angular momentum and parity are operating, this 1^- state might also be produced by an $l_n=0$ neutron, of which there is no sign in the experimental distribution. If one tries to match the $l_n=1$ curve to the experimental distribution by requiring the maxima to coincide, than an $R<2.5\times10^{-13}$ cm must be chosen, a value which seems unreasonably small. However, an $R=10.5\times10^{-13}$ cm is sufficient to cause a coincidence of the $l_n=3$ maximum with that of experiment. Although this is considerably larger than the value 6.7×10^{-13} cm, which is based on other experience with fitting the Bhatia formula to experiment, one is perhaps a little hesitant in excluding this possibility in view of the large radius parameters necessary to improve the fits for the first and third excited state distributions.

ACKNOWLEDGMENTS

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Nuclear Alignment of Co^{58}†

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By using a two-stage adiabatic demagnetization technique, Co^{58} nuclei were aligned in single crystals of (22.7% Cu 77.3% Zn) $K_2(SO_4)_2\cdot6H_2O$ containing a trace of cobalt. Both the 0.805-Mev and 1.6-Mev gamma rays were measured at three angles as a function of the paramagnetic susceptibility. The 1.6-Mev gamma ray was determined to be quadrupole. A parameter λ related to the mixture of Fermi and Gamow-Teller interactions in the allowed beta decay was determined to be 0.11 ± 0.04 in the transition to the 0.805-Mev level in Fe^{58} and 0.45 ± 0.11 in the transition to the 1.6-Mev level. By using the 0.805-Mev counting rate as a thermometer, the paramagnetic susceptibility was determined as a function of the absolute temperature and fitted to the formula $\chi(T)=(c/T)F(T)_{\text{hfs}}(1+\Delta/T+\epsilon/T^2)$. The data indicate an antiferromagnetic interaction between copper ions, in disagreement with previous work.

I. INTRODUCTION

In the last few years the method of nuclear orientation has been applied extensively to the study of the nuclear properties of the cobalt isotopes. Following a suggestion by Bleaney,[1] the first successful nuclear alignment experiments were carried out on Co^{60} by Daniels *et al.*[2] and by Gorter *et al.*[3] In these experiments large anisotropies were observed in the gamma radiation emitted by the aligned nuclei, and it was possible to determine the magnetic moment of Co^{60}. Somewhat later Co^{58} was aligned by Daniels *et al.*[4] In Daniels' experiment it was possible to determine approximately the character, i.e., the mixture of Fermi and Gamow-Teller interactions, of the allowed beta transition preceding the gamma transition as well as to determine the magnetic moment of Co^{58}. Subsequently these methods have been used for the study of other cobalt isotopes.[5]

The present experiment will determine certain parameters connected with the beta transitions leading to the first and second excited states of Fe^{58}, and will determine the multipolarity of the gamma transition from the second excited state to the ground state of Fe^{58}. In addition to investigating these properties of the decay of Co^{58}, this experiment is concerned with the possibility of using the angular distribution of the gamma radiation emitted by oriented nuclei as a thermometer.

The decay scheme of Co^{58} has been investigated by Deutsch *et al.*,[6] Strauch,[7] Cork *et al.*,[8] and Frauenfelder *et al.*[9] The decay scheme according to the latter authors is given in Fig. 1. Gamma-ray γ_2 nearly coincides in energy with γ_1 and consequently is counted along with γ_1. However, γ_2 is so weak compared with γ_1 that it has a negligible effect on the results. It will not be

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1 B. Bleaney, Proc. Phys. Soc. (London) **A64**, 315 (1951).
2 Daniels, Grace, and Robinson, Nature **168**, 780 (1951).
3 Gorter, Poppema, Steenland, and Beun, Physica **17**, 1050 (1951).
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6 M. Deutsch and L. G. Elliott, Phys. Rev. **65**, 211 (1944).
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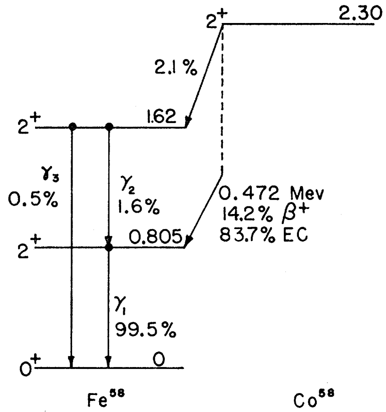

FIG. 1. Decay scheme of Co^{58}.

discussed further. Since the spin assignments of the various nuclear levels and the multipolarities of the gamma rays in the decay of Co^{58} seem to be well established, Co^{58} offers a case where the relative contributions of the Fermi and Gamow-Teller interactions to the allowed beta decay can be studied more or less unambiguously with nuclear alignment techniques.

