, with the K and (L+M) peaks clearly resolved.

Thirty-four hour Ce$^{137m}$ was made from natural La$_2$O$_3$ by the reaction La$^{139}$(p,3n), using 30 MeV protons in the Berkeley 88-in- cyclotron. This energy was below the threshold of the La$^{139}$(p,5n)Ce$^{135}$ reaction. Ce$^{139}$ was produced, but had no interfering radiations. A Ce-La chemical separation was made by solvent extraction of Ce$^{4+}$, following Glendenin,$^5$ giving reasonably mass-free Ce$^{3+}$. The procedure is:

1. The target was dissolved in a minimum amount of conc. HNO$_3$. A few cc of 1M KBrO$_3$ in 10M HNO$_3$ (oxidant) was added to oxidize the Ce$^{3+}$ to Ce$^{4+}$.

2. The solution was placed in a separatory funnel with methylisobutyl ketone (MIBK), pre-equilibrated with the oxidant. The funnel was agitated vigorously for a few minutes. The phases were allowed to separate.

3. The aqueous (lower) phase was drawn out of the funnel. A few cc of 8M HNO$_3$ containing 2 drops of the oxidant were added and the funnel agitated. After the phases had separated, the organic (upper phase) was drawn off with a pipet.

4. It was placed in another separatory funnel containing an equal volume of 1.5M H$_2$O$_2$ in 8M HNO$_3$ (reductant). The funnel was agitated, the aqueous phase drawn off the bottom. The organic phase was discarded.

5. The aqueous solution was evaporated to dryness (slowly!). The residue was taken up in a few drops of conc HCl and evaporated to dryness several times. It was then taken up in 0.1M HCl and transferred to a small Dowex 50 anion exchange column, where it was washed with 30 column
volumes of 0.1M HCl, 2 cc of 2M HCl. The $\text{Ce}^{3+}$ was stripped off the column with 6M HCl, and evaporated to dryness.

For particle measurements, a source with minimum scattering and absorption is required. A large NES crystal was selected and sanded to produce a face at 45° to the axis. The surface of this face was dissolved away with water to remove damage caused by the sanding. The $\text{Ce}^{137m}$ activity was taken up in small drop ($10^{-4}$ cc) of water with a micro pipet and was deposited on an area 1 mm$^2$ on this face. The drop was removed and replaced several times without being allowed to evaporate. In this way $\text{Ce}^{3+}$ replaced some $\text{Nd}^{3+}$ in the lattice. The crystal was then mounted in the cryostat (see Fig. II.13) with the active area on the line of intersection of the detector axes. The surface deposition of the cerium activity provides a "thin" source in which absorption and scattering are small. The energy resolution of the detectors also reduces the detection of scattered electrons.

3. Measurements and Results

Upon demagnetization of the NES crystal from magnetic fields up to 23 kgauss at 0.97°K, the $\gamma$-ray and conversion-electron intensities were measured at both 0° and 90° to the alignment axis as the crystal warmed for about 1.5 hours. (We note that small temperature inhomogeneities in the source are of little consequence here as we were measuring the relative anisotropies of electrons and $\gamma$-rays. In the steady state the average temperature of the radioactive spot was about 0.01°K higher than that of the bulk crystal.) Heat-exchange gas was then admitted, warming the crystal to 0.97°K, and the $\gamma$-ray and electron intensities from the "warm" unaligned nuclei were measured for normalization. "Cold" and "warm" electron and $\gamma$-ray spectra from the 0° counters are shown in Fig. III.2. After small corrections were made for source decay and background, the normalized intensities, $W(\theta)$, were calculated for the 255 keV $\gamma$-ray peak, the 215 keV K-electron peak, and the (L+M)-electron peak at 249 keV. The results obtained from the 90° data were in good agreement with those from the 0° data. However, the 90° counter gave poorer
Fig. III.2. Simultaneous counts of equal duration at 0.020K (cold) and 10K (warm) of (a) the conversion electrons and (b) the \(\gamma\)-rays, from the 255 keV transition, at 0° to the c axis. For comparison the warm counts have been scaled to give the same peak count.
resolution and results of lower statistical accuracy. The poorer resolution required larger background corrections, entailing possible systematic errors. For this reason the 0° results alone were used to derive particle parameters. The 0° data are shown in Table I.

In Figs. III.3 and III.4 1-W(0), for each electron peak, is plotted against 1-W(0)_γ. Figures III.5 and III.6 show the ratios [1-W(0)_K]/[1-W(0)_γ] and [1-W(0)_K/(L+M)])/[1-W(0)_γ] as functions of 1-W(0)_γ. Note that the determination of the ratios is in no way dependent on knowledge of the temperature scale for NES. It is necessary for the interpretation, however, that there should be no gross temperature inhomogeneities over the 1 mm² active area. The long "warm-up" times of hours and large γ-ray anisotropies are excellent evidence that this criterion is satisfied.

4. Discussion

The general expression for the observed angular distribution of conversion electrons following the decay of oriented nuclei was introduced in Sec. II.A,

\[ W(θ) = \sum_{k \text{even}} B_k U_k Q_k g_k h_k F_k P_k (\cosθ) \]  

(III.1)

The \( b_k \) are "particle parameters" introduced in Sec. II.A which modify \( F_k \) for the observed transition. For γ-rays, and for all radiations (of pure or mixed multipolarity) in the high energy limit, \( b_β = 1.53 \). The particle parameters for conversion electrons may be substantially different from unity, especially for low energies and/or multipolarities.\(^5\) In contrast to γ-rays, the electrons are spin-1/2 particles and are ejected from discrete atomic (or molecular) orbitals. They undergo a phase shift on leaving the electromagnetic potential of the atom. Biedenharn and Rose have tabulated particle parameters for k conversion electrons ejected from relativistic atomic orbitals, using the point-nucleus approximation.\(^2\) For a 255 keV M4 transition in cerium (element 58) their theoretical \( b_2 \) is 1.055. Church et al.\(^5\) have pointed out a sign error in the theory. This error does not affect the results reported here.
Table I. Anisotropy of conversion electrons from Ce$^{137m}$ in NES.

<table>
<thead>
<tr>
<th></th>
<th>Experimental Data</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>[1-W(0)$_{\gamma}$]</td>
<td>[1-W(0)$_{K}$]</td>
<td>[1-W(0)$_{L+M}$]</td>
<td>[1-W(0)$_{\gamma}$]</td>
<td>[1-W(0)$_{K}$]</td>
<td>[1-W(0)$_{L+M}$]</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>0.500</td>
<td>0.514</td>
<td>0.531</td>
<td>22</td>
<td>0.530</td>
<td>0.599</td>
<td>0.594</td>
</tr>
<tr>
<td>2</td>
<td></td>
<td>0.412</td>
<td>0.465</td>
<td>0.474</td>
<td>23</td>
<td>0.562</td>
<td>0.546</td>
<td>0.560</td>
</tr>
<tr>
<td>3</td>
<td></td>
<td>0.398</td>
<td>0.437</td>
<td>0.443</td>
<td>24</td>
<td>0.535</td>
<td>0.535</td>
<td>0.543</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td>0.377</td>
<td>0.420</td>
<td>0.414</td>
<td>25</td>
<td>0.496</td>
<td>0.500</td>
<td>0.490</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td>0.360</td>
<td>0.415</td>
<td>0.400</td>
<td>26</td>
<td>0.455</td>
<td>0.484</td>
<td>0.485</td>
</tr>
<tr>
<td>6</td>
<td></td>
<td>0.332</td>
<td>0.381</td>
<td>0.372</td>
<td>27</td>
<td>0.433</td>
<td>0.452</td>
<td>0.449</td>
</tr>
<tr>
<td>7</td>
<td></td>
<td>0.296</td>
<td>0.338</td>
<td>0.332</td>
<td>28</td>
<td>0.429</td>
<td>0.437</td>
<td>0.438</td>
</tr>
<tr>
<td>8</td>
<td></td>
<td>0.270</td>
<td>0.301</td>
<td>0.300</td>
<td>29</td>
<td>0.365</td>
<td>0.407</td>
<td>0.405</td>
</tr>
<tr>
<td>9</td>
<td></td>
<td>0.267</td>
<td>0.296</td>
<td>0.308</td>
<td>30</td>
<td>0.363</td>
<td>0.385</td>
<td>0.394</td>
</tr>
<tr>
<td>10</td>
<td></td>
<td>0.275</td>
<td>0.305</td>
<td>0.298</td>
<td>31</td>
<td>0.349</td>
<td>0.376</td>
<td>0.370</td>
</tr>
<tr>
<td>11</td>
<td></td>
<td>0.244</td>
<td>0.309</td>
<td>0.298</td>
<td>32</td>
<td>0.348</td>
<td>0.384</td>
<td>0.378</td>
</tr>
<tr>
<td>12</td>
<td></td>
<td>0.489</td>
<td>0.521</td>
<td>0.564</td>
<td>33</td>
<td>0.315</td>
<td>0.349</td>
<td>0.340</td>
</tr>
<tr>
<td>13</td>
<td></td>
<td>0.439</td>
<td>0.472</td>
<td>0.496</td>
<td>34</td>
<td>0.335</td>
<td>0.357</td>
<td>0.340</td>
</tr>
<tr>
<td>14</td>
<td></td>
<td>0.430</td>
<td>0.467</td>
<td>0.505</td>
<td>35</td>
<td>0.330</td>
<td>0.351</td>
<td>0.345</td>
</tr>
<tr>
<td>15</td>
<td></td>
<td>0.375</td>
<td>0.435</td>
<td>0.471</td>
<td>36</td>
<td>0.250</td>
<td>0.250</td>
<td>0.220</td>
</tr>
<tr>
<td>16</td>
<td></td>
<td>0.354</td>
<td>0.385</td>
<td>0.400</td>
<td>37</td>
<td>0.220</td>
<td>0.250</td>
<td>0.256</td>
</tr>
<tr>
<td>17</td>
<td></td>
<td>0.325</td>
<td>0.350</td>
<td>0.385</td>
<td>38</td>
<td>0.198</td>
<td>0.230</td>
<td>0.241</td>
</tr>
<tr>
<td>18</td>
<td></td>
<td>0.310</td>
<td>0.327</td>
<td>0.334</td>
<td>39</td>
<td>0.202</td>
<td>0.222</td>
<td>0.212</td>
</tr>
<tr>
<td>19</td>
<td></td>
<td>0.278</td>
<td>0.324</td>
<td>0.325</td>
<td>40</td>
<td>0.210</td>
<td>0.215</td>
<td>0.188</td>
</tr>
<tr>
<td>20</td>
<td></td>
<td>0.295</td>
<td>0.345</td>
<td>0.344</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>21</td>
<td></td>
<td>0.277</td>
<td>0.280</td>
<td>0.273</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Solid Angle Corrections:

<table>
<thead>
<tr>
<th></th>
<th>$\mathcal{E}_2$</th>
<th>$\mathcal{E}_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\gamma$ detector</td>
<td>0.903 ± 0.005</td>
<td>0.700 ± 0.010</td>
</tr>
<tr>
<td>electron detector</td>
<td>0.934 ± 0.015</td>
<td>0.797 ± 0.044</td>
</tr>
</tbody>
</table>
Fig. III.3. Plot of \([1-W(0)]_K\) for K electrons vs \([1-W(0)]_\gamma\) for \(\gamma\)-rays. The solid curve is calculated using the experimentally determined value of \(b_2=1.061\). The dotted line has a slope of unity corresponding to \(b_2=1.0\).
Fig. III.4. $[1-W(O)]$ for L+M electrons plotted against $[1-W(O)]$ for $\gamma$-rays. The solid curve is calculated using the experimentally determined value of $b_2=1.059$. The dotted line corresponds to $b_2=1.0$. 
Fig. III.5. Blocked experimental values of \([1-W(O)_K]/[1-W(O)_\gamma]\) plotted against \([1-W(O)_\gamma]\). The curves are calculated using selected values of \(b_2\), and are adjusted for the different solid angles subtended by the electron and \(\gamma\)-ray detectors.
Fig. III.6. Blocked experimental values of \( \frac{[1-W(0)(L+M)]}{[1-W(0)_\gamma]} \) plotted against \([1-W(0)_\gamma]\). The curves are calculated using selected values of \( b_2 \).
Some \( b_v \) are sensitive to certain subtle features of nuclear structure. Of particular importance is the "penetration matrix element" for hindered M1 transitions.\(^{55}\) Thus it is desirable to check the theory of particle parameters by measuring several \( b_v \) with high accuracy. That the theory has not been very critically tested is evident from the fact that, until the recent work of Geiger,\(^{56}\) all the theoretical mixed-transition particle parameters in the literature had the wrong sign.

For pure multipolarities several particle-parameter measurements of 5-10\% accuracy have been reported.\(^ {57,58} \) They are set out in Table II. These measurements were made by observing the angular correlations of successive radiations and are thus less direct than nuclear orientation experiments, in which only one transition is studied. Angular correlation experiments, and those nuclear orientation experiments that involve intermediate states, are always subject to whatever uncertainty the perturbation factors \( Q_k \) entail. The effects of these two features (observation of two radiations and intermediate-state perturbation) can be eliminated by direct comparison of conversion-electron and photon intensities for the transition of interest.\(^ {57} \)

A serious difficulty encountered in a precise experimental test of the particle parameter theory is the extreme sensitivity of both \( F_2 \) and \( b_2 \) to small admixtures of higher multipolarity in the transition. Except for transitions to spin-zero states, for which angular-momentum conservation requires a unique multipolarity, it is only for very rare cases that higher multipolarity admixtures of less than a few tenths percent can be ruled out. It would, of course, be inappropriate to test the theory by observations on a transition of mixed multipolarity because two more particle parameters (unknowns) would be introduced.

The Ce\(^{137} \) isomeric transition should be very pure M\(^4\). Evidence was obtained from the \( \gamma \)-ray anisotropy (Sec. III.B). The lifetime is only a factor of three below the single particle estimates,\(^ {59} \) and even if the E5 strength were enhanced by as much as a factor of 50 over single proton estimates, which is extremely unlikely, the E5/M\(^4\) intensity mixing ratio would be only \( 2 \times 10^{-4} \) and would change the theoretical \( b_2 \) by less than the experimental error.
Table II. Conversion-electron particle parameters for transitions of "pure" multipolarity.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>$E_{\gamma}$/keV</th>
<th>Multipolarity</th>
<th>$b_2^{\text{theo.}}$</th>
<th>$b_2^{\text{expt.}}$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cd$^{111}$</td>
<td>247</td>
<td>E2</td>
<td>1.93</td>
<td>1.9 ± 0.1</td>
<td>23</td>
</tr>
<tr>
<td>Ta$^{181}$</td>
<td>133</td>
<td>E2</td>
<td>1.84</td>
<td>1.8 ± 0.2</td>
<td>24</td>
</tr>
<tr>
<td>Ce$^{137m}$</td>
<td>255</td>
<td>M4</td>
<td>1.055</td>
<td>1.061 ± 0.018</td>
<td>this work</td>
</tr>
</tbody>
</table>
There were no intermediate states in this experiment, so the
\( U_{k}Q_{k} \) factors in Eq. (III.1) can be replaced by unity. The \( B_{k} \) are
identical for conversion electrons and \( \gamma \)-ray emission. The \( g_{k} \) correc-
tions are accurately known, and \( P_{k}(1) = 1 \) for all \( k \). The \( B_{k} \) are all
interrelated through the spin Hamiltonian for \( \text{Ce}^{5+} \) in \( \text{Nes}^{49} \) (Sec. III.B),
and all \( B_{k} \) for \( k \geq 6 \) are negligible (in fact \( B_{6} = +0.0016 \) at \( T=0.02^\circ K \)).
The relationship between \( B_{2} \) and \( B_{4} \) was accurately known from independent
experiments on the \( \gamma \)-ray angular distributions at \( \theta = 0 \) and \( \theta = \pi/2 \)
(Sec. III.B). Therefore we know \( B_{2} \) and \( B_{4} \) for any \( W(0) \), and
Eq. (III.1) may be simplified to

\[
W(0)_{e} = 1 + B_{2}g_{2}(e)b_{2}F_{2} + B_{4}g_{4}(e)b_{4}F_{4}, \quad (\text{III.2a})
\]

\[
W(0)_{\gamma} = 1 + B_{2}g_{2}(\gamma)F_{2} + B_{4}g_{4}(\gamma)F_{4}, \quad (\text{III.2b})
\]

For this experiment \( F_{2} = -0.889, \quad F_{4} = +0.443, \quad g_{2}(e) = 0.935 \pm 0.015, \)
\( g_{2}(\gamma) = 0.905 \pm 0.005, \) and \( g_{4}(\gamma) = 0.700 \pm 0.010. \) Thus

\[
\frac{1-W(0)_{e}}{1-W(0)_{\gamma}} = \frac{0.831(13)B_{2}b_{2} + 0.354(18)B_{4}b_{4}}{0.802(4)B_{2} + 0.310(4)B_{4}}, \quad (\text{III.3})
\]

where \( b_{2} \) and \( b_{4} \) are related (see Eq. 98 of Ref. 21), for this case by

\[
b_{4} = \frac{17}{3} b_{2} - \frac{14}{3} \quad \text{.} \quad (\text{III.4})
\]

Substituting Eq. (III.4) into Eq. (III.3) we obtain, for any specific:
value of \( 1-W(0)_{\gamma} \), an equation with only one unknown, \( b_{2} \). We have cal-
culated \( [1-W(0)_{e}]/[1-W(0)_{\gamma}] \) as a function of \( 1-W(0)_{\gamma} \) for several values
of \( b_{2} \). These curves are compared with the data in Figs. III.5 and III.6.
From all the \( 0^\circ \) data we obtain the values (standard deviations)
\( b_{2}(K) = 1.061(6), \quad b_{2}(L+M) = 1.059(8). \) The largest known systematic
ersors arise from uncertainties in the background and solid angle correc-
tions. These uncertainties add up to about two standard deviations. Thus
we quote the final values
\[ b_2(K) = 1.061(18), \]
\[ b_2(L+M) = 1.059(20). \]

The theoretical value\(^{21}\) of 1.055 for \( b_2(K) \) is in excellent agreement with our result. No theoretical values are available for the L and M shells.

The results indicate that the theory is accurate to about \( \pm 2\% \) for this case. Of course, this single result does not establish the theory for cases in which the \( b \) are sensitive to details of nuclear and electronic structure (i.e., low energies and/or multipolarities), but it may serve as a point of reference for the study of such cases.

