Angular Distributions of Conversion Electrons from Oriented Ce$^{137m}$ Nuclei*

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The angular distributions of K- and of (L+M)- conversion electrons from oriented Ce$^{137m}$ nuclei were observed. Precise simultaneous intensity measurements of conversion electrons and $\gamma$ rays from the same sample provided an accurate, direct determination of the particle parameters $b_2$ associated with the 255-keV isomeric transition in Ce$^{137m}$. The K-electron result, $b_2 = 1.061 (18)$, is in excellent agreement with a theoretical prediction based on a “point nucleus” model. Techniques and parameters in this experiment, the first of its kind, are discussed. A more careful determination of the temperature dependence of anisotropy of the 255-keV $\gamma$ ray yielded the value $A = 0.0147 (7)$ cm$^{-1}$ for the hyperfine structure constant of Ce$^{137m}$ in neodymium ethylsulfate. This is slightly higher than the older value of 0.0129 (12) cm$^{-1}$ found in this laboratory and is to be preferred on the basis of internal evidence. Using Bleaney’s value of $(r^{-2}n_{0})$ = 4.44 au, the derived magnetic moment is changed from 0.96 (09) nm to 0.89 (05) nm. Other radial integrals give different values. A possible discrepancy in the magnetic temperature scale for neodymium ethylsulfate is discussed.

I. INTRODUCTION

THE measurement of $\gamma$-ray angular distributions from nuclei oriented at very low temperatures has become an established technique for the study of problems in both nuclear and solid-state physics. The use of semiconductor particle detectors has also made possible the measurement of alpha-particle anisotropies.†,‡ Perhaps the most striking application of nuclear orientation, the proof of parity nonconservation in beta decay,† has not been developed into a general study of beta decay, largely because of the hitherto qualitative nature of electron energy measurements with plastic scintillators.

Measurement of the energy dependence of charged-particle anisotropies from oriented nuclei would be valuable for many problems in nuclear physics. Special experimental techniques and counters have been developed to make such measurements possible. These are described in Sec. II. As a first experiment the angular distribution of conversion electrons from oriented Ce$^{137m}$ was chosen. This experiment was free from ambiguity of interpretation and had valuable internal checks. At the same time it allowed the first really direct determination of a “particle parameter” for conversion electrons.‡ The results are given in Sec. III. In Sec. IV the measurement of particle parameters is discussed and the results are compared with theory. The hyperfine structure constant for Ce$^{137m}$ and the magnetic temperature scale for neodymium ethylsulfate are discussed in Sec. V. Finally in Sec. VI future applications are given for the techniques described herein.

II. EXPERIMENTAL

A. The Electron Detectors

The electron measurements were made using gold surface-barrier germanium detectors, which Navarro,†,‡ had found to give good resolution for electron counting at 1 K. A wafer of 40 $\Omega$-cm germanium, 8 mm X 8 mm X 0.8 mm was lapped and etched. A back contact was made by evaporation and alloying of gold. The front surface was a thin (about 400 A) gold sheet, also

‡ Recently J. Chin, A. T. Hirshfeld, and D. D. Hoppes [Rev. Sci. Instr. 34, 1288 (1963)] have used silicon counters to detect electrons below 4.2 K.

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Manganous ammonium sulfite pill

![Fig. 1. Cryostat arrangement for conversion-electron anisotropy studies. NaI counters are mounted outside the Dewar system at 0 and 90° to the crystalline c axis.](image)

evaporated. The detector was mounted on a molybdenum strip with a thin conducting layer of gold epoxy resin. A thin gold wire was pressure-bonded to the front surface and this wire was connected with the conducting epoxy to a B.S. 36 manganin wire which ran up the center of the cryostat pumping line to a connector at the top of the apparatus. Negative bias up to 30 V was applied and the pulses due to the conversion electrons striking the detector were fed into a charge-sensitive preamplifier, followed by a linear amplifier and multichannel analyzer.

Two detectors were placed inside the demagnetization cryostat, one at 0 and one at 90° to the crystalline c axis of the neodymium ethylsulfate (NES) crystal containing the Ce$^{137m}$ activity (Sec. II. C). The c axis of this salt is the axis of nuclear alignment. The cryostat arrangement is shown in Fig. 1. Sodium iodide counters were mounted at 0 and 90° outside the cryostat. The conversion-electron spectrum taken at 1.0 K with an electron detector is shown in Fig. 2, with the K and (L+M) peaks clearly resolved.