According to the decay scheme proposed by Frauenfelder and his co-workers for Co^{58}, both γ_1 and γ_3 are in $2-\overset{\beta}{\rightarrow}2-\overset{\gamma}{\rightarrow}0$ cascades. Therefore, according to the theory of Tolhoek and Cox,[10] the angular distribution of these two gamma rays as observed from oriented nuclei should be identical if the mixture of Fermi and Gamow-Teller beta interactions is the same in the two cases. Conversely, any differences in the angular distributions must be ascribed to differences in the two beta interactions. The effect of the beta interaction on the anisotropy of the gamma radiation emitted from oriented nuclei will be considered in detail in the section on the theory.

The theory of nuclear orientation has been worked out by Tolhoek and Cox.[10] For the case of quadrupole radiation, the angular distribution can be expressed as

$$W=1+A_2f_2(\beta,B/A)P_2(\cos\theta)+A_4f_4(\beta,B/A)P_4(\cos\theta), \quad (1)$$

where θ is the angle between the direction of the gamma ray and the alignment axis; A_2 and A_4 are constants which depend on the spins of the states involved in the decay, the difference in spin between these states, and the angular momentum carried away in each transition; P_2 and P_4 are the Legendre polynomials; and f_2 and f_4 are the orientation parameters which describe the degree of alignment. The parameter β compares the separation between nuclear magnetic substates with kT and is defined by

$$\beta=A/2kT. \quad (2)$$

A and B are constants in the spin Hamiltonian, Eq. (4).

[10] H. A. Tolhoek and J. A. M. Cox, Physica **19**, 101 (1953).

The orientation parameters are defined[10] in general as

$$f_2=j^{-2}[(\sum m^2a_m)-\tfrac{1}{3}j(j+1)],$$
$$f_4=j^{-4}[(\sum m^4a_m)-(1/7)(6j^2+6j-5)(\sum m^2a_m)+(3/35)j(j-1)(j+1)(j+2)], \quad (3)$$

where j is the spin of the parent nucleus, m is the z component of the spin, and a_m is the probability that the z component of nuclear spin is m. The resulting angular distribution obtained in a nuclear orientation experiment depends not only on the details of the decay scheme of the radioactive nucleus as described in the theory of Tolhoek and Cox, but also, via the a_m, on the details of the energy levels in the crystal and the types of interactions present between the paramagnetic ion and its environment. A detailed theory of the energy levels of paramagnetic ions has been developed by Abragam and Pryce.[11] They point out that the low-lying levels of paramagnetic ions can be described by a "spin Hamiltonian." For the case of effective spin $S=\frac{1}{2}$, this Hamiltonian may be written

$$\mathcal{H}=g_{\parallel}\beta H_zS_z+g_{\perp}\beta(H_xS_x+H_yS_y)+AS_zI_z+B(S_xI_x+S_yI_y)+P[I_z^2-\tfrac{1}{3}I(I+1)], \quad (4)$$

where the g's are proportional to the magnetic moment of the ion; A and B are proportional to the nuclear magnetic moment of the paramagnetic ion and to the components of the internal magnetic field parallel and perpendicular, respectively, to the z axis; and where P is proportional to the nuclear electric quadrupole moment and to the electric field gradients present at the site of the nucleus.

In order to determine the angular distribution for a specific case, one must determine the orientation parameters, the f_k, as functions of β. This means that one must diagonalize the appropriate spin Hamiltonian and determine the eigenfunctions and eigenvalues. This calculation normally yields the f_k as complicated transcendental functions of β which have to be evaluated numerically.

The z axis for the nuclei in the case of the Tutton salts is either one of two tetragonal axes in the crystal. The crystal structure of the monoclinic Tutton salts has been discussed in relation to nuclear alignment by Bleaney *et al.*[12] In these salts the paramagnetic ion is surrounded by an octahedron of water molecules. The octahedron is stretched along one diagonal which defines a tetragonal axis. In these crystals there are two ions and two tetragonal axes per unit cell. In Fig. 2 are shown the two mutually perpendicular planes containing the axes of the crystal. $\mathbf{K}_1$, $\mathbf{K}_2$, and $\mathbf{K}_3$ are the principal susceptibility axes; **a**, **b**, and **c** are the crystallographic axes; and $\mathbf{T}_1$ and $\mathbf{T}_2$ are the tetragonal axes. In an alignment experiment the crystals are oriented

[11] A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1951).