B. The Angular Distribution of \( \gamma \)-rays from Ce\(^{137m}\) Oriented in NES and the NES Temperature Scale

1. Introduction

The determination of the particle parameter is independent of temperature, being a direct comparison of simultaneously measured electron and \( \gamma \)-ray anisotropies. In the analysis, however, the relative sizes of the \( P_2 \) and \( P_4 \) terms must be known. This ratio is determined by the form of the spin Hamiltonian (which gives \( B_2 \) and \( B_4 \)) and is independent of temperature measurement.

However, the temperature variation of the 255 keV \( \gamma \)-ray anisotropy was redetermined in order to examine certain systematic discrepancies between theory and experiment noted in the work of Haag et al.\(^{49}\) These discrepancies were attributed to a possible error in the temperature scale for NES.

2. Experimental

Ce\(^{137m}\) was obtained as described in Sec. III.A.2 and activity was incorporated into the bulk of a single NES crystal by growing a seed crystal in a saturated solution of neodymium ethylsulfate containing
the activity. This was done to avoid local heating due to the Ce\textsuperscript{137m} conversion electrons. The γ-ray anisotropy was measured as a function of the magnetic temperature $T^*$ of the crystal, which was corrected to absolute temperatures using the $(1/T-1/T_0)$ correlation of Meyer (Sec.II.B.4) and an empirically determined demagnetization correction. For this experiment,

$$T_0 = T^* + 0.0070.$$ 

The effect of the manganous ammonium sulfate and (chrome alum)-glycerin slurry guard salts on the $T^*$ measurements was determined by removing the NES crystal from the cryostat and recalibrating the coils from $4^\circ\text{K}$ to $1^\circ\text{K}$. The result was a 14% adjustment in the slope of the calibration curve.

3. Results and Discussion

The resulting anisotropies (corrected for background and source decay) as a function of $1/T$, are tabulated in Table III. They are shown in Fig. III.7. A plot of $W(0)$ against $W(\pi/2)$ is shown in Fig. III.8. This plot is sensitive to the relative magnitude and signs of the $P_2$ and $P_4$ terms but does not depend on the magnitude of $\mu^*_N$, the nuclear moment. This quantity only determines the temperature to which a given point on the line corresponds. The solid curve is the theoretical relationship for a pure $M_4$ transition and using the effective spin Hamiltonian, adopted by Haag,

$$\hat{H} = \frac{A S_z I_z}{I} + B (S_x I_x + S_y I_y),$$

where

$$A = \frac{0.074}{I} \text{ \, } \mu\text{N cm}^{-1} \text{ and } B = \frac{0.002}{I} \text{ \, } \mu\text{N cm}^{-1},$$

taking $I = \frac{11}{2}$. The curve is corrected for solid angle ($g_2 = 0.918$, $g_4 = 0.744$). The theory and experiment are in good agreement, providing evidence that the radiation is indeed pure $M_4$ with an upper limit of $\delta = \pm0.010$ for the amplitude of a possible $E5$ admixture in the transition. (Fig. III.9)
Table III. \( W(\theta) \) vs \( \frac{1}{T} \) for 255 keV \( \gamma \)-rays from Ce\(^{137m}\) in NES.

<table>
<thead>
<tr>
<th>( \frac{1}{T} ) ( \theta )</th>
<th>( \frac{1}{T} ) ( \text{(Meyer)} )</th>
<th>( W(\theta) )</th>
<th>( W(\pi/2) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>36</td>
<td>56</td>
<td>0.370</td>
</tr>
<tr>
<td>2</td>
<td>35</td>
<td>53</td>
<td>0.391</td>
</tr>
<tr>
<td>3</td>
<td>34</td>
<td>48.5</td>
<td>0.428</td>
</tr>
<tr>
<td>4</td>
<td>31.8</td>
<td>41.4</td>
<td>0.486</td>
</tr>
<tr>
<td>5</td>
<td>33</td>
<td>44.1</td>
<td>0.435</td>
</tr>
<tr>
<td>6</td>
<td>32</td>
<td>41.8</td>
<td>0.514</td>
</tr>
<tr>
<td>7</td>
<td>29.8</td>
<td>36.5</td>
<td>0.601</td>
</tr>
<tr>
<td>8</td>
<td>30.7</td>
<td>39.0</td>
<td>0.540</td>
</tr>
<tr>
<td>9</td>
<td>30</td>
<td>37.7</td>
<td>0.568</td>
</tr>
<tr>
<td>10</td>
<td>29.5</td>
<td>35.4</td>
<td>0.621</td>
</tr>
<tr>
<td>11</td>
<td>26.6</td>
<td>31.2</td>
<td>0.707</td>
</tr>
<tr>
<td>12</td>
<td>26</td>
<td>30.0</td>
<td>0.724</td>
</tr>
<tr>
<td>13</td>
<td>24</td>
<td>27.9</td>
<td>0.770</td>
</tr>
<tr>
<td>14</td>
<td>16.3</td>
<td>18.1</td>
<td>0.927</td>
</tr>
<tr>
<td>15</td>
<td>15.3</td>
<td>17.1</td>
<td>0.916</td>
</tr>
<tr>
<td>16</td>
<td>24.7</td>
<td>28.6</td>
<td>0.751</td>
</tr>
<tr>
<td>17</td>
<td>23</td>
<td>26.3</td>
<td>0.783</td>
</tr>
<tr>
<td>18</td>
<td>30.0</td>
<td>37.1</td>
<td>0.597</td>
</tr>
<tr>
<td>19</td>
<td>29.7</td>
<td>36.3</td>
<td>0.618</td>
</tr>
<tr>
<td>20</td>
<td>28.7</td>
<td>34.0</td>
<td>0.651</td>
</tr>
<tr>
<td>21</td>
<td>26.4</td>
<td>30.8</td>
<td>0.709</td>
</tr>
<tr>
<td>22</td>
<td>24.3</td>
<td>28.0</td>
<td>0.759</td>
</tr>
<tr>
<td>23</td>
<td>19.2</td>
<td>22.1</td>
<td>0.851</td>
</tr>
<tr>
<td>24</td>
<td>18</td>
<td>20.5</td>
<td>0.869</td>
</tr>
<tr>
<td>25</td>
<td>16.5</td>
<td>18.3</td>
<td>0.838</td>
</tr>
<tr>
<td>26</td>
<td>14.4</td>
<td>16.1</td>
<td>0.911</td>
</tr>
<tr>
<td>27</td>
<td>38</td>
<td>64.0</td>
<td>0.247</td>
</tr>
</tbody>
</table>
### Table III. (Cont)

<table>
<thead>
<tr>
<th>$\frac{1}{T^@}$</th>
<th>$\frac{1}{T}$ (Meyer)</th>
<th>$W(0)$</th>
<th>$W(\pi/2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>28</td>
<td>37</td>
<td>63.0</td>
<td>0.277</td>
</tr>
<tr>
<td>29</td>
<td>36.3</td>
<td>58.3</td>
<td>0.380</td>
</tr>
<tr>
<td>30</td>
<td>35.6</td>
<td>55.4</td>
<td>0.418</td>
</tr>
<tr>
<td>31</td>
<td>34</td>
<td>47.8</td>
<td>0.446</td>
</tr>
<tr>
<td>32</td>
<td>32.7</td>
<td>44.0</td>
<td>0.480</td>
</tr>
<tr>
<td>33</td>
<td>31.8</td>
<td>41.2</td>
<td>0.500</td>
</tr>
</tbody>
</table>

**Corrections:**

**Temperature**

\[ T^@ = T^* + 0.0070 \]

**Solid Angle**

\[ \varepsilon_2 = 0.918 \]
\[ \varepsilon_4 = 0.744 \]
Fig. III.7. $W(\theta)$ for $\theta = 0$ and $\pi/2$, as a function of $1/T$ for the 255 keV $\gamma$-ray in Ce$^{137m}$. The solid curve is theoretical for a hyperfine structure constant $|A| = 0.0147$ cm$^{-1}$. 
Fig. III.8. $W(0)$ plotted against simultaneously measured values of $W(\pi/2)$ for the 255 keV $\gamma$-ray in Ce$^{137m}$. The dotted line shows the calculated result for a pure $P_2$ distribution. The solid curve is calculated assuming a pure $M^4$ transition.
Fig. III.9. $F_2$ as a function of $\delta$ for small values of $\delta$. The shaded area corresponds to the experimental value.
In Fig. III.10 is plotted the anisotropy $\epsilon = 1 - \frac{W(0^+)}{W(\pi/2)}$ as a function of $\frac{1}{T}$. At all temperatures the anisotropies measured were larger than those of Haag et al., requiring that the hfs constant be somewhat larger than their derived value of $A = 0.0129(12)$ cm$^{-1}$. The theoretical curve shown in Fig. III.10 is for $A = 0.0147$ cm$^{-1}$. The detailed agreement of theory with experiment is still poor. The theoretical curve shows definite discrepancies, being too high in the region $10 \leq \frac{1}{T} \leq 25$ and too low for $\frac{1}{40} \leq \frac{1}{T} \leq 50$. Similar deviations were found previously.

In general the form of the $\epsilon$ vs $\frac{1}{T}$ plot depends upon the $F_\nu$, the ratio of the hyperfine structure constants $A$ and $B$, and the temperature measurement. The small admissible $E5$ admixture allows very little change in $F_2$ and $F_4$, and we have found it impossible to obtain a better fit by altering the $A$ to $B$ ratio. Thus the measurement indicates systematic errors in the temperature scale, giving temperatures too low by about 0.01°K in the region of 0.05°K, and too high by about 0.001°K at 0.02°K. Further evidence for such errors has been found in studies of electric quadrupole hyperfine coupling in rare earth ions.60

However, taking the theoretical curve shown as the best fit to our data a new value of $A = 0.0147(7)$ cm$^{-1}$ may be derived for the hfs constant of Ce$^{137m}$ in NES. This error is statistical and no contribution from the temperature scale errors in included. This new value of $A$ may be preferred over the earlier result on the internal evidence of a larger anisotropy at each magnetic temperature. Note, however, that both values depend on the 1957 temperature scale of Meyer; an improved temperature scale will alter this value somewhat.

To derive a nuclear moment from this hfs constant a value of $\langle r^{-3} \rangle_{4f}$ is required. Recently the value of 3.64 a.u. for Ce$^{3+}$, which was used to derive Eq. (III.6), has been seriously challenged by Bleaney and by Freeman and Watson.63 In Table IV are listed the various proposed values of $\langle r^{-3} \rangle$ and the magnetic moment values which each would imply.
Fig. III.10. Anisotropy $\epsilon = 1 - \frac{W(0)}{W(\pi/2)}$ plotted as a function of $1/T$, showing the deviations from the "best" calculated fit, which are attributed to systematic deviations in the $T-T^*$ correlation for NES.
Table IV. Magnetic moment for Ce\textsuperscript{137m} implied by various values of \(\langle r^{-3}\rangle_{4f}\) for Ce\textsuperscript{3+}.

<table>
<thead>
<tr>
<th>(\langle r^{-3}\rangle) a.u.</th>
<th>(\mu_{137m}) n.m.</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.64</td>
<td>1.09(6)</td>
<td>27</td>
</tr>
<tr>
<td>4.74</td>
<td>0.84(5)</td>
<td>29</td>
</tr>
<tr>
<td>4.44</td>
<td>0.89(5)</td>
<td>28</td>
</tr>
</tbody>
</table>
IV. NUCLEAR ALIGNMENT OF Ce\(^{137m}\) AND THE CMN TEMPERATURE SCALE

A. Nuclear Alignment of Ce\(^{137m}\)

1. Introduction

The variation of the 255 keV \(\gamma\)-ray anisotropy as a function of temperature for Ce\(^{137m}\) in cerium magnesium nitrate (CMN) was determined to check the temperature scale for this salt. CMN is of great importance in low temperature work because it can be demagnetized to very low temperatures (\(<0.005^{\circ}\)K) and because the susceptibility deviates only slightly from Curie's Law down to 0.005\(^{\circ}\)K.

The \(\frac{1}{T\Omega}-\frac{1}{T}\) correlation for CMN was made by Daniels and Robinson\(^45\), using methods derived from the second law of thermodynamics (see Sec. II.B.6). Using their data Deklerk recalculated the correlation.\(^43\) Figure IV.1 shows \(1/T\Omega\) vs \(1/T\) according to Daniels and Robinson and according to Deklerk. Hudson et al. have performed entropy experiments which apparently do not agree with Daniels and Robinson's results.\(^64\)

Ce\(^{137m}\) is a singularly good choice for the elucidation of the temperature scale by nuclear alignment. As shown in Sec. III.B, the 255 keV \(\gamma\)-rays from the isomeric transition exhibit a large anisotropy at low temperatures and the multipolarity of the transition is well established as pure M4. An earlier experiment by Haag, Shirley, and Templeton showed that the anisotropy was not saturated in CMN even at \(1/T = 300^{\circ}\)K.

The Hamiltonian for Ce\(^{137m}\) in CMN may be inferred from the paramagnetic resonance experiments of Kedzie et al.\(^65\) on Ce\(^{141}\) in lanthanum magnesium nitrate. Ce\(^{1+}\) has the configuration \(4f^{1+}\) and the lowest level is \(\frac{2}{3}F_{5/2}\). In the double nitrate, the crystalline field has \(C_{3V}\) symmetry which splits the ground level into doublets which may be characterized approximately by \(\pm J_z\). Kedzie et al. found that only the lowest doublet was appreciably populated at \(4^{\circ}\)K and that it was mainly \(\pm 1/2\) with a small admixture of \(\pm 5/2\). They also saw hyperfine structure due to the Ce\(^{141}\) and interpreted their spectra with the Hamiltonian:

\[
H = g_\| \beta H_z S_z + g_\perp \beta (H_x S_x + H_y S_y) \quad (IV.1)
+ A S_z \mathbf{I}_z + B(S_x \mathbf{I}_x + S_y \mathbf{I}_y)
\]
Fig. IV.1. \((1/T^\nu - 1/T)\) correlations for cerium magnesium nitrate. For \(1 \leq 1/T^\nu \leq 100\), \(1/T^\nu = 1/T\).

Curve A: Daniels and Robinson
Curve B: DeKlerk (using Daniels and Robinson's data)
Curve C: derived in this work
Curve D: \(1/T^\nu - 1/T\) (shown for reference)
where $S = 1/2$, $B(\text{Ce}^{141}) = 0.0126 \, \text{cm}^{-1}$, and $B/A \ll 1$. The $g$-factor measurements gave $g_\perp = 1.84$, $g_\parallel = 0.023$. The same values were obtained by Cooke et al. for CMN. \(^6\) Hudson et al. report $g_\parallel = 0$ in CMN. This implies that Ce\(^{4+}\) has the same electronic structure in CMN as LMN, and therefore we expect the Hamiltonian adopted by Kedzie et al. to apply to Ce\(^{137m}\) in CMN.

2. Experimental

The anisotropy of the 255 keV gamma was measured as a function of temperature in three series of demagnetizations, using three separate samples. The first two samples were prepared by growing a CMN seed crystal in a saturated solution of CMN containing Ce\(^{137m}\) activity. The crystal used in the first experiment was thicker than that used in the second experiment. The third sample consisted of three CMN crystals, shaped and glued together with Duco cement to make a rough sphere. The samples were demagnetized from fields up to 22kG at 1\(^\circ\)K. Counting runs of one minute were started upon demagnetization, as were $T^*$ measurements. $T^*$ was measured as a function of time and an average $T^*$ was determined for each counting run. The counts were normalized at 1\(^\circ\)K. Only the first count after demagnetization was used in the temperature analysis. For all three samples, the time required to warm the crystal from the lowest temperatures to the bath temperature (from stray heat influx and radioactive heating) was about 1.5 h.

The counts were corrected for background and decay. The average $T^*$'s were corrected to $T^*$'s as described in Sec. II.B.6. The anisotropy at 0\(^\circ\) as a function of $(1/T)_{DR}$ (where $(1/T)_{DR}$ was obtained from $1/T^*$ using the curve given by Daniels and Robinson) is shown in Fig. IV.2 for the spherical crystal and in Fig. IV.3 for all three runs. The data are tabulated in Table V. Using the $(1/T^*, 1/T)$ relation given by DeKlerk, the anisotropy as a function of $1/T$ is shown in Fig. IV.4 for the spherical crystal. The solid curve is theoretical

$$W(0) = 1 + g_2 B_2 F_2 + g_4 B_4 F_4$$
Fig. IV.2. $W(\theta)$ for $\theta = 0$ for the 255 keV $\gamma$-ray in $\text{Ce}^{137\text{m}}$ plotted as a function of $1/T$, using Daniels and Robinson's $(1/T^2 - 1/T)$ correlation (for spherical crystal).
Fig. IV.3. $W(\theta)$ for $\theta = 0$ for the 255 keV $\gamma$-ray in Ce$^{137m}$ aligned in three different samples of CMN. $1/T$ was derived from $1/T^\theta$ using Daniels and Robinson's correlation.
Table Va. Ce$^{137m}$ in CMN 255 keV γ-ray anisotropy vs 1/T.
Experimental data 4/15/64

<table>
<thead>
<tr>
<th>Run</th>
<th>$W(\theta)$</th>
<th>$W(\pi/2)$</th>
<th>$1/T^\circ$</th>
<th>$(1/T)_{D.R.}$</th>
<th>$(1/T)$ this work</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.773</td>
<td>0.726</td>
<td>287</td>
<td>324</td>
<td>377</td>
</tr>
<tr>
<td>2</td>
<td>1.811</td>
<td>0.7730</td>
<td>307</td>
<td>325</td>
<td>480</td>
</tr>
<tr>
<td>3</td>
<td>1.476</td>
<td>0.7824</td>
<td>157</td>
<td>159</td>
<td>157</td>
</tr>
<tr>
<td>4</td>
<td>1.295</td>
<td>0.8726</td>
<td>90</td>
<td>90</td>
<td>90</td>
</tr>
<tr>
<td>5</td>
<td>1.578</td>
<td>0.8084</td>
<td>190</td>
<td>195</td>
<td>196</td>
</tr>
<tr>
<td>6</td>
<td>1.391</td>
<td>0.8230</td>
<td>124</td>
<td>124</td>
<td>124</td>
</tr>
<tr>
<td>7</td>
<td>1.662</td>
<td>0.7714</td>
<td>210</td>
<td>232</td>
<td>222</td>
</tr>
<tr>
<td>8</td>
<td>1.612</td>
<td>0.7415</td>
<td>221</td>
<td>250</td>
<td>236</td>
</tr>
<tr>
<td>9</td>
<td>1.166</td>
<td>0.8928</td>
<td>61</td>
<td>61</td>
<td>62</td>
</tr>
<tr>
<td>10</td>
<td>1.649</td>
<td>0.7498</td>
<td>251</td>
<td>296</td>
<td>288</td>
</tr>
<tr>
<td>11</td>
<td>1.308</td>
<td>0.8562</td>
<td>103</td>
<td>103</td>
<td>103</td>
</tr>
<tr>
<td>12</td>
<td>1.458</td>
<td>0.8382</td>
<td>139</td>
<td>139</td>
<td>139</td>
</tr>
<tr>
<td>13</td>
<td>1.518</td>
<td>0.7906</td>
<td>172</td>
<td>175</td>
<td>172</td>
</tr>
<tr>
<td>14</td>
<td>1.713</td>
<td>0.7353</td>
<td>261</td>
<td>312</td>
<td>310</td>
</tr>
<tr>
<td>15</td>
<td>1.595</td>
<td>0.7717</td>
<td>239</td>
<td>280</td>
<td>266</td>
</tr>
<tr>
<td>16</td>
<td>1.601</td>
<td>0.7772</td>
<td>203</td>
<td>218</td>
<td>213</td>
</tr>
<tr>
<td>17</td>
<td>1.022</td>
<td>0.9924</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>18</td>
<td>1.104</td>
<td>0.9543</td>
<td>40</td>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>19</td>
<td>1.692</td>
<td>0.7381</td>
<td>270</td>
<td>320</td>
<td>330</td>
</tr>
<tr>
<td>20</td>
<td>1.830</td>
<td>0.7095</td>
<td>308</td>
<td>325</td>
<td>490</td>
</tr>
</tbody>
</table>
Table Vb. Ce\textsuperscript{137m} in CNM. Experimental data 9/10/64.