![Fig. 2. Conversion electron spectrum of the 255-keV transition in Ce$^{137m}$ taken with a germanium surface barrier detector at 1°K.](image)

**B. Choice of the Experiment**

In order to establish the quantitative accuracy of measurements made with the germanium electron detectors it was advantageous to study a case in which the electron energy spectrum was discrete, rather than continuous, and for which a quantitative theoretical prediction existed. The case selected satisfies these requirements unusually well. When aligned in NES, for which the temperature-susceptibility correlation is known, Ce$^{137m}$ had been shown to give large γ-ray anisotropies, and the 255-keV isomeric transition is highly converted ($\gamma_1/\gamma = 6$). The existing decay scheme is shown in Fig. 3. A side effect of this investigation has been the discovery of many more transitions, as well as several errors, in this decay scheme. The decay is under intensive study, and the results will be reported separately soon. We note in passing that the changes will have no effect on the work reported here.

The theoretical expression for the γ-ray angular distribution from an assembly of oriented nuclei has the form

$$W(\theta) = 1 + \sum_{\nu \text{ even}} A_\nu P_\nu(\cos \theta),$$

(1)

where $\theta$ is the angle from the quantization axis and $P_\nu$ is a Legendre polynomial of order $\nu$. The $A_\nu$ are discussed in detail in Sec. IV. For conversion electrons the expression is modified by the inclusion of “particle parameters” $b_\nu$ multiplying the $A_\nu$, which account for changes in the angular distribution brought about by consideration of the initial and final electron states.

7 Horst Meyer, Phil. Mag. 2, 521 (1957).
11 A preliminary account of this work is given by N. J. Stone, R. B. Frankel, and D. A. Shirley, Bull. Am. Phys. Soc. 9, 486 (1964).
Theoretical values of the particle parameters have been given by Biedenharn and Rose. For a 255-keV M4 transition in cerium (element 58), their theoretical $b_2$ is 1.055.

C. Source Preparation

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. The accompanying Ce$^{140}$ activity has no interfering radiations. A Ce-La chemical separation was made by solvent extraction of Ce$^{4+}$, following Glendenin, with additional back extraction, followed by an HCl cleanup column, and the resulting triplicate carrier-free Ce$^{137m}$ activity was used.

For electron measurements a source with minimum scattering and absorption is required. A large NES crystal was selected and was sanded to produce a face at 45° to the $c$ axis. The Ce$^{137m}$ activity was taken up in a small drop of water and was deposited on an area of 1 mm$^2$ on this face. The water drop was removed and replaced several times without being allowed to evaporate. In this way the Ce$^{3+}$ replaced some Nd$^{3+}$ in the lattice. The crystal was then mounted in the cryostat as shown in Fig. 1 with the active area on the line of intersection of the detector axes. 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.

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. 4. After small corrections were made for source decay and background, the normalized intensities, $W(O)$, 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 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.

III. MEASUREMENTS AND RESULTS

Upon demagnetization of the NES crystal from magnetic fields up to 23 kG 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 h. (We note that small temperature

\[ L. E. Glendenin, Anal. Chem. 27, 50 (1955). \]
In Figs. 5 and 6 $[1-W(0)]$, for each electron peak, is plotted against $[1-W(0)]$. Figures 7 and 8 show the ratios $[1-W(0)]/[1-W(0)]$ and $[1-W(0)]/[1-W(0)]$ as functions of $[1-W(0)]$. We note that the determination of the ratios is in no way dependent on our knowledge of the temperature scale for NES. It is necessary for our interpretation however, that there should be no gross temperature inhomogeneities over the 1-mm$^2$ active area. The long “warm-up” times of hours and large $\gamma$-ray anisotropies are excellent evidence that this criterion is satisfied.