[12] Bleaney, Daniels, Grace, Halban, Kurti, Robinson, and Simon, Proc. Roy. Soc. (London) **A221**, 170 (1954).

with the b axes vertical. The detectors of the radiation emitted by the oriented nuclei are then moved in the ac plane. It is clear that in this plane the axis of the counter will always make equal angles with the two tetragonal axes, but that the closest approach of the counting axis to the alignment axes is the angle α.

In an alignment experiment the angular distribution of the gamma radiation emitted by radioactive nuclei is measured as a function of the magnetic susceptibility of the crystals. The susceptibility of most paramagnetic crystals follows Curie's law over a wide range of temperature and this enables one to define a magnetic temperature in terms of the paramagnetic susceptibility. The magnetic and absolute temperatures are the same as long as the susceptibility follows Curie's law, but large deviations from Curie's law occur in the alignment temperature range.

Deviations of the susceptibility from Curie's law arise when the ionic moment is no longer free to turn in the measuring field and when the shape of the paramagnetic sample is not spherical. The interactions responsible for constraining the freedom of orientation of the ionic moments are the coupling of the nuclear spin to the ionic moment via the hyperfine interaction, and the magnetic dipole-dipole and exchange interactions between the paramagnetic ions in the crystal. The deviations due to the effects of the anisotropic hyperfine interaction can be calculated from the spin Hamiltonian, and those due to the magnetic dipole-dipole interactions can be calculated from the known g values and the crystal structure. However, the deviations due to the exchange interaction can only be estimated from the specific heat data, so it is not possible to determine completely the theoretical relation between the magnetic and absolute temperature scales accurately. This problem of measuring the absolute temperature is considered in detail by DeKlerk and Steenland,[13] and the methods that have been used heretofore are discussed. Unfortunately it has not been possible to obtain unequivocal results in measurements of the absolute temperature to date. Now that the magnetic moments and interactions of several of the cobalt isotopes are well known through the work of the paramagnetic resonance groups at Berkeley[14] and Oxford,[15] it should be possible to determine the absolute temperature scale from the radioactive counting data and thereby establish the absolute temperature dependence of the paramagnetic susceptibility.

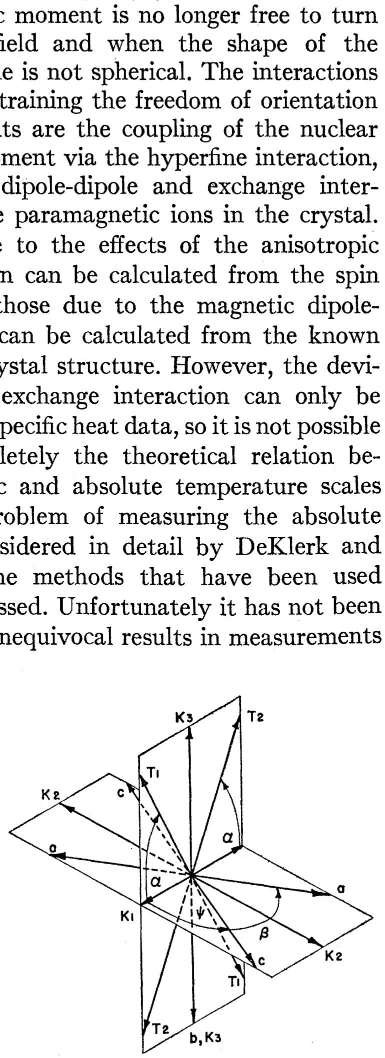

FIG. 2. Diagram of crystal axes in the Tutton salts.

[13] D. DeKlerk and M. J. Steenland, *Progress in Low Temperature Physics* (North Holland Publishing Company, Amsterdam, 1955), Vol. I, p. 273.