<table>
<thead>
<tr>
<th>Run</th>
<th>W(0)</th>
<th>1/T*</th>
<th>(1/T)\textsubscript{D.R.}</th>
<th>(1/T)\textsubscript{this work}</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.857</td>
<td>312</td>
<td>325</td>
<td>520</td>
</tr>
<tr>
<td>2</td>
<td>1.752</td>
<td>286</td>
<td>324</td>
<td>370</td>
</tr>
<tr>
<td>3</td>
<td>1.633</td>
<td>247</td>
<td>293</td>
<td>280</td>
</tr>
<tr>
<td>4</td>
<td>1.717</td>
<td>258</td>
<td>308</td>
<td>302</td>
</tr>
<tr>
<td>5</td>
<td>1.583</td>
<td>208</td>
<td>230</td>
<td>219</td>
</tr>
<tr>
<td>6</td>
<td>1.423</td>
<td>140</td>
<td>140</td>
<td>136</td>
</tr>
<tr>
<td>7</td>
<td>1.143</td>
<td>54</td>
<td>54</td>
<td>54</td>
</tr>
<tr>
<td>8</td>
<td>1.587</td>
<td>205</td>
<td>224</td>
<td>216</td>
</tr>
<tr>
<td>9</td>
<td>1.828</td>
<td>292</td>
<td>325</td>
<td>392</td>
</tr>
<tr>
<td>10</td>
<td>1.621</td>
<td>217</td>
<td>240</td>
<td>231</td>
</tr>
<tr>
<td>11</td>
<td>1.437</td>
<td>148</td>
<td>148</td>
<td>146</td>
</tr>
<tr>
<td>12</td>
<td>1.887</td>
<td>312</td>
<td>325</td>
<td>520</td>
</tr>
<tr>
<td>13</td>
<td>1.863</td>
<td>309</td>
<td>325</td>
<td>494</td>
</tr>
<tr>
<td>14</td>
<td>1.870</td>
<td>302</td>
<td>325</td>
<td>440</td>
</tr>
<tr>
<td>15</td>
<td>1.647</td>
<td>218</td>
<td>244</td>
<td>232</td>
</tr>
<tr>
<td>16</td>
<td>1.432</td>
<td>142</td>
<td>142</td>
<td>140</td>
</tr>
<tr>
<td>17</td>
<td>1.863</td>
<td>304</td>
<td>325</td>
<td>450</td>
</tr>
<tr>
<td>18</td>
<td>1.142</td>
<td>52</td>
<td>52</td>
<td>52</td>
</tr>
<tr>
<td>19</td>
<td>1.818</td>
<td>300</td>
<td>325</td>
<td>428</td>
</tr>
<tr>
<td>20</td>
<td>1.621</td>
<td>215</td>
<td>240</td>
<td>228</td>
</tr>
<tr>
<td>21</td>
<td>1.761</td>
<td>268</td>
<td>316</td>
<td>328</td>
</tr>
<tr>
<td>22</td>
<td>1.824</td>
<td>292</td>
<td>324</td>
<td>392</td>
</tr>
<tr>
<td>23</td>
<td>1.735</td>
<td>258</td>
<td>308</td>
<td>302</td>
</tr>
<tr>
<td>24</td>
<td>1.800</td>
<td>291</td>
<td>323</td>
<td>388</td>
</tr>
<tr>
<td>25</td>
<td>1.348</td>
<td>106</td>
<td>106</td>
<td>106</td>
</tr>
<tr>
<td>26</td>
<td>1.317</td>
<td>106</td>
<td>106</td>
<td>106</td>
</tr>
</tbody>
</table>

\( T^\circ = T^\circ + 0.00194 \)
Table Vc. Ce\sup{137}m in CMN. Experimental data 11/17/64.

<table>
<thead>
<tr>
<th>Run</th>
<th>W(0)</th>
<th>1/(T^\oplus)</th>
<th>(1/(T))\text{D.R.}</th>
<th>(1/(T))\text{DeK}</th>
<th>(1/(T))\text{this work}</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.875</td>
<td>312</td>
<td>325</td>
<td>380</td>
<td>520</td>
</tr>
<tr>
<td>2</td>
<td>1.783</td>
<td>295</td>
<td>325</td>
<td>310</td>
<td>400</td>
</tr>
<tr>
<td>3</td>
<td>1.695</td>
<td>269</td>
<td>319</td>
<td>278</td>
<td>330</td>
</tr>
<tr>
<td>4</td>
<td>1.614</td>
<td>219</td>
<td>247</td>
<td>223</td>
<td>233</td>
</tr>
<tr>
<td>5</td>
<td>1.458</td>
<td>151</td>
<td>152</td>
<td>152</td>
<td>151</td>
</tr>
<tr>
<td>6</td>
<td>1.203</td>
<td>66</td>
<td>66</td>
<td>66</td>
<td>66</td>
</tr>
<tr>
<td>7</td>
<td>1.742</td>
<td>269</td>
<td>318</td>
<td>276</td>
<td>330</td>
</tr>
<tr>
<td>8</td>
<td>1.881</td>
<td>312</td>
<td>325</td>
<td>380</td>
<td>520</td>
</tr>
<tr>
<td>9</td>
<td>1.161</td>
<td>60</td>
<td>60</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>10</td>
<td>1.635</td>
<td>221</td>
<td>252</td>
<td>225</td>
<td>256</td>
</tr>
<tr>
<td>11</td>
<td>1.785</td>
<td>300</td>
<td>325</td>
<td>328</td>
<td>428</td>
</tr>
<tr>
<td>12</td>
<td>1.618</td>
<td>222</td>
<td>254</td>
<td>226</td>
<td>237</td>
</tr>
<tr>
<td>13</td>
<td>1.446</td>
<td>150</td>
<td>150</td>
<td>152</td>
<td>150</td>
</tr>
<tr>
<td>14</td>
<td>1.733</td>
<td>267</td>
<td>317</td>
<td>274</td>
<td>324</td>
</tr>
<tr>
<td>15</td>
<td>1.210</td>
<td>58</td>
<td>58</td>
<td>58</td>
<td>58</td>
</tr>
<tr>
<td>16</td>
<td>1.442</td>
<td>154</td>
<td>155</td>
<td>155</td>
<td>154</td>
</tr>
<tr>
<td>17</td>
<td>1.851</td>
<td>305</td>
<td>325</td>
<td>345</td>
<td>460</td>
</tr>
<tr>
<td>18</td>
<td>1.789</td>
<td>289</td>
<td>325</td>
<td>300</td>
<td>381</td>
</tr>
<tr>
<td>19</td>
<td>1.807</td>
<td>302</td>
<td>325</td>
<td>330</td>
<td>440</td>
</tr>
<tr>
<td>20</td>
<td>1.785</td>
<td>278</td>
<td>322</td>
<td>297</td>
<td>352</td>
</tr>
<tr>
<td>21</td>
<td>1.756</td>
<td>304</td>
<td>325</td>
<td>342</td>
<td>450</td>
</tr>
<tr>
<td>22</td>
<td>1.880</td>
<td>312</td>
<td>325</td>
<td>380</td>
<td>520</td>
</tr>
<tr>
<td>23</td>
<td>1.851</td>
<td>300</td>
<td>325</td>
<td>328</td>
<td>430</td>
</tr>
</tbody>
</table>

\(T^\oplus = T^\ominus + 0.00022\)
Fig. IV.4. $W(\theta)$ for $\theta = 0$ for the 255 keV $\gamma$-ray in $\text{Ce}^{137m}$ plotted as a function of $1/T$, using DeKlerk's ($1/T^2 - 1/T$) correlation.
where $g_2 = 0.915$, $g_4 = 0.735$ (the solid angle was the same in all three experiments) and $F_2$ and $F_4$ were given in Sec. III. $B_2$ and $B_4$ were calculated as functions of $1/T$ using the Hamiltonian given in Eq. (IV.1), with $B = 0.0060$ cm$^{-1}$. This gives the best fit for $1 < \frac{1}{T} < 200$. Independent evidence for the validity of this Hamiltonian is shown in Fig. IV.5, where $W(0)$ is plotted against $W(\pi/2)$ for data taken in the first experiment. The solid curve is theoretical and depends on the relative sizes of $B_2$ and $B_4$, which depend on the form of the Hamiltonian. (For $H = AS_z I_z$, see Fig. III.8 in Sec. III.B.) Figure IV.6 shows $(H/T)_{\text{initial}}$ against $(J/T)_{\text{final}}$. 

3. Discussion

Apparently, neither $(1/T^2-1/T)$ correlation is suitable below $T = 0.005$, although the correlation given by DeKlerk gives smoother results than that given by Daniels and Robinson.

The regions of deviation of the experimental points corrected with Daniels' and Robinson's $(1/T^2-1/T)$ correlation from the theoretical curve is also the region where collective electronic effects become important and where a ferro- or antiferromagnetic transition probably occurs. This may be expected to change the Hamiltonian and lead to a change in $B_2$ and $B_4$.

For the purposes of calculation, the collective transition was simulated by a fictitious magnetic field applied in the $x$ direction. The Hamiltonian

$$H = g \beta H_x S_x + B(S_x I_x + S_y I_y)$$

(IV.2)

was then used to calculate the energy levels and $B_2$ and $B_4$. $B$ was taken as constant and $H_x$ was varied. Since the transition does not set in until about $1/T = 300$,

$$\mu H_x = kT$$

implies $H_x \approx 25$ gauss. For the sake of comparison, $B_2$ and $B_4$ were
Fig. IV.5. $W(\theta)$ for $\theta = 0$ plotted against simultaneously measured values of $W(\theta)$ for $\theta = \pi/2$. The solid curve is theoretical for an M4 transition and planar alignment. The dotted curve is theoretical for a pure P2 distribution.
Fig. IV.6. Magnetic temperatures $(1/T^*_T)$ obtained by demagnetizing from given initial conditions $[(H/T)_{\text{initial}}]$. The solid curve was drawn from data given by Daniels and Robinson.
calculated for $H_x = 0$ to $H_x = 10000$ G. The calculations showed that $|B_2|$ and $|B_4|$ were maximum for $H_x = 0$ and decreased monotonically as $H_x$ increased. At $1/T = 300$, $B_2$ went from $-0.751$ to $-0.705$ over this range while $B_4$ went from $+0.297$ to $+0.245$. Since $F_2$ is negative and $F_4$ is positive, this means that a collective transition may be expected to decrease the anisotropy slightly. This does not explain the observed effect, which is an increase in anisotropy.

A $(1/T^0 - 1/T)$ correlation can be derived from the nuclear alignment experiment. Only the spherical crystal data were used because the demagnetization correction was the smallest for this sample. A smooth curve was fitted to the data plotted as anisotropy vs $1/T^0$. The calculated curve shown in Figs. IV.2 and IV.7 gave the $1/T$ corresponding to a given anisotropy. The resulting anisotropy correlation is shown in Fig. IV.1 and is given in Table VI. Using this correlation, the data from all three experiments are plotted in Fig. IV.7.

The results suggest that CMN can be demagnetized to $T = 0.0019^0K$, confirming the observations of Hudson, Kaeser and Radford. This is of great importance to experiments using CMN as a coolant and as a temperature scale in the milli-degree range. Nuclear alignment experiments will be influenced, and nuclear moments determined by alignment in CMN will have to be reexamined. In addition, the proposed temperature scale is particularly crucial to the search for superfluidity in liquid He$^3$, predicted by Breuckner et al. Anderson, Salinger, Steyert, and Wheatley report no evidence for such a transition down to $0.008^0K$, based on specific heat measurements with CMN as a coolant and thermometer, but in a recent experiment of the same kind, V. P. Peshkov reports a superfluid transition at $0.0055^0K$. These data will have to be reexamined in the light of the new temperature scale before any conclusions can be reached.
Fig. IV.7. \( W(\theta) \) for \( \theta = 0 \) for the 255 keV \( \gamma \)-ray in Ce\(^{137m}\) plotted as a function of \( 1/T \), using the \( (1/T^* - 1/T) \) correlation derived from the spherical crystal data.
Table VI. \((1/T\oplus - 1/T)\) correlations for CMN.

<table>
<thead>
<tr>
<th>1/T(\oplus)</th>
<th>((1/T)_{DR}^{45})</th>
<th>((1/T)_{this\ work})</th>
<th>((H/T)_{initial}) (kG (\cdot) K(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>20</td>
<td>20</td>
<td>1.0</td>
</tr>
<tr>
<td>40</td>
<td>40</td>
<td>40</td>
<td>1.9</td>
</tr>
<tr>
<td>60</td>
<td>60</td>
<td>60</td>
<td>2.9</td>
</tr>
<tr>
<td>80</td>
<td>80</td>
<td>80</td>
<td>3.8</td>
</tr>
<tr>
<td>100</td>
<td>100</td>
<td>100</td>
<td>4.6</td>
</tr>
<tr>
<td>120</td>
<td>120</td>
<td>120</td>
<td>5.4</td>
</tr>
<tr>
<td>140</td>
<td>140</td>
<td>140</td>
<td>6.2</td>
</tr>
<tr>
<td>160</td>
<td>160</td>
<td>160</td>
<td>6.9</td>
</tr>
<tr>
<td>180</td>
<td>182</td>
<td>181</td>
<td>7.8</td>
</tr>
<tr>
<td>200</td>
<td>223</td>
<td>210</td>
<td>8.75</td>
</tr>
<tr>
<td>210</td>
<td>231</td>
<td>221</td>
<td>9.2</td>
</tr>
<tr>
<td>220</td>
<td>249</td>
<td>232</td>
<td>9.7</td>
</tr>
<tr>
<td>230</td>
<td>266</td>
<td>249</td>
<td>10.2</td>
</tr>
<tr>
<td>240</td>
<td>284</td>
<td>266</td>
<td>10.8</td>
</tr>
<tr>
<td>250</td>
<td>300</td>
<td>287</td>
<td>11.4</td>
</tr>
<tr>
<td>260</td>
<td>312.5</td>
<td>305</td>
<td>12.1</td>
</tr>
<tr>
<td>280</td>
<td>319</td>
<td>322</td>
<td>12.8</td>
</tr>
<tr>
<td>290</td>
<td>322.7</td>
<td>358</td>
<td>13.5</td>
</tr>
<tr>
<td>300</td>
<td>324</td>
<td>383</td>
<td>14.2</td>
</tr>
<tr>
<td>310</td>
<td>324</td>
<td>430</td>
<td>15.5</td>
</tr>
<tr>
<td>312</td>
<td>324</td>
<td>500</td>
<td>18.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>520</td>
<td>18.8</td>
</tr>
</tbody>
</table>
B. Nuclear Alignment of Pm\(^{144}\) in CMN: A Test of the Proposed Temperature Scale

1. Introduction

In order to test the validity of the \((1/T_0^2-1/T)\) correlation derived from the Ce\(^{137m}\) nuclear orientation data, the alignment of 450 day Pm\(^{144}\) in CMN was determined as a function of temperature. Pm\(^{144}\) was chosen because the ground state electronic level for Pm\(^{3+}\) in CMN is a singlet and this level only is appreciably populated at 1°K.\(^{70}\) Thus, it will not participate in any possible collective effects between the Ce\(^{3+}\) ions in CMN at the lowest temperatures (0.002°K). Furthermore, Pm isotopes have been aligned in CMN\(^{70,71}\) and the form of the Hamiltonian is known, viz.,

\[
H = P(I_z^2 - \frac{1}{3} I(I+1)). \tag{IV.3}
\]

This Hamiltonian has the form of quadrupolar interaction, but crystal field calculations by Grant and Shirley\(^{70}\) show that the major contribution to \(P\) is a magnetic interaction, termed the "pseudo-quadrupole" interaction. Finally, Pm\(^{144}\) was aligned in CMN by Grant and Shirley\(^{70}\) who concluded from their measurement that the \((1/T_0^2-1/T)\) correlation of Daniels and Robinson was correct.

2. Experimental

The electron capture decay of Pm\(^{144}\) to Nd\(^{144}\) was studied by Ofer\(^{72}\) and by Funk et al.\(^{73}\). The decay scheme is shown in Fig. IV.8. The spin and multipolarity assignments were confirmed by nuclear alignment experiments on Pm\(^{144}\) in NES.\(^{74}\) The Pm\(^{144}\) was produced by bombarding Pr\(^{144}\) as praseodymium oxide with alpha particles in the Berkeley 88-in. cyclotron. The beam energy was kept below 20 MeV to minimize Pr\(^{143}\) production. The Pm\(^{43}\) was separated from the Pr\(^{43}\) using a Dowex-50 cation exchange column with 0.4 M alpha hydroxy butyric acid as the eluant. The separated Pm\(^{144}\) was grown into a single crystal of CMN. The anisotropes of the 615 and 695 keV \(\gamma\)-rays were measured using NaI(Tl) crystals at 0° and 90° to the
Fig. IV.8. Decay scheme of $^{144}$Pm.
crystalline c. axis. Grant and Shirley showed that these two γ-rays have the same anisotropy. This was checked by measuring the anisotropies with a Ge(Li) detector, which clearly resolved the two γ-rays (Fig. IV.9). The Ge(Li) determinations confirmed the observation by Shirley et al. 74 that the anisotropy of the 474 keV transition is attenuated by 20%, from the theoretical predictions based on the decay scheme. The results of the Ge(Li) runs are present in Table VII. These runs also showed that the 740 keV γ-ray from the Pm 143 contamination was very small and had vanishingly small effect on the 615-695 keV γ-ray's anisotropies.