IV. DISCUSSION

A general expression for the observed angular distribution of conversion electrons following the decay of oriented nuclei is

$$W(\theta) = \sum_{\text{even}} B_s U_s g_{s} b_s P_{s} \cos(\theta). \quad (2)$$

Here $B_s$ are orientation functions, $P_s$ are Legendre polynomials, and $F_s$ are the angular correlation coefficients. The $g_{s}$ are attenuation coefficients which account for the solid angle subtended by the counter. The composite parameters $Q_s$ and $U_s$ describe, respectively, the reorientation in intermediate states and the reorientation due to unobserved transitions between the original state in which the nuclei are oriented and the state from which the observed radiations are emitted. For several successive unobserved transitions the parameters are just the products of the corresponding functions for the transitions taken singly. Thus if $n$ successive unobserved transitions precede the state from which emitted radiation is observed, we may write

$$U_s = \prod_{i=1}^{n} U_{s,i}^{(0)}, \quad (3a)$$

$$Q_s = \prod_{i=1}^{n} Q_{s,i}^{(0)}. \quad (3b)$$

The $U_s^{(0)}$ are simply geometrical functions of the intermediate spins and multipolarities. They arise from angular-momentum conservation and are found in the theory of angular correlation in multiple cascades and in the theory of the angular distribution of $\gamma$ rays from oriented nuclei. The $U_s^{(0)}$ may be calculated exactly.\(^{12}\)

The $Q_s^{(0)}$ (attenuation) factors arise from interactions of intermediate nuclear states with extranuclear fields. They are less well understood. For intermediate-state lifetimes of $\sim 1$ nsec or longer it is not possible in practice to predict the value of $Q_s^{(0)}$ with any confidence, though a theory is available, because of their sensitivity to details of source preparation.\(^{15}\) It is generally accepted that intermediate-state perturbation is negligible ($Q_s^{(0)}=1$) for lifetimes much shorter than 1 nsec, although some evidence to the contrary exists.\(^{16}\) Certainly substantial attenuation has been observed for lifetimes of the order of a few nsec.\(^{15,17,18}\)

The $b_s$ are “particle parameters” which modify $F_s$ for the observed transition. For $\gamma$ rays, and for all radiations (of pure or mixed multipolarity) in the high-energy limit, $b_s=1$.\(^{19}\) The particle parameters for

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conversion electrons may be substantially different from unity, especially for low energies and/or multipoles. In contrast to γ rays, the electrons are spin-½ 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. Church et al. have pointed out a sign error in the theory.

Some $b_s$ are sensitive to certain subtle features of nuclear structure. Of particular importance is the "penetration matrix element" for hindered $M1$ transitions. Thus it is desirable to check the theory of particle parameters by measuring several $b_s$ with high accuracy. That the theory has not been very critically tested is evident from the fact that, until the recent work of Geiger, all the theoretical mixed-transition particle parameters in the literature had the wrong signs.

For pure multipoles several particle-parameter measurements of 5–10% accuracy have been reported. They are set out in Table I. 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, in which only one intermediate state, are always subject to whatever uncertainty the perturbation factors $Q_e$ 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.

A serious difficulty encountered in a precise experimental test of the particle parameter theory is the extreme sensitivity of both $F_2$ and $b_s$ 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$^{37}$ isomeric transition should be very pure $M4$. The lifetime is only a factor of 3 below the single-particle estimates, 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 $E5/M4$ intensity mixing ratio would be only $2 \times 10^{-4}$ and would change the theoretical $b_4$ by less than the experimental error.

There were no intermediate states in this experiment, so the $U$, $Q_e$ factors in Eq. (2) can be replaced by unity. The $B_s$ are identical for conversion electrons and γ-ray emission. The $g_s$ corrections are accurately known, and $P_s(1) = 1$ for all $v$. The $B_s$ are all interrelated through the spin Hamiltonian for Ce$^{3+}$ in NBS (Sec. V), and all $B_s$ for $s \geq 6$ are negligible (in fact $B_s = +0.0016$ at $T = 0.02^\circ$). The relationship between $B_2$ and $B_4$ was accurately known from independent experiments on the γ-ray angular distributions at $\theta = 0$ and $\theta = 4\pi$ (Sec. V). Therefore we know $B_2$ and $B_4$ for any $W(0)_\gamma$, and Eq. (2) may be simplified to