II. THEORY

(a) Angular Distribution

According to Cox and Tolhoek,[16] the angular distribution of either γ_1 or γ_3 in Fig. 1 is given by

$$W(\theta)=1-(15/14)(1+\lambda)N_2f_2P_2(\cos\theta) \\ -(5/3)(-2+5\lambda)N_4f_4P_4(\cos\theta), \quad (5)$$

where λ is a parameter which describes the beta interaction in the decay of Co^{58}. In general, λ is defined[10] as

$$\lambda=\left[1+K\left|\int\sigma\right|^2\Big/\left|\int 1\right|^2\right]^{-1}, \quad (6)$$

where $K=(C_T^2+C_A^2)/(C_S^2+C_V^2)$ is the ratio of constants which appear in the interaction Hamiltonian in the theory of allowed beta decay. C_S, C_V, C_T, and C_A are, respectively, the fractions of the total Hamiltonian due to scalar, vector, tensor, and axial vector interactions. $\int\sigma$ and $\int 1$ are, respectively, the nuclear matrix elements for the Gamow-Teller and Fermi interactions. Empirical values for K have been obtained[17] in cases where $|\int\sigma|^2/|\int 1|^2$ could be estimated. K seemed to be about unity. When K is unity, λ is just the fraction of the total nuclear matrix element due to Fermi interaction. In any case, if $\int\sigma$ is zero, λ is unity, while if $\int 1$ is zero, λ is zero.

Since for the Co^{++} ion, $P=0$, the spin Hamiltonian for Co-K_2 Tutton salt in the absence of an external magnetic field is

$$\mathcal{H}=AS_zI_z+B(S_xI_x+S_yI_y), \quad (7)$$

where $S=\frac{1}{2}$, $I=2$, and $A/B\sim 4$.[18] This Hamiltonian was diagonalized both for the case of $B=0$ and for the case $A/B=4$, and the corresponding energy levels and wave functions determined. In both cases each energy level is doubly degenerate, but there is considerable mixing of the magnetic substates and shifting of energy levels when B is not zero. The temperature dependence

[14] Dobrowolski, Jones, and Jeffries, Phys. Rev. **101**, 1001 (1956); W. Dobrowolski and C. D. Jeffries (to be published).

[15] Bleaney, Bowers, and Ingram, Proc. Roy. Soc. (London) **A228**, 147 (1955).

[16] J. A. M. Cox and H. A. Tolhoek, Physica **19**, 673 (1953).

[17] A. Winther and O. Kofoed-Hansen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 14 (1953); C. S. Wu, *Beta- and Gamma-Ray Spectroscopy* (North Holland Publishing Company, Amsterdam, 1955), p. 314.

[18] B. Bleaney and D. J. E. Ingram, Proc. Phys. Soc. (London) **A65**, 953 (1952).

of the angular distribution derived from the $A/B=4$ case is quite different from that for $B=0$.

(b) Magnetic Susceptibility (hfs)

The anisotropic hyperfine structure (hfs) interaction of the ionic magnetic moment with its own nucleus is partly responsible for the deviation of the paramagnetic susceptibility from Curie's law in the temperature region below 1°K. In the crystals used in this experiment, $(22.7\%\text{Cu}77.3\%\text{Zn})K_2(SO_4)_2\cdot 6H_2O$, the concentration of cobalt ions was measured analytically and found to be less than 10^{-4} of the concentration of the copper ions. The magnetic properties then are due to the copper ions alone. The spin Hamiltonian for the Cu-K_2 Tutton salt is given by Eq. (4) with $S=\frac{1}{2}$ and $I=\frac{3}{2}$. The constants used were those given by Bowers and Owen[19] for this salt. This Hamiltonian was diagonalized just as in the case of the Co-K_2 Tutton salt discussed above and the energy eigenvalues and eigenfunctions determined. From these the single ion partition function, Z,

$$Z=\sum_i \exp(-E_i/kT), \tag{8}$$

was constructed.[20] The susceptibility tensor is completely determined by this partition function. Because of the (assumed) axial symmetry, two of the principal values of the susceptibility are equal so that the values parallel and perpendicular to the z axis completely determine the susceptibility tensor.

To calculate the principal values of the susceptibility, one evaluates the quantity

$$\chi=\lim_{H=0}\frac{kT}{H}\frac{\partial}{\partial H}\ln Z, \tag{9}$$

where H is taken successively parallel and perpendicular to the z axis. The susceptibility components are evaluated with respect to the tetragonal axes, whereas the value along the b-crystallographic axis is desired. However, the susceptibility transforms like a tensor, and since the angle between the tetragonal axes and the b axis is $(90°-\alpha)$, the value along the b axis is given by

$$\chi_b=\chi_{\parallel}\sin^2\alpha+\chi_{\perp}\cos^2\alpha. \tag{10}$$

This function was evaluated numerically, and it followed Curie's law very well down to temperatures of about 0.1°K. At 0.01°K, the deviation from Curie's law amounts to 12.5%.