The anisotropy as a function of temperature for the combined 615 and 695 keV peaks, at 0° and 90° to the crystal c axis, is given in Table VIII. Only the first count after demagnetization was used in the temperature scale analysis. Additional data were taken for use in the determination of the relative sizes of B₂ and B₄. The temperature was measured as described in Sec. II.B.6. The validity of these measurements is indicated by the fact that H(initial)/T(initial) vs 1/T° (final) points were in good agreement with the measurements of Daniels and Robinson. 45

3. Results and Discussion

In Fig. IV.10, the 0° data are plotted against 1/T, using (1/T° - 1/T)DR. These data are in fairly good agreement with Grant and Shirley's measurements. Figure IV.11 shows W(0) vs W(π/2). This figure shows evidence for a large P₄ contribution to the anisotropy. This does not agree with Grant and Shirley, who found a very small or zero P₄ contribution. Curve A in Fig. IV.11 is theoretical for a pure P₂ distribution.

The W(0) vs W(π/2) curve is independent of temperature, but does depend on the ratio of B₂ and B₄. This is determined by the form of the Hamiltonian and the spin. Each point on the curve corresponds to some temperature determined by the size of the coupling constant, P. Assuming the decay sequence
Fig. IV.9. Gamma-ray spectrum of $\text{Pm}^{144}$ in a Ge(Li) detector.
Table VII. Relative Anisotropies for $^{144}_{\text{Pm}}$ in CMN

$(1/T) = 380 \,(\text{K}^{-1})$.

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$W(0)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>474</td>
<td>1.096 ± 0.007</td>
</tr>
<tr>
<td>615</td>
<td>1.122 ± 0.003</td>
</tr>
<tr>
<td>695</td>
<td>1.124 ± 0.004</td>
</tr>
<tr>
<td>740 ($^{143}_{\text{Pm}}$)</td>
<td>1.080 ± 0.013</td>
</tr>
</tbody>
</table>
Table VIIIa. $\text{Pm}^{144}$ in CMN. Anisotropies of the 615 and 695 keV $\gamma$-rays vs $1/T$. Experimental data.

<table>
<thead>
<tr>
<th>Run</th>
<th>$W(0)$</th>
<th>$W(\pi/2)$</th>
<th>$1/T^{\oplus}$</th>
<th>$(1/T)_{DR}$</th>
<th>$(1/T)_{\text{this work}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.144</td>
<td>0.906</td>
<td>311</td>
<td>324</td>
<td>510</td>
</tr>
<tr>
<td>2</td>
<td>1.123</td>
<td>0.929</td>
<td>285</td>
<td>323</td>
<td>370</td>
</tr>
<tr>
<td>3</td>
<td>1.083</td>
<td>0.952</td>
<td>196</td>
<td>206</td>
<td>204</td>
</tr>
<tr>
<td>4</td>
<td>1.054</td>
<td>0.977</td>
<td>128</td>
<td>128</td>
<td>128</td>
</tr>
<tr>
<td>5</td>
<td>1.029</td>
<td>0.985</td>
<td>60</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>6</td>
<td>1.089</td>
<td>0.948</td>
<td>200</td>
<td>213</td>
<td>210</td>
</tr>
<tr>
<td>7</td>
<td>1.030</td>
<td>0.991</td>
<td>67</td>
<td>67</td>
<td>67</td>
</tr>
<tr>
<td>8</td>
<td>1.061</td>
<td>0.967</td>
<td>135</td>
<td>135</td>
<td>135</td>
</tr>
<tr>
<td>9</td>
<td>1.142</td>
<td>0.909</td>
<td>312</td>
<td>324</td>
<td>515</td>
</tr>
<tr>
<td>10</td>
<td>1.134</td>
<td>0.923</td>
<td>293</td>
<td>324</td>
<td>394</td>
</tr>
<tr>
<td>11</td>
<td>1.087</td>
<td>0.949</td>
<td>205</td>
<td>323</td>
<td>216</td>
</tr>
<tr>
<td>12</td>
<td>1.051</td>
<td>0.971</td>
<td>137</td>
<td>137</td>
<td>137</td>
</tr>
<tr>
<td>13</td>
<td>1.023</td>
<td>0.988</td>
<td>60</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>14</td>
<td>1.130</td>
<td>0.919</td>
<td>296</td>
<td>324</td>
<td>402</td>
</tr>
<tr>
<td>15</td>
<td>1.100</td>
<td>0.944</td>
<td>257</td>
<td>309</td>
<td>300</td>
</tr>
<tr>
<td>16</td>
<td>1.107</td>
<td>0.950</td>
<td>257</td>
<td>309</td>
<td>300</td>
</tr>
<tr>
<td>17</td>
<td>1.123</td>
<td>0.924</td>
<td>273</td>
<td>320</td>
<td>314</td>
</tr>
<tr>
<td>18</td>
<td>1.133</td>
<td>0.912</td>
<td>306</td>
<td>324</td>
<td>460</td>
</tr>
<tr>
<td>19</td>
<td>1.113</td>
<td>0.938</td>
<td>302</td>
<td>324</td>
<td>440</td>
</tr>
<tr>
<td>20</td>
<td>1.128</td>
<td>0.924</td>
<td>277</td>
<td>322</td>
<td>350</td>
</tr>
<tr>
<td>21</td>
<td>1.141</td>
<td>0.918</td>
<td>312</td>
<td>324</td>
<td>520</td>
</tr>
<tr>
<td>22</td>
<td>1.119</td>
<td>0.938</td>
<td>255</td>
<td>307</td>
<td>296</td>
</tr>
<tr>
<td>23</td>
<td>1.100</td>
<td>0.943</td>
<td>232</td>
<td>269</td>
<td>256</td>
</tr>
<tr>
<td>24</td>
<td>1.139</td>
<td>0.911</td>
<td>312</td>
<td>324</td>
<td>520</td>
</tr>
</tbody>
</table>

$T^{\oplus} = T^{\oplus} + 0.00116$

$g_2 = 0.944$

$g_4 = 0.820$
Table VIIIb. \( ^{144}\text{Pm} \) in CMW. Additional \( W(0) \) vs \( W(\pi/2) \) data.

<table>
<thead>
<tr>
<th>( W(0) )</th>
<th>( W(\pi/2) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.055</td>
<td>0.964</td>
</tr>
<tr>
<td>1.113</td>
<td>0.912</td>
</tr>
<tr>
<td>1.125</td>
<td>0.921</td>
</tr>
<tr>
<td>1.082</td>
<td>0.952</td>
</tr>
<tr>
<td>1.020</td>
<td>0.995</td>
</tr>
<tr>
<td>1.123</td>
<td>0.916</td>
</tr>
<tr>
<td>1.122</td>
<td>0.923</td>
</tr>
<tr>
<td>1.131</td>
<td>0.919</td>
</tr>
<tr>
<td>1.109</td>
<td>0.938</td>
</tr>
<tr>
<td>1.118</td>
<td>0.924</td>
</tr>
<tr>
<td>1.122</td>
<td>0.919</td>
</tr>
<tr>
<td>1.110</td>
<td>0.934</td>
</tr>
<tr>
<td>1.096</td>
<td>0.940</td>
</tr>
<tr>
<td>1.146</td>
<td>0.915</td>
</tr>
<tr>
<td>1.135</td>
<td>0.923</td>
</tr>
</tbody>
</table>
Fig. IV.10. $W(\theta)$ for $\theta = 0$ for the 615 and 695 keV $\gamma$-rays in Pm$^{144}$ plotted as a function of $1/T$, using Daniels and Robinson's $(1/T \times 1/T)$ correlation.
Fig. IV.11. $W(\theta)$ for $\theta = 0$ for the 615 and 695 keV $\gamma$-rays in Pm$^{144}$ plotted against simultaneously measured $W(\theta)$ for $\theta = \pi/2$. Curve A is theoretical for a pure $P_2$ distribution. Curve B is theoretical assuming no attenuation. Curve C is a representative fit assuming attenuation of the anisotropy.
the angular distributions for the 615 and 695 keV γ-rays are

\[ W(\theta) = 1 - 0.441 B_2 P_2(\cos\theta) - 0.214 B_4 P_4(\cos\theta) \]

Using the Hamiltonian in Eq. (IV.3), \( B_2 \) and \( B_4 \) were calculated as a function of \( 1/T \), allowing a theoretical prediction for \( W(0) \) vs \( W(\pi/2) \). This is shown in Fig. IV.11 as curve B (including solid angle corrections \( g_2 = 0.944, g_4 = 0.820 \)). The deviation of the experimental points from this curve suggests an attenuation of the anisotropy. Precedence for attenuation is found in the work of Grant and Shirley on the 740 keV γ-ray from the decay of \( \text{Pm}^{143} \) in CMN. They found a saturation anisotropy of 1.09 [\( W(0) \)]. The multipolarity of the 740 keV transition has recently been remeasured as E2. This makes the probable decay sequence for \( \text{Pm}^{143} \)

\[ \frac{5/2^+}{j_B=1} \rightarrow \frac{3/2^+}{j_B=1} \rightarrow \frac{740 \text{ keV}}{E_2} \rightarrow 5/2^+ \]

for which \( W(0) = 1.112 \). Since \( B_4 (I=3/2) = 0 \), this gives \( Q_2 = 0.8 \), where \( Q_2 \) is the attenuation factor. (Section II.A)

For \( \text{Pm}^{144} \) then,

\[ W(0) = 1 + g_2 Q_2 B_2 U_2 F_2 + g_4 Q_4 B_4 U_4 F_4 \]

These two equations have two explicit unknowns, \( Q_2 \) and \( Q_4 \), and one implicit unknown, \( P \). \( P \) will determine the temperature to which a given ratio \( B_2/B_4 \) will correspond. Substituting the values of \( g \), \( U \) and \( F \) and solving,

\[ Q_2 B_2 = 0.412 \pm 0.011 \]

\[ Q_4 B_4 = 0.148 \pm 0.011 \]

for \( W(0) = 1.144, W(\pi/20) = 0.905 \).
A $B_2/B_4$ ratio implies a unique $Q_2/Q_4$ ratio. In addition, each $B_2/B_4$ ratio corresponds to a unique value of $B_2$. For a given value of $1/T$, this $B_2$ is related to a value of $P$. Thus, by assigning the value of $1/T$ at which $W(0) = 1.144$, the ratio $Q_2/Q_4$ may be related directly to values of $P$. This relationship is shown in Fig. IV.12 for $1/T_{\text{final}} = 520$.

Figure IV.13 shows the experimental $W(0)$ data vs $1/T$, using the $(1/T^2 - 1/T)$ correlation derived from the Ce$^{137}$m data (Sec. IV.A). The solid curve is theoretical for $Q_2 = Q_4 = 0.63$ and is shown as a representative fit to the data. In Fig. IV.11, curve C is the theoretical fit to $W(0)$ vs $W(\pi/2^\circ)$ data for the same parameters. The same $W(0)$ vs 1/T curve is shown in Fig. IV.10.

An upper limit to $P$ may be set from the limiting ratio of $B_2/B_4$, but a better limit on $P$ may be found by demanding that the calculated curve for $W(0)$ vs $1/T$ fit the experimental data over the entire range of $1/T$ from 1 to 520. With this condition, the upper limit to $Q_2/Q_4$ is 1.18 and hence

$$P \leq (0.50 \pm 0.20) \times 10^{-3} \text{ (°K)}.$$  

If $Q_4$ is allowed to be bigger than $Q_2$, but both $Q_2$ and $Q_4$ are required to be less than one,

$$P \geq (0.15 \pm 0.003) \times 10^{-3} \text{ (°K)}.$$  

At this lower limit to $P$, $Q_2 = 0.76$, in agreement with the $Q_2$ derived above for Pm$^{143}$ in CMN.

Several conclusions may be drawn from this experiment:

1. The experimental data show an anomalous behavior at low temperatures, using the $(1/T^2 - 1/T)$ correlation of Daniels and Robinson.

2. A "better" fit is obtained using the $(1/T^2 - 1/T)$ correlation derived from the Ce$^{137}$m data, qualitatively confirming the validity of that correlation.

3. The angular distributions of $\gamma$-rays from Pm isotopes aligned in CMN show a considerable attenuation of anisotropy. If this attenuation exists for Pm isotopes in NES as well, some measurements of Pm nuclear moments by nuclear alignment may require revision in the direction of larger moments.
Fig. IV.12. Values of the coupling constant implied by different values of $Q_2/Q_4$. The shaded area includes values allowed by demanding a good theoretical fit to the data, and by restricting $Q_2$ and $Q_4$ to values less than one.
Fig. IV.13. $W(\theta)$ for $\theta = 0$ for the 615 and 695 keV $\gamma$-rays in Pm$^{144}$ plotted as a function of $1/T$, using the $(1/T^2 - 1/T)$ correlation derived from the Ce$^{152}$ data.
V. THE DECAY OF Ce$^{137}$g AND Ce$^{137}$m AND THE LEVEL STRUCTURE OF La$^{137}$

1. Introduction

As techniques become more sophisticated, nuclear decay schemes become more complex. This is abundantly illustrated by the decay of Ce$^{137}$ to La$^{137}$. The decay scheme based on previous studies was shown in Fig. III.1. A revised version based on the work reported here is shown in Fig. V.16. Gamma-ray spectroscopy of Ce$^{137m+g}$ using NaI and Ge(Li) detectors is reviewed in Sec. V.2, and energy levels in La$^{137}$ are presented. Conversion electron measurements with Si(Li) detectors and multipolarity determinations are discussed in Sec. V.4. Section V.5 describes coincidence studies using NaI and Ge(Li) detectors. Spin determinations for several levels were made by studying the anisotropy of gamma-rays emitted from Ce$^{137}$ in NES and CMN, using Ge(Li) detectors. These measurements and spin assignments are discussed in Sec. V.6.

Kisslinger and Sorenson have calculated energy levels in spherical nuclei. Their calculations for La$^{137}$ are compared with the experimental results in Sec. V.7.

2. Gamma-ray Spectroscopy

The photon spectrum in NaI from the decay of Ce$^{137m+g}$ is shown in Fig. V.1. The analyzer was biased so that the Ce and La x-rays from conversion of the 255-keV isomeric transition in Ce and the electron capture to La are not shown. The spectrum shows several prominent features. The photopeak at 166 keV was assigned to Ce$^{139}$ on the basis of the half-life of its decay. The photopeak at 255 keV and 440 keV, and the complex peaks at ~800 keV are all due to Ce$^{137m+g}$ decay. An additional transition at 10 keV was reported by Brosi and Ketelle, who detected the gamma-ray and L and M conversion electrons with a krypton-filled proportional counter. From its conversion coefficient, they concluded the transition was almost pure M1. These observations have been confirmed by other workers and a lifetime measurement is consistent with an M1 transition.
Fig. V.1. $^{137m+g}$ photon spectrum, with a NaI spectrometer.
The $^{137m+g}\text{Ce}$ photon spectrum was also taken with a Ge(Li) detector. Figure V.2 shows the spectrum from 20-1050 keV. There are several surprises. The 445-keV transition is a doublet, 10 keV apart. The complex peak at ~800 keV is seen to be 11 transitions, four of them doublets spaced by 10 keV. Not shown is a weak singlet at 1160 keV. Using well-known standard gamma-ray sources, the energies of the peaks were determined to within one keV. The energies are tabulated in Table IX, with the 10 keV transition reported by other investigators. Using the efficiency curve for Ge shown in Fig. II.8, the relative photon intensities were calculated and are given in Table X.

Recently, discrepancies in the efficiency curve for Ge(Li) detectors have been found. Count rate, source position and collimation may all affect relative efficiencies. The relative intensities within doublets determined here should be valid, but relative efficiencies for different parts of the spectrum may be in doubt.

Certain regions of the spectrum were investigated with higher resolution. The peak at 166 keV formerly assigned to $^{139}\text{Ce}$ was found to be a doublet, with a peak at 166 keV and one at 168 keV. The 168 keV peak was found to decay with a 3½-hour half-life. The peaks are shown in Fig. V.3 for a source 7 days old and 8 days old. Evidence for peaks at 433 keV and 479 keV was obtained from investigations of the 400 keV region. These spectra are shown in Fig. V.4.

On the basis of these spectra, a tentative decay scheme was constructed. Brosi and Ketelle showed that the 10-keV transition they observed was from a level 10 keV above the ground state. They assigned the spins of these states as 7/2 (ground) and 5/2 (10 keV). The ground state spin was assigned on the basis of the half-life for decay to the spin 3/2 state in $^{137}\text{Ba}$. The doublets observed in Ge(Li) spectra spaced by 10 keV may be taken as decays to the ground and first excited states. This would suggest levels at 446, 492, 781, 836, 925, and 1004 keV. Taking energy differences, it was seen that two prominent transitions, those at 698 and 762 keV, were not accounted for by these levels. The 762 is definitely not a doublet. Evidence discussed below shows that the
Fig. V.2. $^{137m+g}$ photon spectrum, with a Ge(Li) spectrometer.
Table IX. Energies of transitions following the electron capture and positron decay of Ce$^{137}$.

<table>
<thead>
<tr>
<th>Transition energy (keV)</th>
<th>Following Ce$^{137}$ ground decay, g</th>
<th>Following Ce$^{137m}$ decay, m</th>
<th>From energy level (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1160.3</td>
<td>g</td>
<td></td>
<td>1170.3</td>
</tr>
<tr>
<td>1004.0</td>
<td>m</td>
<td></td>
<td>1004.0</td>
</tr>
<tr>
<td>994.1</td>
<td>m</td>
<td></td>
<td>1004.0</td>
</tr>
<tr>
<td>925.8</td>
<td>g</td>
<td></td>
<td>925.8</td>
</tr>
<tr>
<td>915.8</td>
<td>g</td>
<td></td>
<td>925.8</td>
</tr>
<tr>
<td>835.8</td>
<td>m</td>
<td></td>
<td>835.8</td>
</tr>
<tr>
<td>825.0</td>
<td>m</td>
<td></td>
<td>835.8</td>
</tr>
<tr>
<td>781.5</td>
<td>g</td>
<td></td>
<td>781.5</td>
</tr>
<tr>
<td>771.1</td>
<td>g</td>
<td></td>
<td>781.5</td>
</tr>
<tr>
<td>762.1</td>
<td>m</td>
<td></td>
<td>762.1</td>
</tr>
<tr>
<td>698.0</td>
<td>g</td>
<td></td>
<td>708.0</td>
</tr>
<tr>
<td>511.0</td>
<td></td>
<td></td>
<td>β⁺</td>
</tr>
<tr>
<td>492.5</td>
<td>g</td>
<td></td>
<td>492.5</td>
</tr>
<tr>
<td>481.5</td>
<td>g</td>
<td></td>
<td>492.5</td>
</tr>
<tr>
<td>479.0</td>
<td>g</td>
<td></td>
<td>925.8</td>
</tr>
<tr>
<td>446.5</td>
<td>g</td>
<td></td>
<td>446.5</td>
</tr>
<tr>
<td>436.1</td>
<td>g</td>
<td></td>
<td>446.5</td>
</tr>
<tr>
<td>433.0</td>
<td>g</td>
<td></td>
<td>925.8</td>
</tr>
<tr>
<td>255.8</td>
<td>m</td>
<td></td>
<td>255.8(ce$^{137m}$)</td>
</tr>
<tr>
<td>168</td>
<td>m</td>
<td></td>
<td>1004.0</td>
</tr>
<tr>
<td>10.0</td>
<td>g</td>
<td></td>
<td>10.077</td>
</tr>
</tbody>
</table>
Table X. Photon and conversion electron intensities in Ce$^{137}$ and La$^{137}$.