$$W(0)_\gamma = 1 + B_{2g}(\gamma)B_2 + B_{4g}(\gamma)B_4,$$

For this experiment,

$$F_2 = -0.889, ~ F_4 = +0.443, ~ g_s(e) = 0.935 \pm 0.015, ~ g_s(\gamma) = 0.903 \pm 0.005,$$

and

$$g_s(\gamma) = 0.700 \pm 0.010.$$  

Thus,

$$1 - W(0) = \frac{0.831(13)B_2b_2 + 0.354(18)B_4b_4}{1 - W(0)} = \frac{0.802(4)B_2 + 0.310(4)B_4}{1 - W(0)},$$

where $b_2$ and $b_4$ are related [see Eq. (98) of Ref. 4] for this case by

$$b_4 = \frac{(17/3)b_2 - 14/3}{1}. \tag{6}$$

Substituting Eq. (6) into Eq. (5) we obtain, for any specific value of $1 - W(0)_\gamma$, an equation with only one unknown, $b_2$. We have calculated $[1 - W(0)_\gamma]/[1 - W(0)_\gamma]$ as a function of $1 - W(0)_\gamma$ for several values of $b_2$. These curves are compared with the data in Figs. 7 and 8. From all the $0^\circ$ data we obtain the

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**Table I. Conversion-electron particle parameters for transitions of "pure" multipolarity.**

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>$F_2$ (keV)</th>
<th>Multi. polarity $b_s$ (theo)</th>
<th>$b_s$ (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>
</tr>
<tr>
<td>Ta$^{181}$</td>
<td>133</td>
<td>$E2$</td>
<td>1.84</td>
<td>1.8±0.2</td>
</tr>
<tr>
<td>Ce$^{137}$</td>
<td>255</td>
<td>$M4$</td>
<td>1.055</td>
<td>1.06±0.018</td>
</tr>
</tbody>
</table>

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* See Ref. 4.

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**Notes:**

1. From Ref. 4 the extreme limits of $b_s$ for $K$ electrons and pure multipolarity are encountered in the low energy, low-$Z$ limit for $E1$ and $E2$ transitions, where the values of $b_s$ are $-2(E1)$ and $+2(E2)$. For mixed-multipolarity transitions the ratios $F_2(e, mixed)/F_2(e, pure)$ have singularities at those points for which $F_2(e)$ is zero.


values (standard deviations) $b_2(K) = 1.061(6)$, $b_2(L+M) = 1.059(8)$. The largest known systematic errors arise from uncertainties in the background and solid angle corrections. 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 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.

We conclude that the theory is accurate to about 2% for this case. Of course this single result does not establish the theory for cases in which the $b_v$ 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.

V. THE NES TEMPERATURE SCALE AND THE MAGNETIC MOMENT OF Ce$^{137m}$

The particle parameter determination is independent of temperature measurement, being a direct comparison of simultaneously measured electron and $\gamma$-ray anisotropies. Although in the analysis the relative sizes of the $P_2$ and $P_4$ terms in the angular distribution must be known, this ratio is determined by the form of the spin Hamiltonian and does not depend upon temperature measurements.

However, as an auxiliary experiment the variation of the 255-keV $\gamma$-ray anisotropy with temperature was remeasured in order to re-examine the temperature dependence found by the previous study. At the time this earlier work was done the systematic discrepancy between theory and experiment was noted and was attributed to a possible error in the temperature scale for NES. When the present work was undertaken a completely different apparatus was available and it semed worthwhile to repeat the work with a stronger source. A second NES crystal was grown, homogeneous in Ce$^{137m}$ activity to avoid uneven local heating, and the $\gamma$-ray anisotropy measured as a function of the magnetic temperature $T^*$ of the crystal. This was corrected to absolute temperature using the $T-T^*$ correlation of Meyer and making a demagnetization correction for crystal shape. The results (corrected for background and source decay) are shown in Fig. 9. Figure 10 is a plot of $W(0)$ against $W(\frac{3}{2}\pi)$; this plot is sensitive to the relative magnitudes and signs of the coefficients of the $P_2$ and $P_4$ terms. The solid curve is the theoretical relationship for pure $M4$ radiation, using the effective spin Hamiltonian adopted by Haag et al.,

$$3c = AS_xS_z + B(S_xS_y + S_yS_z),$$

with

$$A = 0.074\mu_N/I \text{ cm}^{-1} \quad \text{and} \quad B = 0.002\mu_N/I \text{ cm}^{-1},$$

and taking $I = \frac{11}{2}$. In this plot the size of $\mu_N$ need not be assumed as it does not affect the form of the theoretical curve, but only the temperature to which a given point on the line corresponds. The excellent agreement found between theory and experiment (the theoretical curve is corrected for finite counter aperture) is evidence that the radiation is indeed pure $M4$, and an upper limit of $\delta = \pm 0.10$ can be placed upon the ES amplitude admixture in the transition.