(c) Magnetic Susceptibility (Interactions)

Daniels[21] has worked out a refined treatment of the effect of the magnetic dipole-dipole (d−d) and exchange (ex) interactions on the paramagnetic susceptibility. The total susceptibility may be written

$$\chi(T)=\chi_0(T)(1+\Delta/T+\epsilon/T^2+\cdots)F(T)_{\text{hfs}}, \tag{11}$$

where $\chi_0(T)$ is the Curie law part of the susceptibility, $(1+\Delta/T+\epsilon/T^2)$ is the modification of the Curie law due to the presence of the interactions, and $F(T)_{\text{hfs}}$ is the modification due to the hfs interaction. The latter has already been discussed and was calculated exactly. This separation of the hfs and interaction contributions to the susceptibility is not strictly valid, but is accurate through terms in $(1/T)^3$.[21] In calculating the effect of the (d−d) and (ex) interactions from Daniel's theory, the exchange energy was estimated from the specific heat data of Benzie *et al.*[22]; the (d−d) interaction was assumed to be isotropic; and the lattice sums were carried out within a sphere of radius $b\sqrt{10}$, where b is the largest dimension of the unit cell. The numerical results of the susceptibility calculations, assuming an antiferromagnetic exchange interaction, are $\Delta=-f(0.039\text{ K}°)$ and $\epsilon=f^2(8.6\times 10^{-4}\text{ K}°^2)$. The quantity f is the fraction of the divalent ions which are copper (0.227 in this experiment). The value of g was taken to be that along the b crystallographic axis. The (ex) term contributes 92% and the (d−d) term 8% to Δ. In the numerical value for ϵ, 9.34 is due to (ex) alone, −0.735 is due to (d−d) alone, and −0.021 is due to a mixture of (ex) and (d−d). From this it is clear that in the crystals used in this experiment the (d−d) contribution to the susceptibility was less than 10% of that due to the (ex). Furthermore the correction introduced by these interactions is large compared with the correction due to the hfs interaction. At 0.01°K, the correction to the susceptibility due to the interactions is 45% while that due to hfs interaction is only 12.5%.

In addition to the contribution to the susceptibility of ions within a sphere with center at the ion in question, the contribution of the ions outside the sphere must be considered. This leads to a correction which takes into account the geometrical shape of the sample. This correction is added to Δ in Eq. (11) and may be written

$$\Delta_s=(4\pi/3-\text{D.F.})c, \tag{12}$$

where D.F. is the "demagnetizing factor" ($4\pi/3$ for a spherical sample) and $c\simeq 10^{-3}$ K° is the Curie constant. A demagnetizing factor is not defined for nonellipsoidal shapes, so that a correction by means of Eq. (12) may not strictly be applied. However, on the basis of the general shape of the group of crystals used in the present work, this correction is estimated to be positive and at most 0.002 K°. Thus it is less than 25% of the other corrections already discussed.

[19] K. D. Bowers and J. Owen, Repts. Progr. in Phys. **18**, 304 (1955).
[20] B. Bleaney, Phil. Mag. **42**, 441 (1951).
[21] J. M. Daniels, Proc. Phys. Soc. (London) **A65**, 673 (1953).
[22] Benzie, Cooke, and Whitley, Proc. Roy. Soc. (London) **A232**, 277 (1952).

III. EXPERIMENTAL METHOD

(a) Production of Co^{58}

Radioactive Co^{58} was produced by an (α,n) reaction in Mn^{55} using the 15-Mev internal beam of the University of Illinois cyclotron. The cobalt was chemically separated (carrier-free) from the manganese target so that only cobalt activities were present. The energy of the beam was well below the threshold for the $(\alpha,3n)$ reaction[23] so that no Co^{56} was present in the source. Co^{57} was present, but since the energies of the gamma rays in Co^{57} are so low in comparison with those in Co^{58} this presented no difficulty.

(b) Construction of the Source

About 100 microcuries of Co^{58} were grown in 19 single crystals of diluted Cu-K_2 Tutton salt having a total mass of 2.7 grams. The glass support for the paramagnetic crystals is shown in Fig. 3. The radioactive crystals were glued on 0.005-in. silver foils of 99.95% purity which had been glued to the glass plate *D* by a 1:1 mixture of toluene and General Electric 7031 varnish. About 5 grams of $Mn(NH_4)_2(SO_4)_2 \cdot 6H_2O$ were glued on similar silver foils which had been glued to the glass plate *B*. A lead wire having dimensions 3.5 cm $\times$0.04 cm $\times$0.01 cm was soldered to the silver foils and formed a thermal switch[24] between the glass plates *B* and *D*. The glass plates were separated from one another and from the main glass support by the glass tubes *A* and *C* which were 0.2 cm in diameter and had 0.015 cm wall thickness.