<table>
<thead>
<tr>
<th>Energy</th>
<th>Relative photon intensity (Ge)$^a$</th>
<th>$(\alpha_x)_{k}^{\text{expt.}}$</th>
<th>$(\alpha_m)<em>{M1}(\alpha_P)</em>{E2}$</th>
<th>Multipolarity</th>
</tr>
</thead>
<tbody>
<tr>
<td>168</td>
<td>4.1</td>
<td>$5.4 \times 10^{-2}$</td>
<td></td>
<td>E1</td>
</tr>
<tr>
<td>255</td>
<td>100.0</td>
<td>$2.5 \times 10^{-2}$</td>
<td>$1.6 \times 10^{-2}$</td>
<td>M4</td>
</tr>
<tr>
<td>436.1</td>
<td>8.0</td>
<td>$1.5 \times 10^{-2}$</td>
<td>$1.2 \times 10^{-2}$</td>
<td>M1+E2</td>
</tr>
<tr>
<td>446.5</td>
<td>49.5</td>
<td>$1.3 \times 10^{-2}$</td>
<td>$1.2 \times 10^{-2}$</td>
<td>M1+E2</td>
</tr>
<tr>
<td>481.5</td>
<td>1.6</td>
<td>--</td>
<td></td>
<td>--</td>
</tr>
<tr>
<td>492.5</td>
<td>0.3</td>
<td>--</td>
<td></td>
<td>--</td>
</tr>
<tr>
<td>511.0</td>
<td>0.6</td>
<td>--</td>
<td></td>
<td>--</td>
</tr>
<tr>
<td>698.0</td>
<td>1.2</td>
<td>$4.6 \times 10^{-3}$</td>
<td>$3.6 \times 10^{-3}$</td>
<td>M1+E2</td>
</tr>
<tr>
<td>762.1</td>
<td>6.7</td>
<td>$1.9 \times 10^{-3}$</td>
<td></td>
<td>M1+E2</td>
</tr>
<tr>
<td>771.1</td>
<td>0.4</td>
<td>--</td>
<td></td>
<td>--</td>
</tr>
<tr>
<td>781.5</td>
<td>0.16</td>
<td>--</td>
<td></td>
<td>--</td>
</tr>
<tr>
<td>825.0</td>
<td>16.6</td>
<td>$2.2 \times 10^{-3}$</td>
<td>$2.5 \times 10^{-3}$</td>
<td>M1+E2</td>
</tr>
<tr>
<td>835.8</td>
<td>3.6</td>
<td>$1.75 \times 10^{-3}$</td>
<td>$2.5 \times 10^{-3}$</td>
<td>M1+E2</td>
</tr>
<tr>
<td>915.8</td>
<td>2.7</td>
<td>$2.0 \times 10^{-3}$</td>
<td>$1.9 \times 10^{-3}$</td>
<td>M1+E2</td>
</tr>
<tr>
<td>925.8</td>
<td>1.6</td>
<td>$2.4 \times 10^{-3}$</td>
<td>$1.9 \times 10^{-3}$</td>
<td>M1+E2</td>
</tr>
<tr>
<td>994.1</td>
<td>0.06</td>
<td>--</td>
<td></td>
<td>--</td>
</tr>
<tr>
<td>1004.0</td>
<td>0.84</td>
<td>$4.7 \times 10^{-3}$</td>
<td>$1.6 \times 10^{-3}$</td>
<td>M2+E3</td>
</tr>
<tr>
<td>1160.3</td>
<td>0.08</td>
<td>--</td>
<td></td>
<td>--</td>
</tr>
</tbody>
</table>

$^a$Using efficiency curve for Ge shown in Fig. II.8.

$^b$E3: $3.4 \times 10^{-3}$
M2: $5.8 \times 10^{-3}$
Fig. V.3. Photon spectrum showing 166 keV γ-ray in $\text{Ce}^{139}$ and 168 keV γ-ray in $\text{Ce}^{137/m}$, taken with a Ge(Li) detector.
Fig. V.4a. Photon spectrum of Ce$^{137m+g}$ showing 433 keV shoulder on 436 keV γ-ray. The 433 keV transition is a stopover from the 925 keV level to the 492 keV level.

Fig. V.4b. Photon spectrum of Ce$^{137m+g}$ showing 479 keV γ-ray. The 479 keV transition is a stopover from the 925 keV level to the 446 keV level.
762 keV transition follows the decay of Ce\(^{137m}\) to La\(^{137}\). This may mean that the 762 represents the decay of a high-spin level, at 762 keV, to the 7/2 ground state, but not the 5/2 first excited state. The 698 and 1160 keV \(\gamma\)-rays follow the decay of Ce\(^{137g}\) to La\(^{137}\). They may be due to the decay of low-spin levels at 708 keV and 1170 keV to the first excited state. Unfortunately, a weak transition at 708 keV might not be seen because it would be obscured by the Compton edges from the higher \(\gamma\)-rays.

In order to determine which levels were fed by the Ce\(^{137g}\) decay, 25\% enriched Ce\(^{136}\) as CeO\(_2\) (from Oak Ridge Isotopes Division) was irradiated with neutrons for six hours at a flux of \(10^{13}\) neutrons cm\(^{-2}\) sec\(^{-1}\) in the Livermore Pool-Type Reactor. This sample was purified by the procedure in Sec. III.A, with 10 mg of La carrier. The ratio of cross sections for thermal neutron capture leading to the ground state and to the 255 keV state has been measured as 6.0 ± 0.6. This was qualitatively confirmed by the ratio of the 436-446 doublet to the 255 keV peak. A spectrum taken 18 hours after the end of the irradiation is shown in Fig. V.5. The complexity of the spectrum is due to \(\gamma\)-rays from Ce\(^{139}\), Ce\(^{141}\), and especially Ce\(^{143}\), also formed in the neutron irradiation. The activity was followed for 10 days. The peaks assigned to Ce\(^{137}\) decayed with a nine-hour half-life. The peaks assigned to Ce\(^{143}\) decayed with a 33-hour half-life. The assignments in Ce\(^{143}\) were checked by irradiating enriched Ce\(^{142}\) as CeO\(_2\) with neutrons. The peaks due to Ce\(^{143}\) are indicated in Fig. V.5.

The \(\gamma\)-rays following the decay of Ce\(^{137g}\) and Ce\(^{137m}\) (by inference) are given in Table IX. Since the spin of Ce\(^{137m}\) is 11/2- and the spin of Ce\(^{137g}\) is 3/2+, the levels in La\(^{137}\) may be expected to divide into two groups; a) levels with spins 1/2, 3/2, and 5/2, fed by the Ce\(^{137g}\) decay and b) levels with spins 9/2, 11/2, 13/2, fed by the Ce\(^{137m}\) decay. A level with spin 7/2 could be very weakly fed by either parent, but the decay would involve 2 units of angular momentum and would be first forbidden (unique) in one case and second forbidden in the other. The low-spin levels (above ground and 10 keV first excited state) are at 446, 492, (708), and 925 and (1170) keV. The high spin levels are at (762), 781, 835, and 1004 keV.
Fig. V.5. Photon spectrum following neutron irradiation of \textsuperscript{93}Ce \textsuperscript{136} [Ge(Li) detector].
3. Positron Branching
A weak 511-keV gamma ray was observed, presumably due to a weak 
β+ branching in the decay of the ground and isomeric states in Ce\textsuperscript{137}. 
The energy difference between the ground states of La\textsuperscript{137} and Ce\textsuperscript{137} has 
been estimated by systematics to be 1.2 MeV.\textsuperscript{81} The β+ branch must occur 
in the decay of the Ce\textsuperscript{137} ground state to the La\textsuperscript{137} 10-keV state. Since 
the ground state is observed to decay to an 1170-keV state in La\textsuperscript{137}, 
there is a lower limit of 1170 keV between the ground states. An upper 
limit on the decay energy may be set using Zweifel's\textsuperscript{82} calculations for 
relative electron capture and positron emission as a function of Z and 
\( W_0 \), the maximum positron energy. For 300 keV and Z=58,

\[
\frac{\lambda_{EC}}{\lambda_{\beta^+}} = 2000.
\]

Since the observed ratio is less than 2000, the positron branch has an 
energy less than 300 keV (\( \lambda_{EC}/\lambda_{\beta^+} \) increases as \( W_0 \) decreases), and an 
upper limit of 1.332 may be set on the ground state energy differences.

Using the upper limit, log ft values for the various levels 
populated in La\textsuperscript{137} were calculated (Table XI). The total number of 
transitions was taken as the 255-keV photon intensity \( \times 8.2 \) (to correct 
for internal conversion) plus the photon intensities for the 1004-, 835.8- 
and 762.1-keV levels, corrected for internal conversion.

4. Conversion Electron Spectroscopy
The conversion electrons from Ce\textsuperscript{137} were studied using a lithium-
drived silicon detector. The detector and electron source were cooled 
to 77\textdegree K to improve the resolution of the detector. The source consisted 
of mass-free Ce\textsuperscript{137} as the chloride, dried onto a Mylar tape backing from 
a drop of solution.

The conversion electron spectrum from the 255 keV transition in 
Ce\textsuperscript{137} is shown in Fig. V.6. The K/L+M ratio was measured as 2.0. The 
higher energy spectra are shown in Figs. V.7 and V.8. These measurements 
were difficult because of the low intensities of the lines and because of 
the pile-up due to the very intense 255-keV transition. Weak sources and 
long runs were required.
Table XI. Log ft for the decay of Ce$^{137m}$ + Ce$^{137g}$ to La$^{137}$.

<table>
<thead>
<tr>
<th>Level</th>
<th>% population</th>
<th>Half-life for decay to level</th>
<th>log (ft) [Ce$^{137m}$ $^{1.3}$ MeV $\rightarrow$ La$^{137g}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1170.3</td>
<td>0.01</td>
<td>39 y</td>
<td>7.7</td>
</tr>
<tr>
<td>1004.0</td>
<td>0.59</td>
<td>240 d</td>
<td>7.3</td>
</tr>
<tr>
<td>925.8</td>
<td>0.51</td>
<td>280 d</td>
<td>7.1</td>
</tr>
<tr>
<td>835.8</td>
<td>1.9</td>
<td>75 d</td>
<td>7.1</td>
</tr>
<tr>
<td>781.5</td>
<td>0.07</td>
<td>5.5 y</td>
<td>8.2</td>
</tr>
<tr>
<td>762.1</td>
<td>0.79</td>
<td>180 d</td>
<td>7.6</td>
</tr>
<tr>
<td>708.0</td>
<td>0.14</td>
<td>2.7 y</td>
<td>8.2</td>
</tr>
<tr>
<td>492.5</td>
<td>0.22</td>
<td>1.8 y</td>
<td>8.1</td>
</tr>
<tr>
<td>446.0</td>
<td>6.8</td>
<td>21 d</td>
<td>6.8</td>
</tr>
<tr>
<td>100.0</td>
<td>89.0</td>
<td>1.6 d</td>
<td>6.2</td>
</tr>
</tbody>
</table>
Fig. V.6. Conversion electron spectrum of the 255 keV transition in Ce$^{137m}$, taken with a Si(Li) detector.
Fig. V.7. Conversion electron spectrum from transitions in Ce$^{137m+g}$ (300-500 keV).
Fig. V.8. Conversion electron spectrum from transitions in Ce$^{137m+g}$ (600-1000 keV).
Using the experimental arrangement described in Sec. II, the conversion coefficients of the prominent transition were directly measured. The results are tabulated and compared with theoretical values \(^{83,84}\) in Table X. All the transitions measured were M1 or E2 or M1+E2, with the exception of the 1004-keV transition which is apparently E3 or M2. The 168-keV conversion coefficient was measured using a source in which the 168 \(\gamma\)-ray intensity was 10 times larger than the 166 \(\gamma\)-ray from Ce\(^{139}\). The background from the 255-keV lines obscured the electron peak, but the \(\gamma\)-ray peak was seen clearly. Using the \(\gamma\)-ray intensity, the electron intensity for an M1, E2 transition was calculated from Rose, \(^{83}\) with the 255 K line as a standard shape. This was subtracted from the background at the position of the expected 168 K line. The resulting background shape was very anomalous as shown in Fig. V.9. All multipolarities greater than E2 require larger coefficients and would therefore be excluded on the same grounds. This is evidence for the multipolarity of the 168-keV transition being E1.

5. Coincidence Studies

Strong coincidences between the 168 keV \(\gamma\)-ray and the 825, 835 pair were recorded with NaI-NaI and Ge(Li)-Ge(Li) detectors. The Ge spectrum is shown in Fig. V.10. No other coincidences except x-rays were observed for either \(\gamma\)-ray.

The 436, 446 pair were found to be in weak coincidence with a transition at 479 keV. Evidence for this transition was seen in the Ge singles spectrum. When the gate was set on the 436, 446 (in Ge or NaI) a peak at 168 keV was seen, presumably due to coincidences with the Compton-scattered \(\gamma\)-rays from the 825, 835 transitions. The 482,492 pair was in coincidence with the 433 keV \(\gamma\)-ray, seen in the Ge singles spectrum. As expected, the 255 keV \(\gamma\)-ray was not in coincidence with any portion of the spectrum.

Stopovers in the high energy group were searched for using NaI as a gate detector and Ge(Li) as the spectrum detector. When the gate was set mainly on the 915, 925 keV peaks, a \(\gamma\)-ray at 85 keV appeared in
Fig. V.9. 168 keV transition conversion electron measurement. The points are experimental. The solid curve is theoretical for an E1 transition. The dashed curve is theoretical for an M1 or E2 transition.
Fig. V.10. Spectrum in coincidence of 168 keV transition in Ce$^{37m}$, showing 825, 825 keV γ-rays (Ge(Li)-Ge(Li) coincidence).
very weak coincidence, along with the 168 keV \( \gamma \)-ray from the coincidences with the forward edge of the 825-835 pair, and x-rays. Shifting the gate higher increased the 85 relative to 168 in the coincidence spectrum. Setting the gate on the 1004 decreased the 85 relative to the x-rays. Thus, the 85 keV \( \gamma \)-ray is in coincidence with the 915, 925 pair.

The 168, 835 coincidence is due to a stopover from the 1004 level to the 835 level. The 436, 446-479 and the 433-482, 492 coincidences are due to stopovers from the 925 level at the 492 and 446 keV levels. The 85-915, 925 coincidence is tentatively assigned to a stopover from the 1004 level to the 925 level. The assignment is tentative because the energy is not quite right—the 1004-925 = 79 keV spacing is known to within 1 keV. Also, this transition is not expected on the basis of the spin assignments discussed below, especially because of transitions that are conspicuously absent, such as stopovers from the 1004 and 835 levels to the 762 level.

6. Spin Measurements: Nuclear Orientation and Angular Correlation

The separation of the La\(^{137}\) energy levels into high-spin levels populated by the decay of Ce\(^{137m}\) and low-spin levels populated by the decay of Ce\(^{137g}\), and the measurement of the multipolarities of the transitions from these levels, allows assignments of spins based on selection rules and systematics. In an attempt to directly measure spins of some of the levels, nuclear alignment experiments were performed using Ge(Li) detectors. This allowed anisotropy measurements to be made on the 436, 446, 698, 762, 835, 825, 915 and 925 keV \( \gamma \)-rays. The 915, 925, 1004, and 1160 keV \( \gamma \)-rays were studied using NaI.

Ce\(^{137m}\) was grown into the bulk of a large, single NES crystal. The crystal was demagnetized and counts were taken at 0\(^\circ\) and 90\(^\circ\) to the crystal axis. No susceptibility readings were taken but the anisotropy of the 255-keV transition was used to monitor temperature. The cold counts were taken for up to 20 minutes. From the anisotropy in the 255 keV \( \gamma \)-ray, \( 1/T \) and \( B_2(11/2) \) were known. This same \( B_2 \) applied to all transitions populated by the decay of the Ce\(^{137}\) 11/2 level. Given \( 1/T \)
and the hyperfine structure constant measured by Haag et al.\textsuperscript{49} \(B_2(3/2)\) could be calculated for all the levels populated by the decay of the Ce\textsuperscript{137} 3/2 level. In this way, \((u_2p_2)\) was obtained for all measured anisotropies. These were compared with \((u_2p_2)\) calculations.

\subsection*{446 Level}

On the basis of the population of this state from the Ce\textsuperscript{137} 3/2 state and the decay to the 7/2 ground state and 5/2 first excited state in La\textsuperscript{137}, this state may have spin 3/2 or 5/2. Both the 446 and 436 keV gamma rays show small positive anisotropies when aligned in NES. This is consistent with either spin assignment as indicated in Table XII and Figs. V.11 V.12, and V.13. The conversion coefficients indicate that the transitions are M1 or E2. This is also consistent with either assignment. Danby et al.\textsuperscript{76} report the 446 transition is E2, but they did not see the 436 keV transition and this compromised their data. Haag\textsuperscript{36,49} measured the linear polarization of the combined 436 and 446 keV \(\gamma\)-rays, using a Compton polarimeter. He found a polarization \(P \approx 1.5\) at \(1/T = 50\), although there was quite a bit of scatter in the data. A polarization \(P > 1.0\) excludes a pure E2 transition (for a pure E2, \(0.80 \leq P \leq 0.94\), depending on \(u_2\)). Since the 446 keV \(\gamma\)-ray is 6 times larger than the 436 keV \(\gamma\)-ray, we may assume that the polarization result excludes E2 for the 446 keV \(\gamma\)-ray. This excludes spin 3/2 for the 446 keV level. On the basis of Haag's polarization data, then, \(I(446) = 5/2\).