In Fig. 11 is plotted the anisotropy $\epsilon = 1 - W(0)/W(\frac{3}{2}\pi)$ as a function of $1/T$. 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 $A = 0.0129(12) \text{ cm}^{-1}$. The theoretical curve shown in Fig. 11 is for $A = 0.0147 \text{ cm}^{-1}$. The detailed agreement of theory with experiment is still poor. The theoretical curve shows definite discrepancies, being too high in the region $10 \leq 1/T \leq 25$.
TABLE II. Magnetic moment for Ce$^{137m}$ implied by various values of $(r^{-4})_{M}$ for Ce$^{2+}$.

<table>
<thead>
<tr>
<th>$(r^{-4})$, au</th>
<th>$\mu_{M_{\text{mom}}, \text{nm}}$</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>

and too low for $40 \leq 1/T \leq 50$. Similar deviations were found previously.

In general, the form of the $\epsilon$ versus $1/T$ plot depends upon the $F_{2}$, 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°C in the region of 0.05°C, and too high by about 0.001°C at 0.02°C. Further evidence for such errors has been found in studies of electric quadrupole hyperfine coupling in rare-earth ions.

However, taking the theoretical curve shown as the best fit to our data we may derive a new value of $A = 0.0147(7) \text{ cm}^{-1}$ for the hfs constant of Ce$^{137m}$ in NES. This error is statistical and no contribution from the temperature scale errors is included. We note that this new value of $A$ is to be preferred over the earlier result on the internal evidence of a larger anisotropy at each magnetic temperature.

To derive a nuclear moment from this hfs constant a value of $(r^{-4})_{M}$ is required. Recently the value of 3.64 au$^{27}$ for Ce$^{2+}$, which was used to derive Eq. (8), has been seriously challenged by Bleaney$^{28}$ and by Freeman and Watson. In Table II we list the various proposed values of $(r^{-4})$ and the magnetic moment values which each would imply.

VI. APPLICATIONS

The techniques reported here would be applicable for measuring particle parameters in any nuclear orientation experiments in which intensities permit and for which sufficiently thin samples can be prepared. In these cases this type of measurement can be especially valuable for the following reasons:

(1) The $b_{\epsilon}$ are parity sensitive. For example, except for very high-energy ($>2$ MeV) transitions in the heaviest elements, the $E1$ and $M1$ $b_{\epsilon}$'s are always quite different. Thus an internal parity determination, which is absent in directional correlation of $\gamma$ rays alone, is provided.

(2) The ratio $F_{2}(\epsilon)/F_{2}(\gamma)$ for mixed transitions is in some multipolarity regions extremely sensitive to the multipolarity mixing ratio and can give this ratio with higher accuracy than any other method.

(3) For transitions of a given multipolarity the $b_{\epsilon}$ are independent of the initial- and final-state spins. A familiar dilemma in both angular correlation and nuclear orientation is the situation in which there are more unknowns (spins and multipolarities) than equations (experimental $F$). Each measured $b_{\epsilon}$ provides an addition equation.

The original motivation for this experiment was the development of a precise, energy-sensitive technique for studying beta-decay matrix elements. This field has not grown very rapidly, largely because of the extreme difficulty of the available experimental methods. Nuclear orientation experiments have the difficulties accompanying thick source backings, but they employ singles rather than coincidence counting, thus avoiding the intermediate state with its attendant uncertainties. The multiplicity of matrix elements in most beta decay problems will necessitate full use of all available measurements. We hope that the techniques reported here will prove useful for such problems.

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26 J. Blok, Lawrence Radiation Laboratory, Berkeley, California (private communication).
28 B. Bleaney (private communication, 1963).