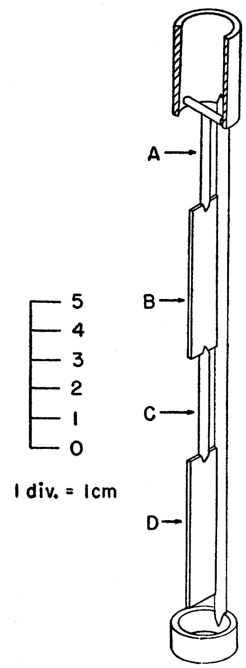

FIG. 3. Glass crystal mount.

(c) Demagnetization Cycle

A qualitative examination of the various possible angular distributions consistent with the decay scheme given in Fig. 1 for either γ_1 or γ_3 will show that at 90° to the alignment axis the maximum effect of the beta decay on the normalized intensity is of the order of 4–5%. Since γ_3 is so low in intensity, 1% counting statistics are about the best possible, so that very low temperatures are essential if definitive results are to be obtained regarding the nature of the beta interaction. With the available apparatus, an initial temperature of 1.3°K and an external magnetic field of 7.5 kilogauss were attainable. Under these conditions final temperatures of about 0.03°K could be reached by single-stage demagnetization. Since this was clearly inadequate for this experiment, it was necessary to make use of the two-stage adiabatic demagnetization technique, discussed in some detail by Daunt *et al.*[24] and by Darby *et al.*[25] By using a simplified scheme, the initial temperature was decreased from 1.3°K to about 0.3°K in the first stage of the demagnetization, thereby decreasing the attainable temperature to about the lowest possible, 0.012°K. The cycle required 30 minutes to complete and the radioactive crystals warmed up to the bath temperature in about 23 minutes. The total heat input was estimated from the calculated entropy to be about 75 ergs/min. The 100 microcuries of radioactivity accounted for nearly all of this heat input.

(d) Gamma-Ray Spectroscopy

The scintillation counter crystals were cylinders of NaI(Tl) 3 cm long and 3 cm in diameter, and were located 11 cm from the source throughout the experiment. The measured angular acceptance of the counter was 14.5°, and was determined to have a negligible effect on the measurement of the angular distribution. Just one counter was used for all the measurements, and the data for the two gamma-ray energies were taken simultaneously with this one counter.

The pulse-height selector (PHS) spectrum of the gamma radiation from the source is shown in Fig. 4. The photopeaks due to the 0.511-Mev annihilation radiation, 0.805-Mev gamma ray, and 1.6-Mev gamma ray are indicated on the figure. The PHS was set from *A–B* for γ_1, and from *C–D* for γ_3 during the measurement of the angular distribution in the alignment experiment.

The intensity of γ_1 was so much stronger than that of γ_3 that chance coincidences of two 0.805-Mev gamma rays resulting in photoelectric events in the scintillation counter appeared as single 1.6-Mev photoelectric

[23] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).
[24] C. V. Heer and J. G. Daunt, Phys. Rev. **76**, 854 (1949); J. G. Daunt and C. V. Heer, Phys. Rev. **76**, 985 (1949).
[25] Darby, Hatton, Rollin, Seymour, and Silsbee, Proc. Phys. Soc. (London) **A64**, 861 (1951).

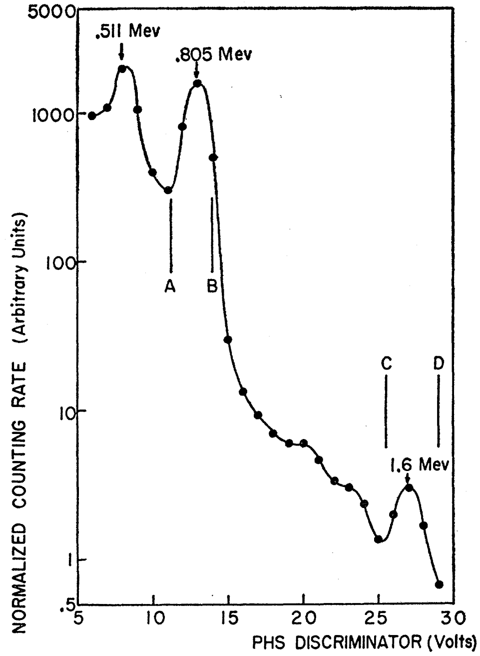

FIG. 4. Pulse-height spectrum of Co^{58} decay showing the annihilation radiation photopeak, 0.805-Mev gamma-ray photopeak, and 1.6-Mev gamma-ray photopeak.