\subsection*{492 Level}

This level is weakly fed from the decay of Ce\textsuperscript{137}g and it decays to both the ground and first excited states in La\textsuperscript{137}. It is also fed by stopovers from the 925 keV level. Little more can be said about this level. It is too weakly populated for conversion coefficient or nuclear alignment measurements. The fact that it decays to a 5/2\(^+\) and a 7/2\(^+\) level, and is fed from a 3/2\(^+\) level suggests that its spin is \(I_{492} = 3/2^+, 5/2^+,\) or 7/2\(^+\).
Table XII. Nuclear alignment results for the 436, 446 keV γ-rays following decay of Ce$^{137g}(3/2^+)$.  

<table>
<thead>
<tr>
<th></th>
<th>436 keV</th>
<th>446 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(B_{2}U_{2}F_{2})_{\text{ext}}$</td>
<td>$-0.0875 \pm 0.02$</td>
<td>$-0.054 \pm 0.015$</td>
</tr>
<tr>
<td>$(U_{2}F_{2})_{\text{expt}^{(\mu}\text{Ce}^{137g}=1}^{(1/T=46)}$</td>
<td>$-0.120 \pm 0.02$</td>
<td>$-0.074 \pm 0.015$</td>
</tr>
</tbody>
</table>

If $I_{446} = 3/2$,

<table>
<thead>
<tr>
<th>$(U_{2})_{\text{theor}}$</th>
<th>See Fig. V.11 for $F(8)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(F_{2})_{\text{theor}}$</td>
<td>$-0.143$ (EZ)</td>
</tr>
</tbody>
</table>

If $I_{446} = 5/2$,

<table>
<thead>
<tr>
<th>$(U_{2})_{\text{theor}}$</th>
<th>$0.75$ $(j_{\mu}=1)$</th>
<th>$0.75$ $(j_{\mu}=1)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(F_{2})_{\text{theor}}$</td>
<td>See Fig. V.13 for $F(8)$</td>
<td>See Fig. V.14 for $F(8)$</td>
</tr>
</tbody>
</table>
Fig. V.11. Calculated $F_2$ vs $\delta$ curves for a $\frac{3}{2} \rightarrow \frac{1}{2}$ transition. The shaded region corresponds to the experimental $F_2$. 
Fig. V.12. Calculated $F_2$ vs $\delta$ curves for a $5/2 \rightarrow \frac{7}{2}$ transition. The shaded region corresponds to the experimental $F_2$. 
Fig. V.13. Calculated $F_2$ vs $\delta$ curves for a $5/2 \rightarrow 3/2 \rightarrow 5/2$ transition. The shaded region corresponds to the experimental $F_2$. 

- $F_2 (111/2, 5/2) = -0.428$
- $F_2 (121/2, 5/2) = -0.507$
- $F_2 (221/2, 5/2) = +0.191$
708 Level

The 708 keV level is populated by the decay of the Ce\textsuperscript{137} ground state. It decays by a single transition presumably to the first excited state in La\textsuperscript{137}. Because no 708 keV transition to ground is observed, this level may be tentatively assigned spin 1/2+. The γ-ray was observed from Ce\textsuperscript{137} nuclei aligned in NES. Because of low counting rate and large background corrections, the data were very poor, but indicated an effect of 0.980 ± 0.040, consistent with a 1/2+ assignment.

762 Level

This level is populated by the decay of Ce\textsuperscript{137m} (11/2-). It de-excites by a single transition, presumably to the La\textsuperscript{137} 7/2+ ground state. Conversion coefficient measurements show that the transition is M1, E2 or M1 + E2. When aligned in NES, the 762 keV γ-ray was found to have a large negative U\textsubscript{2}F\textsubscript{2}, consistent with a 11/2+ → 7/2+ transition (Table XII). It is also consistent with 9/2+ → 7/2+ transition (Fig. V.14), but a 9/2 spin is unlikely on the grounds that the 9/2+ → 5/2+ transition is not observed. Thus the spin of the 762 level is assigned I\textsubscript{762} = 11/2+.

781 Level

Because it is populated by the decay of the 3/2+ state in Ce\textsuperscript{137} and has transitions to both the 7/2+ and 5/2+ ground and first excited states in La\textsuperscript{137}, the 781 keV level may be 3/2+, 5/2+, or 7/2+. In the conversion electron spectrum, the 781 and 771 K lines were obscured by the 762 L-M lines. The state was too weakly populated for spin determination by nuclear alignment.

835 Level

On the basis of systematics, the 835 keV level may be expected to be 9/2+. It is populated by the decay of the 11/2- state in Ce\textsuperscript{137} and in turn decays to the ground and first excited states in La\textsuperscript{137}, with M1, E2 or M1 + E2 transitions. Nuclear alignment in NES showed that the U\textsubscript{2}F\textsubscript{2} for the 835 keV γ-ray was positive, while that for the 825 keV γ-ray was negative. These observations support the 9/2+ assignment. The 825 keV
Table XIIIa. Nuclear alignment results for 762, 825, and 835 keV γ-rays.

<table>
<thead>
<tr>
<th></th>
<th>762</th>
<th>825</th>
<th>835</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(U_2 F_2)_{\text{expt}}$</td>
<td>$-0.41 \pm 0.08$</td>
<td>$-0.39 \pm 0.06$</td>
<td>$+0.53 \pm 0.12$</td>
</tr>
<tr>
<td>$(U_2 F_2)_{\text{theor}}$</td>
<td>$11/2 \rightarrow 9/2$</td>
<td>$11/2 \rightarrow 9/2$</td>
<td>$11/2 \rightarrow 9/2$</td>
</tr>
<tr>
<td></td>
<td>$2 \rightarrow 7/2$</td>
<td>$2 \rightarrow 5/2$</td>
<td>$1/2 \rightarrow 7/2$</td>
</tr>
<tr>
<td></td>
<td>$-0.40 \pm 0.01$</td>
<td>$-0.36$</td>
<td></td>
</tr>
</tbody>
</table>

See Fig. V.15 for $F(\delta)$ for $F(\delta)$.

Gives $\frac{\delta^2}{1+\delta^2} = 0.01 \pm 0.025$ or $\frac{\delta^2}{1+\delta^2} = 0.93 \pm 0.86$.

$\delta < 0$
Fig. V.14. Calculated $F_2$ vs $s$ curves for a $9/2 \rightarrow 1/2 \rightarrow 7/2$ transition. The shaded region corresponds to the experimental $F_2$. 
Fig. V.15. Coincidence rate for 168-825,835 keV cascade in Ce$^{137+}$m as a function of $P_2(\cos \theta)$. 

Normalized coincidence rate

$P_2 (\cos \theta)$
transition is pure E2 and the 835 keV γ-ray is mixed M1, E2, for which \( \beta < 0 \) (Fig. V.14). Further evidence for the 9/2+ assignment is found in an angular correlation experiment between the 835, 825 keV γ-rays and the 168 keV γ-ray, discussed below.

925 Level

Since the 925 keV level is fed from the ground state of Ce\(^{137}\) and transitions from it to the ground and first excited states of La\(^{137}\) are M1, E2, or M1 + E2, the spin of this level may be 5/2+ or 3/2+. Nuclear alignment experiments in NES were inconclusive, for both the 915 and 925 keV γ-rays showed anisotropies very close to zero even at 1/T=60.

1004 Level

After population by the electron capture of the Ce\(^{137}\) 11/2− state, the 1004 keV level is observed to decay to the La\(^{137}\) ground state, with a weak stopover at the 10 keV state. The conversion coefficient measurements show that the 1004 transition is M2 or E3 or M2 + E3. In addition, the 168 transition to the 830 keV level is E1 (see above). Anisotropy measurements were made on the 1004 keV γ-rays (the 994 keV γ-ray was too weak for measurement), following the decay of Ce\(^{137m}\) nuclei aligned in NES, and on the 168 keV γ-ray for Ce\(^{137m}\) in CMN. The 1004 keV γ-ray data were poor statistically, but indicated a \( U_{21}^E < 0 \) and consistent with a 11/2 → 7/2 transition. The 168 keV γ-ray effect was consistent with a 11/2 → 9/2 transition.

The assignments were confirmed by measuring the angular correlation between the 168 and 825, 835 keV γ-rays. For the cascade

\[
11/2^- \rightarrow E1 \rightarrow 9/2^+ \rightarrow E2 \rightarrow 5/2^+, A = -0.07
\]

and \( \epsilon = 1 - C_{180}/C_{90} \), where \( C \) is the coincidence rate at angle \( \theta \), is +0.10. For the cascade

\[
9/2^- \rightarrow E1 \rightarrow 9/2^+ \rightarrow E2 \rightarrow 5/2^+, \epsilon = -50\%
\]

Experimentally, \( \epsilon = +5\% \), confirming the 11/2− spin assignment.
to the 100.4 keV level. The reduction in anisotropy is due to the effect of the weaker 835 keV \( \gamma \)-ray, which has an \( F_2 \) of opposite sign relative to the 825 keV \( \gamma \)-ray. (See Table XIIIb.)

1170 Level

Ce\(^{137}\)g \((3/2^+)\) decay weakly populates the level at 1170 keV. Only one transition from the level is observed, presumably to the first excited state. Conversion lines were not seen, due to the weakness of the transition. Anisotropy measurements from oriented nuclei were attempted using a NaI detector. Statistically poor data indicate zero anisotropy. These observations are consistent with a spin of 1/2+, but the assignment is tentative only.

7. Comparison with Theory

La\(^{137}\) (Z=57, N=80) has two holes in the closed neutron shell at \( N=82 \) and seven protons beyond the closed proton shell at \( Z=50 \). The protons in this region fill into the \( 1g_{7/2}, 2d_{5/2}, 2d_{3/2}, 3s_{1/2}, \) and \( 1h_{11/2} \) single particle states. The La\(^{137}\) ground state would be \( 7/2^+ \), with the \( 5/2^+ \) state close to it. The next state \((446 \text{ keV})\) may be expected to be \( 1/2^+ \) or \( 3/2^+ \). The higher-lying states may be ascribed to collective effects, expected at high energy for spherical nuclei. Clearly, this simple model is inadequate for more than the first two, or possibly three levels.

A more sophisticated approach has been used by Kisslinger and Sorenson \(^{85}\) (KS), who have calculated the energy levels for spherical nuclei using a pairing force between protons and neutrons separately and a quadrupole interaction between all pairs of particles. Using a set of single particle energies determined from experiment (assuming they vary with mass as \( A^{-2/3} \)), and appropriate choices for force constants, they have fitted the low-lying energy levels over a wide portion of the periodic table. Two types of excitations arise from the theory: quasi-particle excitations due to the pairing interaction, and phonon excitations due to the quadrupole force. Rho \(^{86}\) has calculated the theoretical quasi-particle energies for 82 neutron, odd-\( A \) nuclei. He finds a \( 7/2^+ \) lowest,
<table>
<thead>
<tr>
<th>Nuclear alignment:</th>
<th>168 keV</th>
<th>1004 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>((U_{2}F_{2})_{\text{expt}})</td>
<td>0.245 ± 0.008</td>
<td>-0.30 ± 0.15</td>
</tr>
<tr>
<td>(J=2)</td>
<td>7/2 - 1/1 → 9/2⁺</td>
<td>9/2⁺ → 11/2⁻</td>
</tr>
<tr>
<td>+0.136</td>
<td>(J=1)</td>
<td>9/2⁻ → 11/2⁻</td>
</tr>
<tr>
<td>-0.418</td>
<td>(J=1)</td>
<td>9/2⁻ → 11/2⁻</td>
</tr>
<tr>
<td>((U_{2}F_{2})_{\text{theor}})</td>
<td>0.288 ((J=0))</td>
<td>-0.40 ± 0.01</td>
</tr>
<tr>
<td>0.274 ((J=1))</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

| Angular correlation for 168 keV - 825,835 keV cascade: |
| Theory | \(A_2\) | \(\epsilon(=1-C_{180}/C_{90})\)% |
| 11/2⁻ → 9/2⁺ | \(J=2\) | 5/2⁺ |
| \(E_{2,M1}\) | 825 | 7/2⁺ |
| -0.043 | = +6.34% |
| 9/2⁻ → 9/2⁺ | \(J=2\) | 5/2⁺ |
| \(E_{2,M1}\) | 825 | 7/2⁺ |
| +0.115 | = -17.2% |
| Experiment | -0.033 ± 0.10 | +5.0 ± 1.5% |
followed by $d_{5/2}$, $h_{11/2}$, and $d_{3/2}$, for La$^{139}$. In the absence of the quadrupole force, KS find $g_{7/2}$ lowest, followed by $d_{5/2}$ and $s_{1/2}$, the latter between 1 and 2 MeV. Introduction of the quadrupole force brings $5/2$ lower than $7/2$ for La$^{137}$, in disagreement with the experiment. The energy levels for La$^{137}$ according to KS are shown in Fig. V.17.

Comparison with the experimental levels shown in Fig. V.17 shows qualitative similarities. There is a gap of about 500 keV between the ground and first excited states and the next level. The level density increases above about 700 keV, although this is hardly unexpected. Eleven levels are found experimentally, while twelve are predicted. Of particular interest is the $11/2^-$ state found at 1004 keV. This is undoubtedly the $h_{11/2}$ single quasi-particle state found at about 1 MeV by Rho for La$^{139}$. Comparing the 168 keV $E1$ transition rate and the 1004 keV $E3$ transition rate with the theoretical rates for single particles, we see that the $E1$ transition is hindered by two orders of magnitude relative to the $E3$. This is unusual because the $E3$ transition would be expected to be $\ell$-forbidden in the single particle model.
Fig. V.16. Decay scheme for Ce$^{137m+g}$. 
Fig. V.17. Level structure of La$^{137}$ from calculations by Kisslinger and Sorensen. Also shown are experimentally determined levels and possible correlations.
VI. THE ANGULAR DISTRIBUTION OF ALPHA PARTICLES FROM E\textsuperscript{253} ALIGNED IN NES AND THE RELATIVE PHASES OF THE L=0 AND L=4 WAVES

1. Introduction

E\textsuperscript{253} was aligned in NES by Navarro\textsuperscript{2,87} who studied the temperature dependence of the angular distribution of alpha particles in the decay to Bk\textsuperscript{249}. He determined that the L=0 and L=2 waves were in phase. Further experiments were conducted in order to determine the relative phases of the L=0 and L=4 waves.

Section VI.2 below constitutes a short theoretical and experimental survey of alpha particle emission by prolately deformed nuclei. Section VI.3 treats the angular distribution of alpha particles from aligned nuclei, and Sec. VI.4 considers previous experiments. The present experiments are presented in Sec. VI.5 and discussed in Sec. VI.6.

2. Background

Alpha particles from a nuclear decay are emitted with characteristic energies, corresponding to the energy levels populated in the daughter nucleus by the decay. Thus, alpha spectra have provided a wealth of information about the level structure of nuclei in the regions where alpha emission is a predominant mode of nuclear disintegration.\textsuperscript{88} In addition to carrying off energy, the alpha particle can also carry off angular momentum. Since alpha particles are bosons, with no spin, they carry only orbital angular momentum, L. Consider \( I_p \rightarrow L \rightarrow I_f \), where \( I_p \) is the spin of the parent, \( I_f \) is the spin of the level populated in the daughter and \( L \) is the angular momentum carried off the alpha particle. Conservation of angular momentum requires that:

\[
|L - I_f| \leq L \leq I_p + I_f \quad \text{ (VI.1)}
\]

In \( \gamma \)-ray emission, the interaction is strongly dependent on L, so that only lowest allowed multipolarity is found, with vanishingly small probabilities for higher multipoarities. For example, where L=1 and L=2 are allowed, they will compete with each other, but L=3,4 etc. will not
compete in general. In alpha decay, essentially a barrier penetration process, the only difference between L and L+2 emission is a centrifugal barrier which is small compared to Coulomb barrier. This additional barrier is large enough to ensure that the lowest value of L will have the largest probability of emission, but is small enough to allow considerable admixture of L+2 and L+4 into the decay (when they are allowed by Eq. (VI.1)). Furthermore, since alpha decay is a strong interaction, parity is conserved. The parity of the outgoing alpha is given by \((-1)^L\), where \(L\) is the orbital angular momentum of the wave. Therefore, for decays between states of the same parity, only even values of \(L\) are allowed, while for decays between states of different parity, only odd values of \(L\) are allowed.

If \(\pi_i \longrightarrow \pi_f\), where \(\pi_i\) and \(\pi_f\) are the initial and final state parities, respectively, then

\[
\pi_i \pi_f = (-1)^L
\]  

(VI.2)

Simple alpha decay theory defines the decay constant \(\lambda (=\ln2/t_{1/2})\) as a product \(\lambda = fP\), where \(P\) is the penetration factor and \(f\) is the "reduced transition probability." In the simplest theory, the alpha particle is regarded as existing preformed in the daughter and \(f\) is a frequency factor—the number of "collisions" per unit time with the barrier. \(P\) is the probability that each "collision" will lead to emission.

The factor \(P\) can be calculated and the experimental results of alpha spectroscopy given in terms of the \(f\)'s. For the spherical even-even isotopes of a given element, \(\lambda\) shows a smooth functional dependence on \(E\) for the ground state transitions, which is due to the factor \(P\) only. The odd-neutron isotopes of the same element and the excited state transitions of the even isotopes may show deviations in the \(\lambda\) vs \(E\) curve obtained from the ground transition in the even-even isotopes. This deviation is uniformly in the direction of smaller \(\lambda\) and the transitions may be said to be hindered. The amount of deviation is described by a "hinderance factor." For odd-Z nuclei, "theoretical" \(\lambda\) vs \(E\) curves can be obtained by interpolating between neighboring even-even nuclei, and the
experimental λ's relative to these curves are also described in terms of
hindrance factors. The transition coming closest to the "theoretical"
line for a given nucleus is termed the favored transition.

In the region of nuclei for which N > 138, the systematics of
nuclear energy levels and nuclear transition have best been described in
terms of the "strong coupling" model of Bohr and Mottelson, 89 which con-
siders nuclei in this region to have stable, spheroidal deformations.
For even-even nuclei in this region, the lowest-lying levels are due to
rotational excitation and have energies which can be expressed as:

\[ E \propto I(I+1) \]

In this rotational band, the 0+ ground state is followed by levels with
spins and parities 2+, 4+, 6+, etc. (in some cases, a 1- excitation is
also observed 88), and the alpha spectrum of the decay of the 0+ ground
state in the parent to this band in the daughter shows the most intense
line at highest energy, going to the ground state \((0+ \overset{L=0}{\rightarrow} 0^+\)), followed
in intensity by transitions to the first excited state \((0^+ \overset{L=2}{\rightarrow} 2^+)\),
second excited state \((0^+ \overset{L=4}{\rightarrow} 4^+)\) and so forth.

In the case of odd nuclei in the same region, excitation energies
of the odd particle can be of the same order of magnitude as the collective
excitation energies. Rotational spectra are, however, observed, but built
on various single nucleon states. These single nucleon states have been
calculated for the case of a spheroidally deformed potential by Nilsson. 90
Since the potential is no longer spherical, the orbital angular momentum
of the odd nucleon is no longer conserved, but its projection on the
nuclear deformation axis is a constant of the motion and is generally
denoted by K. K is also the projection of I on the symmetry axis because
rotational angular momentum, denoted \( \bar{R} \); is perpendicular to this axis.
States are labeled as \( KI\pi \), where the \( I \) is the spin of the state (odd
nucleon spin plus rotational spin) and \( \pi \) is the parity (determined by
the value of \( I \) the odd nucleon would have in the limit of sphericity).
The rotational bands have energies \( E \propto I(I+1) \), where \( I=K \), \( I=K+1 \), etc.
The alpha spectra for these nuclei are unlike the spectra of even-even alpha emitters in that the most intense transition is not always the highest energy transition—that is, the ground level of the daughter is not always the most highly populated. In view of barrier penetration nature of the decay, this is surprising, but the observation has been shown to be compatible with the theory. Here, the $f$ in the relation $\lambda = fF$ may be thought of as a formation factor. It has been suggested that the formation of the alpha particle before transmission through the barrier would be most probable if it did not involve the single odd nucleon in the parent. The alpha particle would be formed from paired nucleons and would not involve unpairing and recoupling. Thus, the odd nucleon would occupy the same state in the daughter as in the parent and the $K$ quantum number would not change. Then the greatest probability for decay would be to the rotational band for which $\Delta K = 0$. Of course, for some nuclei the intrinsic state for the odd nucleon in the parent might be the ground intrinsic state for the odd nucleon in the daughter.

Bohr, Broman, and Mottelson have introduced some branching relations for relative intensities of decay to the various members of a rotational band in the daughter. In addition to the fact that $I_1^Z = I_f^Z$, the relation $I_1^M + M_L = K_f$ also holds, where $M_L$ is the projection of the alpha orbital angular momentum on the nuclear symmetry axis. The final state wave function can be expanded in terms of the initial state wave function and the alpha wave functions as:

$$\langle I_1K_1K_f-K_1 | I_1K_f \rangle = \sum L \langle I_1LK_1K_f-K_1 | I_1K_f \rangle \langle I_1K_1 | L K_f-K_1 \rangle$$

where $\langle I_1LK_1K_f-K_1 | I_1K_f \rangle$ is a Clebsch-Gordan coefficient. For a given alpha wave of angular momentum $L$,

$$\langle I_1K_f \rangle = \langle I_1LK_1K_f-K_1 | I_1K_f | I_1K_1 \rangle \langle I_1K_1 | L K_f-K_1 \rangle$$.

The "reduced" probability that the transition proceeds to a given state $I_f$ by emission of a wave of angular momentum $L$ is just:
For favored decay, \( K_i = K_f \) and \( \lambda \propto \langle I_L K_\lambda | K_\lambda - K_i | I_L K_f \rangle^2 \).

For a spheroidal shape, the total probability for formation at the nuclear surface of \( L \) waves where \( L > 0 \) may be estimated by introducing the reduced transition probabilities \( C_L \), for the \( L \) wave measured in neighboring even-even nuclei. Thus

\[
\lambda \propto P_0(Z, E, T) \sum L C_L \langle I_L | K_\lambda | I_L \rangle^2.
\]  

(VI.3)

This relation has been widely used to analyze the intensities in the favored transition of odd-\( A \) nuclei. The results give the relative \( L=0, L=2, L=4 \), etc. wave intensities to the various levels in the favored band.

The alpha wave angular momentum is conserved only in the case of a spherically symmetric Coulomb potential barrier. Several authors have considered the effects of the nuclear quadrupole moment in mixing states of different \( L \). The calculation leads to coupled differential equations which have been solved numerically for some cases. For \( U^{233} \), Chasman and Rasmussen find a 20% enhancement of the \( L=2 \) wave in the ground state transition over the Bohr,Froman and Mottelson prediction. This agrees with systematic deviations observed experimentally.

Waves of the same energy can interfere constructively or destructively and such interference is expected between the waves of different \( L \) contributing to a given transition. This interference will be reflected in the distribution of alpha particles at the nuclear surface. Brussard has shown that the distribution at the surface can be related directly to distribution after transmission through the barrier.

From time reversal invariance, it can be shown that the reduced matrix elements for the emission of various alpha \( L \) waves are real. Therefore, the waves can have relative phases of 0 or \( \pi \). Brussard and Tolhoek have considered the phase shift due to transmission through the Coulomb barrier and conclude that the effect is small. Thus, the waves may be taken as in or out of phase.
Another approach to the decay of prolately deformed nuclei was taken by Hill and Wheeler. They calculated that the major effect would be due to the anisotropy of the Coulomb barrier. If the nuclear center of charge coincides with the nuclear center-of-mass, the barrier would be lower at the "tips" of the nucleus and higher at the "waist." If the probability of alpha formation at the nuclear surface is isotropic, then preferential emission occurs at the tips. Steenberg and Sharma have, however, pointed out that this argument is weak in that it assumes an isotropic formation probability inside the nucleus. Brussard suggests that the major effect of spheroidal deformation is an anisotropic probability distribution at the nuclear surface, before transmission.

3. Angular Distribution of Alpha Particles from Oriented Nuclei

The angular distribution of alpha particles from oriented nuclei provides a means of determining the relative amplitudes and phases of the different L waves in a given alpha transition.

As discussed previously (Sec. II) the calculation of the angular distribution of alpha particles from oriented nuclei follows from the expression for the distribution of \( \gamma \)-rays with the introduction of particle parameters, \( b_k^* \),

\[
W(\theta) = 1 + \sum_{k} b_k^{(LL')}(LL') R_k f_k^{(LL'I'I')} P_k(\cos \theta). \tag{II.8}
\]

For alpha particles,

\[
b_k^{(LL')}(LL') = \frac{-2\sqrt{L(L+1)(L')(L'+1)}}{k(k+1)-L(L+1)-L'(L'+1)} \tag{VI.4}
\]

It can be seen that the \( b_k \)'s vanish for interferences involving an \( L=0 \) wave. The \( L=0 \) wave itself is isotropic, and hence contributes only a constant to the angular distribution. It does, however, interfere with other \( L \) waves. To calculate the interference term, it is necessary to go back to the definitions of \( b_k \) and \( f_k \) defined for \( \gamma \)-rays [Eq. (II.4)]
\[ F_k(\ell\ell';ab) = (-1)^{a-b-1/2} 2^{b-1} (\ell\ell';1-1|k0) W(bbL_L,L_1;ka) \]

where \((\ell\ell';1-1|k0)\) is a Clebsch-Gordan coefficient and \(W(bbL_L,L_1;ka)\) is a Racah coefficient. The \(F_k\) was defined for photons, which have spin 1.

The particle parameter for the alpha particle merely reflects its spinless nature, and

\[ b_k = - \frac{(\ell\ell';00|k0)}{(\ell\ell';1-1|k0)} \]  \hspace{1cm} (VI.5)

For \(L'=0\), the denominator is zero. Taking the combined expression

\[ b_k F_k(\ell\ell';ab) = (-1)^{a-b} \sqrt{2b+1} \sqrt{2\ell+1} (\ell\ell';00|k0) W(bbL_L,L_1;ka) \]  \hspace{1cm} (VI.6)

we can calculate the interference.

If the resolution of the detector is sufficiently high so the transitions to the various daughter states are observed independently, the expression for \(W(\theta)\) becomes

\[ W(\theta) = 1 + \sum_{k \text{ even}} \left[ \frac{\sum_{\ell} |a_{\ell}|^2 b_k(\ell\ell)L_F(\ell\ell) + |a_L||a_{\ell}| \phi_{\ell\ell'} b_k^{(\ell\ell')} F_k^{(\ell\ell')} }{1/\sum_{\ell} |a_L|^2} B_{kk}(\cos \theta) \right] \]  \hspace{1cm} (VI.7)

where \(a_{\ell}\) is the amplitude of the \(\ell\)th wave and \(\phi_{\ell\ell'}\) is the relative phase of the \(\ell\) and the \(\ell'\)th waves (=1, in phase; = -1, out of phase). For example, if in a given case \(L=0, 2, 4\) compete,

\[ W(\theta) = 1 + \sum_{k \text{ even}} \left[ |a_0|^2 b_k^{(02)} F_k^{(02)} + |a_1|^2 b_k^{(44)} F_k^{(44)} + 2 |a_2||a_1| b_k F_k(02) \phi_{02} \right. \]

\[ + 2 |a_4||a_0| b_k F_k(04) \phi_{04} + 2 |a_2||a_1| b_k F_k(24) \phi_{24} \]

\[ \left. \times \frac{1}{|a_0|^2 + |a_2|^2 + |a_4|^2} B_{kk}(\cos \theta) \right] \]
Note that only two phase relations are required—that is, if $\phi_{02}$ and $\phi_{04}$ have been assigned, $\phi_{24}$ is determined ($\phi_{24} = \phi_{02} \times \phi_{04}$). In addition, we see from the definition of $b_k^F$ that $(LL'00|k0)$ must be non-zero. This is satisfied only if $L''=k$. Thus the $(0,2)$ term is found for $K=2$ only, with $(2,2), (2,4),$ and $(4,4)$. The $K=4$ term has $(0,4), (2,2), (4,4),$ and $(2,4)$ contributions. The $K=6$ term would have $(4,4)$ and $(2,4)$ contributions but is usually neglected because $B_6$ is generally small.

If the detectors cannot resolve transitions to close-lying states, it is necessary to take a weighted average of the distributions calculated for the various states. If $A_{I'_f}$ indicates the intensity of the transition to the state $I'_f$, then

$$W(\theta) = \sum_{I'_f} \frac{A_{I'_f} w(\theta, I'_f, I''_f, L''_f \rightarrow I''_f)}{\sum A_{I''_f}}$$ (VI.8)

The important point here is that there is no interference between waves going to different final states (incoherent mixture).

4. Previous Experiments

Roberts and Dabbs et al. 98-102 aligned $^{233}\text{U}$ as $\text{UO}_2^{+2}$ and $^{237}\text{Np}$ as $\text{NpO}_2^{+2}$ in uranyl rubidium nitrate. In this case, the alignment was due to coupling between the quadrupole moment of the $^{233}\text{U}$ ($^{237}\text{Np}$) and the field gradient due to the $0-\text{U}-0$ ($0-\text{Np}-0$) bonds which lie parallel to the crystalline $c$ axis. At low temperatures, enhanced alpha emission was observed perpendicular to the $c$ axis. In addition, they measured the quadrupole coupling constant $P$ to be positive for $^{237}\text{Np}$. By assuming $P-\pi$ bonding in the neptunium ion, and a prolate deformation ($Q > 0$), they calculated that the nuclear alignment axis was perpendicular to the $c$ axis and that the alpha particles were preferentially emitted along the nuclear symmetry axis. On the other hand, by invoking predominantly $\sigma$ bonding in the ion, Pryce 103 suggested that the positive $P$ implied the quadrupole moment was negative, and that the nuclear alignment was along the $c$ axis.
Robert and Dabb's interpretation suggests confirmation of the Hill-Wheeler theory, or that the \( L=0 \) and \( L=2 \) waves are in phase at the nuclear surface.

Recently, Navarro et al.\(^{25,87}\) aligned \(^{253}\text{E}^3\) and \(^{249}\text{Cf}^3\) in neodymium ethylsulfate. Experiments on the \( 4f \) homologs of \( \text{E}^3 \) and \( \text{Cf}^3 \), \( \text{Ho}^3 \) and \( \text{Dy}^3 \), respectively, showed that the alignment was axial in each case.\(^{25,104}\) For \( (\text{E}^{253})^3 \), this was confirmed experimentally by measurement of the temperature dependence of the anisotropy while for \( \text{Cf}^3 \), Navarro measured the anisotropy of the 340 and 394 keV \( \gamma \)-rays succeeding the alpha decay. The \( \gamma \)-ray anisotropies were found to be attenuated due to reorientation effects during the nuclear recoil, but from the sign of the anisotropies and their temperature dependence, plus the knowledge that they were both \( M1 \), Navarro inferred that \( B_2 \) was positive, and hence the alignment was axial.

In both cases, anisotropic emission of alpha particles was observed, and in each case there was preferential emission along the nuclear symmetry axis, assuming prolate deformations. From these results, Navarro concluded that the experiments supported the Hill-Wheeler prediction and that the \( L=0,2 \) waves were in phase.

5. **Experiments on \( \text{E}^{253} \)**

Further nuclear alignment experiments on \( \text{E}^{253} \) were conducted as an extension of Navarro's work in order to determine the relative phases of \( L=0 \) and \( L=4 \) waves. This quantity can be determined by a measurement of the sign and magnitude of the coefficient of the \( P_4 \) term in the angular distribution of the alpha particles. This requires simultaneous measurements at \( 0^\circ \) and \( 90^\circ \). Most of Navarro's data were taken at \( 0^\circ \) and \( 90^\circ \) independently. From his simultaneous measurements, he tentatively concluded that the \( L=4 \) wave was out of phase with the \( L=0 \) and \( L=2 \) waves, although the measurement was not conclusive.

Asaro et al.\(^{105}\) have made extensive measurements of the relative intensities of the alpha groups emitted by \( \text{E}^{253} \). The decay scheme is shown in Fig. VI.1. With the intensity data for the rotational band
<table>
<thead>
<tr>
<th>State</th>
<th>Pop. (%)</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Omega = 1 ) \text{[N n_2 \Lambda]} \</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( 9/2 )</td>
<td>0.01</td>
<td>230</td>
</tr>
<tr>
<td>( 11/2 )</td>
<td>0.08</td>
<td>155.8</td>
</tr>
<tr>
<td>( 13/2 )</td>
<td>0.85</td>
<td>93.4</td>
</tr>
<tr>
<td>( 9/2 )</td>
<td>6.6</td>
<td>41.7</td>
</tr>
<tr>
<td>( 7/2 + 3/2 ) [633]</td>
<td>90</td>
<td>3/2 - 1/2 [521]</td>
</tr>
</tbody>
</table>

\[ \text{Fig. VI.1. Decay scheme for } {}^{253}_{\text{E}} \]
populated by the favored decay, they have used the Bohr-Froman-Mottelson (BFM) relations and the reduced L-wave formation probabilities measured in neighboring Cf$^{252}$ and Fm$^{254}$ to calculate the L=0, 2, and 4 wave intensities in the ground transition and the L=2 and 4 wave intensities for the higher states in the band. The results are given in Table XIV.

Using Asaro's calculations, the angular distribution of alphas for aligned E$^{253}$ can be calculated from Eqs. (VI.7 and VI.8), taking \( \phi_{0,2} \) as +1 (determined by Navarro) and \( \phi_{0,4} \) as ±1.

This band accounts for 97.54\% of the total decays. The effect due to decays populating the bands built on states at 8.8 keV and 395 keV can be shown to be negligible and they are therefore omitted from the calculation. The results are:

\[
\begin{align*}
L=0,4 \quad \text{out of phase} & \quad W(\theta) = 1 + 0.594 B_2 P_2(\cos \theta) - 0.0063 B_4 P_4(\cos \theta) \\
L=0,4 \quad \text{in phase} & \quad W(\theta) = 1 + 0.698 B_2 P_2(\cos \theta) + 0.138 B_4 P_4(\cos \theta)
\end{align*}
\]

The major contribution to the \( P_2(\cos \theta) \) term comes from the L=0, L=2 interference. In the coefficient of the \( P_4 \) term, the L=2 term and the L=0, L=4 interference terms are of about equal magnitude. The sign of the L=2 term, however, is unique, while the sign of the L=0, L=4 term depends on the choice of phase. If we included a \( P_6 \) term we would have to consider L=0, L=6 as a possible contribution, with another ambiguity in phase. Since \( B_6 \) is small even at 1/T=60 (\( B_6 = 0.127 \)) and since the L=6 wave intensity is probably small compared to the L=4 wave intensity, this term in the expansion of \( W(\theta) \) has been discarded.

E$^{253}$ has spin 7/2 for axial alignment, only \( I_z = \pm 7/2 \) are populated at lowest temperatures. The limiting or saturation values of \( B_2 \) and \( B_4 \) are

\[
\begin{align*}
B_2(7/2)_{\text{satn}} & = 1.527 \\
B_4(7/2)_{\text{satn}} & = 0.797
\end{align*}
\]
Table XIV. Intensities for partial waves in alpha transitions to the favored band in Bk\textsuperscript{249} (BFM).

<table>
<thead>
<tr>
<th>Final state energy</th>
<th>Final state spin</th>
<th>L=0(%)</th>
<th>L=2(%)</th>
<th>L=4(%)</th>
<th>Total</th>
<th>Expt.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>7/2</td>
<td>79.6</td>
<td>10.0</td>
<td>0.127</td>
<td>89.7</td>
<td>90</td>
</tr>
<tr>
<td>41.7</td>
<td>9/2</td>
<td>--</td>
<td>5.92</td>
<td>0.327</td>
<td>6.24</td>
<td>6.6</td>
</tr>
<tr>
<td>93.4</td>
<td>11/2</td>
<td>--</td>
<td>0.88</td>
<td>0.267</td>
<td>1.15</td>
<td>0.85</td>
</tr>
<tr>
<td>156</td>
<td>13/2</td>
<td>--</td>
<td>--</td>
<td>0.083</td>
<td>0.083</td>
<td>0.08</td>
</tr>
<tr>
<td>250</td>
<td>15/2</td>
<td>--</td>
<td>--</td>
<td>0.0083</td>
<td>0.0083</td>
<td>0.012</td>
</tr>
</tbody>
</table>
From Eq. VI.9, we find at saturation,

\[ L=0, L=4 \text{ out of phase, } W(\theta) = 1 + 0.910 P_2(\cos \theta) - 0.005 P_4(\cos \theta) \]

\[ L=0, L=4 \text{ in phase, } W(\theta) = 1 + 1.07 P_2(\cos \theta) + 0.110 P_4(\cos \theta) \]

**Experimental Procedure**

Twenty-day \( \text{E}^{253} \) was produced by prolonged neutron irradiation, starting with \( \text{Cm}^{244} \). The californium fraction was separated from the other activities by ion-exchange techniques. This fraction contained considerable amounts of \( \text{Cf}^{253} \), which beta decays to \( \text{E}^{253} \) with a twenty day half-life. After a suitable decay period, the \( \text{E}^{253} \) was obtained by ion-exchange separation. It was then passed through several alcohol-HCl clean-up columns, and finally collected mass free on glass planchettes.