events. These chance 1.6-Mev events produce a so-called "sum line." If $R_{0.8}$ and $R_{1.6}$ are the counting rates due to photoelectric events for the 0.805-Mev and 1.6-Mev gamma rays respectively, then the chance coincidence rate, R_c, will be

$$R_c = (R_{0.8})^2 2t, \qquad (13)$$

where t is the time that the pulse from the counter is in the voltage region corresponding to about 0.8 Mev. The counting rates were approximately 320/sec and 1/sec for the regions A–B and C–D respectively. The absorption measurements below show that the chance coincidence rate represented about 20% of the total counting rate observed at 1.6 Mev.

In order to determine quantitatively the fraction of the total counting rate at 1.6 Mev due to these chance coincidences, the PHS was set from C–D and the counting rate taken as a function of absorber thickness. These data are presented in Fig. 5. The resulting absorption curve is the sum of two straight lines. The absorption coefficients of these two lines are 21.6 and 5.6 g/cm² corresponding to gamma-ray energies of 1.9 and 0.45 Mev respectively. The first is in reasonable agreement with the expected value for the 1.6-Mev gamma ray since the absorption coefficient varies slowly with energy in this energy region. The second is in agreement with the value one would expect for the absorption of chance coincidences of two 0.805-Mev gamma rays. The absorption coefficient in the latter case is just half the absorption coefficient for a single 0.805-Mev gamma ray. The absorption coefficient of 11.2 g/cm² corresponds to a gamma-ray energy of 0.78 Mev, in support of the conclusion that the lower absorption coefficient component is a "sum line."

At zero absorber thickness the "sum line" accounts for 20.4% of the total counting rate. As a compromise between total counting rate and sum line contamination, an absorber thickness of 5.6 g/cm² was used during the measurements of the angular distribution. This reduced the sum line contribution to the total counting rate from 20.4% to 10.8%.

In an alignment experiment the counting rate is measured as a function of susceptibility at fixed angle. Depending on the angle of observation, the observed counting rate either increases or decreases through the course of the experiment as it would if the strength of the source were changing. The counting rate for a true gamma line is proportional to the source strength, but for a sum line the counting rate is proportional to the square of the source strength. Let $f_{1.6}$, $f_{0.8}$, and f_s be the counting rates of the 1.6-Mev photopeak, 0.805-Mev photopeak, and sum line respectively, normalized to their values at 1.3°K, as functions of temperature. Then it can be shown that they are related to the normalized total rate, f_t, by

$$f_t = f_s + f_{1.6} = x f_{0.8}^2 + (1-x) f_{1.6}, \qquad (14)$$

where x is the fraction of the total rate at high temperature due to the sum line. The absorption measurements determine x. The alignment experiment determines f_t and $f_{0.8}$. Then $f_{1.6}$ is calculated from Eq. (14).

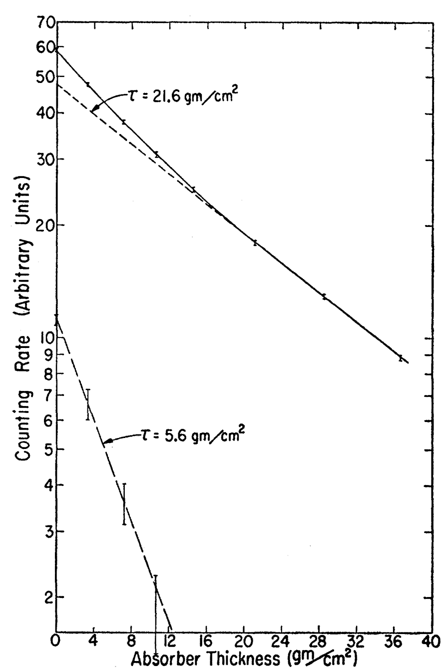

FIG. 5. Graph of absorption measurements of 1.6-Mev photopeak of Co^{58} showing the contributions of the true 1.6-Mev line and sum line to the total absorption curve.