This work was done by the Actinide Chemistry group at the Lawrence Radiation Laboratory, under the direction of Dr. B. B. Cunningham. I am indebted to Dr. Cunningham, Dr. J. Hollander, Mrs. Helen Michel and Mr. Martin Holtz for providing the samples used in this experiment.

The \((\text{E}^{253})^{+3}\) as the chloride was taken up in a minute amount of \( \text{H}_2\text{O} \) and deposited on a small spot on the surface of a single neodymium ethylsulfate crystal with a micro pipet. Both naturally and artificially prepared surfaces were used. The procedure was the same as used for the \( \text{Ce}^{137m} \) conversion-electron experiment described in Sec. III.A. This was done until approximately \((1-3) \times 10^5\) alpha counts per minute were obtained in \(2\pi\) geometry with an alpha survey meter. The crystal was hung in a demagnetization cryostat with collimated alpha detectors at \(0^\circ\) and \(90^\circ\) as shown above (Fig. II.12). Upon demagnetization, simultaneous counts were taken with the detectors, while the bulk temperature was monitored with mutual inductance coils. Because the temperature of the spot probably rose faster than the bulk temperature of the crystal, only one count was taken per demagnetization. The counts were normalized by warming the entire crystal to \(1^\circ\text{K}\). No structure was observed in the alpha spectrum, which is shown for \(0^\circ\) in Fig. VI.2. Occasionally, small shifts in the peak position or in the peak width were observed for the warm count.
Fig. VI.2. Alpha particle spectrum from E$^{253}$ in NES taken with a germanium surface-barrier detector at 10K and at 0° to the crystal c axis. Curve A was taken with the nuclei at 0.010K (aligned), curve B with the nuclei at 10K (isotropic).
relative to the cold. If the shifts were very large, the count was discarded. For the other counts, most of the peak was included. During some of the series of demagnetizations a few of the runs showed shifts, while others did not. By including most of the peak, the runs proved to be internally consistent. In all, five different samples were used with six different counters. One series of demagnetizations was used to measure the temperature dependence, while the others were used to measured the saturation anisotropies only. Solid angle corrections were made to the theory using the formulas given by Rose for finite detectors and finite source areas.

The data, corrected for source decay, are given in Table XV. Since there were no higher energy radiations, there were no background corrections. Figure VI.3 shows the anisotropy measured as a function of temperature. Using the saturation values measured for this series of demagnetizations, and taking the saturation magnitudes of $B_2$ and $B_4$, an experimental measure of the size of the $P_2(\cos \theta)$ coefficient was calculated relative to that of the $P_4(\cos \theta)$ coefficient. Using these values, the "theoretical" temperature dependence was calculated, and is shown as the solid line. The temperature variation of the $B_2$ and $B_4$ parameters was calculated assuming a hyperfine structure constant $|A/R| = 0.40^\circ$K adopted by Navarro. The temperature variation is in good agreement with Navarro's results. Figure VI.4 shows $2[1-W(\pi/2)]/[1-W(0)]$ plotted against $[1-W(\pi/2)]$ for the same series of demagnetizations. The solid line was calculated using the same parameters as used in Fig. VI.3. The dashed curve corresponds to a pure $P_2$ distribution. Figure VI.5 shows $[1-W(\pi/2)]-[W(0)-1]$, blocked and plotted against $1/T$. This quantity is equal to $-7/2 B_4(b_4F_4)$. The solid curve is the $B_4$ variation with $1/T$, normalized to the experimental value at $1/T=60$. The dashed curve shows the $B_2$ variation with $1/T$, normalized to the same value.
### Table XV. $E^{253}$ in NES

Alpha particle anisotropies as a function of temperature.

<table>
<thead>
<tr>
<th>Run</th>
<th>$1/T$</th>
<th>$W(0)$</th>
<th>$W(\pi/2)$</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-1</td>
<td>62.5</td>
<td>1.650</td>
<td>0.590</td>
<td>Crystal with sanded face</td>
</tr>
<tr>
<td>1-2</td>
<td>62.5</td>
<td>1.680</td>
<td>0.547</td>
<td></td>
</tr>
<tr>
<td>1-3</td>
<td>62.5</td>
<td>1.734</td>
<td>0.542</td>
<td></td>
</tr>
<tr>
<td>2-1</td>
<td>62.5</td>
<td>1.679</td>
<td>0.574</td>
<td>Crystal with natural face</td>
</tr>
<tr>
<td>2-2</td>
<td>62.5</td>
<td>1.665</td>
<td>0.546</td>
<td></td>
</tr>
<tr>
<td>2-3</td>
<td>62.5</td>
<td>1.662</td>
<td>0.528</td>
<td></td>
</tr>
<tr>
<td>2-4</td>
<td>62.5</td>
<td>1.623</td>
<td>0.523</td>
<td></td>
</tr>
<tr>
<td>2-5</td>
<td>62.5</td>
<td>1.666</td>
<td>0.514</td>
<td></td>
</tr>
<tr>
<td>3-1</td>
<td>62.5</td>
<td>1.524</td>
<td>0.682</td>
<td>Crystal with sanded face</td>
</tr>
<tr>
<td>3-2</td>
<td>62.5</td>
<td>1.520</td>
<td>0.690</td>
<td></td>
</tr>
<tr>
<td>3-3</td>
<td>37.5</td>
<td>1.525</td>
<td>0.657</td>
<td></td>
</tr>
<tr>
<td>3-4</td>
<td>18.3</td>
<td>1.513</td>
<td>0.714</td>
<td></td>
</tr>
<tr>
<td>3-5</td>
<td>12.1</td>
<td>1.504</td>
<td>0.726</td>
<td></td>
</tr>
<tr>
<td>3-6</td>
<td>8.3</td>
<td>1.417</td>
<td>0.778</td>
<td></td>
</tr>
<tr>
<td>3-7</td>
<td>6.0</td>
<td>1.352</td>
<td>0.825</td>
<td></td>
</tr>
<tr>
<td>3-8</td>
<td>4.0</td>
<td>1.315</td>
<td>0.819</td>
<td></td>
</tr>
<tr>
<td>3-9</td>
<td>2.0</td>
<td>1.036</td>
<td>0.989</td>
<td></td>
</tr>
<tr>
<td>3-10</td>
<td>62.5</td>
<td>1.522</td>
<td>0.658</td>
<td></td>
</tr>
<tr>
<td>3-11</td>
<td>8.0</td>
<td>1.432</td>
<td>0.730</td>
<td></td>
</tr>
<tr>
<td>3-12</td>
<td>2.0</td>
<td>1.075</td>
<td>0.940</td>
<td></td>
</tr>
<tr>
<td>3-13</td>
<td>5</td>
<td>1.208</td>
<td>0.854</td>
<td></td>
</tr>
<tr>
<td>3-14</td>
<td>62.5</td>
<td>1.513</td>
<td>0.646</td>
<td></td>
</tr>
<tr>
<td>3-15</td>
<td>6.25</td>
<td>1.526</td>
<td>0.641</td>
<td></td>
</tr>
<tr>
<td>3-16</td>
<td>44</td>
<td>1.544</td>
<td>0.654</td>
<td></td>
</tr>
<tr>
<td>3-17</td>
<td>273</td>
<td>1.509</td>
<td>0.667</td>
<td></td>
</tr>
<tr>
<td>3-18</td>
<td>205</td>
<td>1.522</td>
<td>0.730</td>
<td></td>
</tr>
<tr>
<td>3-19</td>
<td>39</td>
<td>1.523</td>
<td>0.645</td>
<td></td>
</tr>
<tr>
<td>3-20</td>
<td>25</td>
<td>1.509</td>
<td>0.690</td>
<td></td>
</tr>
</tbody>
</table>
Table XV. (Cont).

<table>
<thead>
<tr>
<th>Run</th>
<th>I/T</th>
<th>W(0)</th>
<th>W(\pi/2)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>4-1</td>
<td>62.5</td>
<td>1.540</td>
<td>0.657</td>
<td>Crystal with sanded face</td>
</tr>
<tr>
<td>4-2</td>
<td>28.5</td>
<td>1.544</td>
<td>0.677</td>
<td></td>
</tr>
<tr>
<td>4-3</td>
<td>34.5</td>
<td>1.520</td>
<td>0.678</td>
<td></td>
</tr>
<tr>
<td>4-4</td>
<td>25.0</td>
<td>1.540</td>
<td>0.692</td>
<td></td>
</tr>
<tr>
<td>4-5</td>
<td>15.5</td>
<td>1.511</td>
<td>0.710</td>
<td></td>
</tr>
<tr>
<td>4-6</td>
<td>16.5</td>
<td>1.502</td>
<td>0.702</td>
<td></td>
</tr>
<tr>
<td>4-7</td>
<td>11.8</td>
<td>1.504</td>
<td>0.710</td>
<td></td>
</tr>
<tr>
<td>4-8</td>
<td>15.3</td>
<td>1.516</td>
<td>0.689</td>
<td></td>
</tr>
<tr>
<td>4-9</td>
<td>12.4</td>
<td>1.487</td>
<td>0.708</td>
<td></td>
</tr>
<tr>
<td>4-10</td>
<td>10</td>
<td>1.491</td>
<td>0.729</td>
<td></td>
</tr>
<tr>
<td>4-11</td>
<td>3</td>
<td>1.111</td>
<td>0.943</td>
<td></td>
</tr>
<tr>
<td>5-1</td>
<td>62.5</td>
<td>1.674</td>
<td>0.529</td>
<td>Crystal with natural face</td>
</tr>
<tr>
<td>5-2</td>
<td>62.5</td>
<td>1.564</td>
<td>0.533</td>
<td></td>
</tr>
<tr>
<td>5-3</td>
<td>62.5</td>
<td>1.661</td>
<td>0.534</td>
<td></td>
</tr>
<tr>
<td>5-4</td>
<td>62.5</td>
<td>1.673</td>
<td>0.542</td>
<td></td>
</tr>
<tr>
<td>5-5</td>
<td>62.5</td>
<td>1.693</td>
<td>0.559</td>
<td></td>
</tr>
</tbody>
</table>
Fig. VI.3. \( W(\theta) \) for \( \theta = 0 \) and \( \pi/2 \) for \( E^{253} \) aligned in NES as a function of \( \log(1/T) \). The solid curve is theoretical for \( |A/k| = 0.40^\circ K \).
Fig. VI.4.  $2[1-W(\pi/2)]/[1-W(0)]$ versus $[1-W(\pi/2)]$ for $E^{\gamma \gamma}$ aligned in NES, showing the effect of the large, negative $P_4$ term. The dashed curve is theoretical for a pure $P_2$ term. The solid curve is theoretical and includes the $P_4$ term derived from the data at $1/T=62.5$. 
Fig. VI.5. $2[1-W(\pi/2)-W(0)-1] = -\frac{7}{4} B_4 b_4 F_4$ NÉS, as a function of $\log(1/T)$. The solid curve is the variation of $B_4$ with $\log(1/T)$ normalized to the $1/T=62.5$ point. The dashed curve is the variation of $B_2$. 
6. Discussion

The sign of the \( P_4 \) term is negative, as may be seen in Figs. VI.4 and VI.5. This implies that the \( L=0 \) and \( L=4 \) waves are out of phase. This conclusion is consistent with predictions made by Rasmussen and Mang\(^{107}\) based on calculations of favored decay using Nilsson wave functions. They find that \( L=4 \) is in phase below a minimum intensity region around mass 244 and out of phase above mass 244. Theoretical studies by Rasmussen and Chasman\(^{95}\) indicate that \( L=4 \) is in phase for \( U^{233} \).

The agreement between the experimental magnitudes of the coefficients of the \( P_2 \) and \( P_4 \) terms and the theoretical predictions based on the BFM hypothesis is not satisfactory. The \( P_2 \) term is smaller than predicted, while the \( P_4 \) term is almost an order of magnitude larger. The direction of the effect suggests an enhancement of the \( L=4 \) wave amplitude and a reduction of the \( L=2 \) wave amplitude from the BFM calculations. This conclusion is based on the observation of a smaller anisotropy at \( \theta=0^\circ \) than predicted. There are several experimental effects that could cause a reduction in anisotropy. One possibility is rapid warmup in the source spot. However, in view of the saturation behavior of the anisotropy, this is unlikely. That is, the temperature may change from \( 1/T=60 \) to \( 1/T=25 \) without an appreciable change in anisotropy. Another potential difficulty might be source preparation. If some of the \( E^{+3} \) is not in the lattice of the large crystal, but is in small, randomly oriented crystallites on the surface, the alpha particles from these crystallites would contribute an isotropic background which would reduce the observed anisotropy. If these crystallites are spread uniformly over the source area, the effect at \( 0^\circ \) and the effect at \( 90^\circ \) will be attenuated by the same fraction. Because of the finite source size, there is the possibility that the effective source seen by the \( 0^\circ \) detector is different from that seen by the \( 90^\circ \) detector. In this case, the effect on the anisotropy at \( 0^\circ \) and \( 90^\circ \) would be different, leading perhaps to an anomalously large \( P_4 \) term and an anomalously small \( P_2 \) term. Because of the possible existence of these attenuations, the experiment was performed using five independently prepared sources. All the sources showed an enhancement of the \( P_4 \) term and a reduction of the \( P_2 \) term compared to BFM, and all showed a
negative sign for the $P_4$ term. All the samples did not show the same saturation anisotropy. The sources used in the temperature dependence study showed a saturation anisotropy at 0° which was 10% less than the maximum anisotropy observed with the other sources. The anisotropy at 90° was also attenuated. This would indicate that any attenuation is the same at 0° and 90°. Similar effects were observed by Navarro.\textsuperscript{25,87} From the temperature dependence run, the variation of the $P_4$ term as a function of temperature was shown in Fig. VI.5. This variation is fitted with a curve having the temperature variation of $B_4$ calculated with $(A/K) = 0.4^\circ\text{K}$. Also shown is the $B_2$ variation. The data follow the $B_4$ variation more closely than the $B_2$ variation. This confirms that the sign of the $P_4$ coefficient is indeed negative. Finally, a misalignment of the detectors from 0° and 90° could give an observed anomaly. To avoid this the detectors and collimators were carefully aligned in each experiment and were at 0° and 90° to within 2°. Since $P_2$ and $P_4$ are relatively flat at 0° and 90°, a small misalignment would have a negligible effect.

Taking an average of the saturation runs and correcting for solid angle ($g_2=0.944$, $g_4=0.812$), the following amplitudes for the L waves in the transition to the ground state of $^{249}$Bk were calculated:

\begin{align*}
a_0 &= 0.796 \\
a_2 &= 0.0886 \\
a_4 &= 0.0127
\end{align*}

The relative phase for the L=2 and L=4 waves to the higher states in the rotational band was taken as negative. The L=2, L=4 amplitudes for these transitions were taken as Asaro's values. In Table XVI these amplitudes are compared with the calculations by Asaro from the intensity data and with a calculation by Poggenburg, Mang, and Rasmussen\textsuperscript{108} based on overlap integrals between the various single-particle wave functions and a superconductivity-model calculation of occupation probabilities. This latter calculation also finds L=0,2 in phase and L=0,4 out of phase.
Table XVI. Relative amplitudes and phases for alpha decay to the ground state of Bk\(^{249}\).

<table>
<thead>
<tr>
<th></th>
<th>(a_0)</th>
<th>(a_2)</th>
<th>(a_4)</th>
<th>(\phi_{02})</th>
<th>(\phi_{04})</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Asaro et al.</td>
<td>1</td>
<td>0.125</td>
<td>0.0016</td>
<td>--</td>
<td>--</td>
<td>105</td>
</tr>
<tr>
<td>Navarro</td>
<td>-</td>
<td>--</td>
<td>--</td>
<td>(+)</td>
<td>--</td>
<td>25, 87</td>
</tr>
<tr>
<td>This work</td>
<td>1</td>
<td>0.110</td>
<td>0.016</td>
<td>(+) (--)</td>
<td>(--)</td>
<td></td>
</tr>
<tr>
<td>Poggenburg et al.</td>
<td>1</td>
<td>0.125</td>
<td>0.00053</td>
<td>(+) (--)</td>
<td>(--)</td>
<td>108</td>
</tr>
</tbody>
</table>
ACKNOWLEDGMENTS

I would like to thank:

Dr. David A. Shirley, who directed this research and who introduced me to these techniques and problems.

Dr. Nicholas J. Stone, who collaborated on many of the experiments described here.

Mr. Gardner G. Young and his shop, Mr. Harry Powell and his shop, Mr. Clint Ward, Mr. Steven Levine, Mr. Don Landis, Dr. Fred Goulding, Mrs. Gertrude Boltz, and Mrs. Winnie Heppler, for excellent technical assistance.

Dr. Eckart Matthias and my long-suffering fellow-graduate students, for their encouragement and assistance with the experiments.

The National Science Foundation, for financial support during my first three years of graduate study.

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 

2. H. Frauenfelder, "Angular Correlation," in Beta and Gamma Ray Spec- 
troscopy, ed. by K. Siegbahn (Interscience Publishers, New York, 
1955).

3. C. D. Jeffries, Dynamic Nuclear Orientation (Interscience Publishers, 

(1955).


6. C. E. Johnson, J. F. Schooley, and D. A. Shirley, Phys. Rev. 120, 
2108 (1960).

S69, Sept. 1958.

8. C. E. Johnson, J. F. Schooley, and D. A. Shirley, Phys. Rev. 120, 
1777 (1960).

9. J. N. Blok (Lawrence Radiation Laboratory, Berkeley), private 
communication, 1964.

135 (1951).


(1955).


16. R. M. Steffen and H. Frauenfelder, Perturbed Angular Corrections, 
ed. by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland 


34. H. T. Easterday and A. J. Haverfield (Lawrence Radiation Laboratory, Berkeley), private communication, 1964.
54. From Ref. 21 the extreme limits of $b_2$ for K electrons and pure multipolarity are encountered in the low energy, low Z limit for $E1$ and $E2$ transitions, where the values of $b_2$ are -2$(E1)$ and +2$(E2)$. For mixed-multipolarity transitions the ratios $F_2(e,\text{mixed})/F_2(\gamma,\text{mixed})$ have singularities at those points for which $F_2(\gamma)$ is zero.
60. J. Blok (Lawrence Radiation Laboratory, Berkeley), private communication, February 1964.
70. R. W. Grant and D. A. Shirley, Phys. Rev. 150, 1100 (1965).
75. N. J. Stone, D. A. Shirley, and R. B. Frankel, unpublished data.
84. L. A. Sliv and I. M. Band, Univ. of Illinois (Trans), Report 571CCK1 Physics Department, Urbana, Ill., 1956.
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.