(e) Preliminary Measurements of the Angular Distribution

In order to determine the plane of the tetragonal axes for the cobalt ions, a radioactive measurement is necessary since the magnetic properties of the crystals are determined by the copper ions. The 0.805-Mev gamma ray is known to produce an increase in counting rate at 90° to the alignment axes.[4] To determine approximately the laboratory angle corresponding to the maximum counting rate, the sample was demagnetized and the counting rate determined at a fixed temperature *versus* the laboratory angle φ. The angle φ is that between the counter axis and the plane of the tetragonal axes. The angle θ between the counter axis and the tetragonal axis is related to φ by the equation

$$\cos\theta = \cos\alpha \cos\varphi, \tag{15}$$

where 2α is the angle between the two tetragonal axes shown in Fig. 2.

IV. TREATMENT OF EXPERIMENTAL DATA

(a) Radioactive Decay

The data for γ_1 were treated first. The counting rate, normalized to that at 1.3°K, was plotted *versus* susceptibility for each angle and a smooth curve drawn through each set of points. The deviation of each point from this curve was determined and these deviations plotted against the number of points in a given deviation interval. The standard deviation determined in this manner agreed with that calculated from the number of counts. All data were rejected which lay outside four times this standard deviation. Two experimental points were rejected in this way out of a total of 561. The susceptibility axis was divided into twenty bins and the ordinates and abcissas averaged for each bin. The resulting experimental points are given in Fig. 6. The statistical errors are not given on the graph for these points since they are so small. The errors for the 86.5° points do not exceed ± 0.0011, while the errors for the other two angles do not exceed ± 0.0020.

Nineteen crystals were used in the source, and some error was no doubt present because of failure to achieve the same orientation for all the crystals. Each crystal had one flat face bearing on the silver foil (see Sec. III-b) and at least one face bearing on an adjacent crystal. The 19 crystals were arranged with 12 on one side of the plate and 7 on the other side. Each crystal within one of these groups was accurately aligned with respect to all the other crystals in the group. The two silver foils were parallel to within 1°. These uncertainties introduce errors small compared to the statistical errors.

The normalized counting rate is changing with angle most rapidly in the region where it does not change with temperature, i.e., at the intermediate position of the counter. Therefore, in applying the theory to the

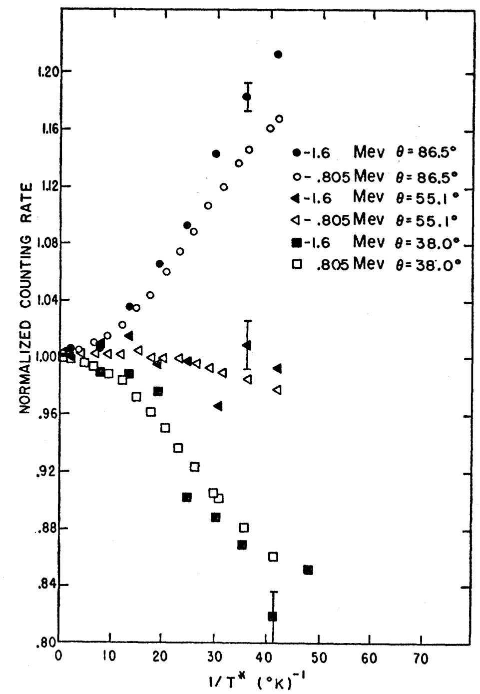

FIG. 6. Normalized counting rate *versus* $1/T^*$ for the 0.805-Mev and 1.6-Mev gamma rays in Co^{58} for three angles between the counting axis and the tetragonal axis.

experimental curves, the data at the intermediate angle were most sensitive to the assumptions made in choosing the theoretical parameters, particularly the parameter λ, defined in Eq. (6), describing the character of the beta transition. As pointed out in Sec. III-g, the orientation of the alignment axes with respect to the laboratory was determined initially only approximately. By using internal consistency arguments, however, the accuracy of the data enabled the angles θ and α, and the parameter λ to be determined. The method used to determine these quantities is described in the following paragraphs.

Since the susceptibility and $\beta = A/4kT$ are in 1:1 correspondence, the three values of normalized counting rate for a particular susceptibility all correspond to the same value of β. Now the initial selection of angles (Sec. III-e) insures that one position corresponds to an angle with the alignment axes of about 90° while a second corresponds to an angle with the alignment axes of about α. In these regions of angle, the normalized counting rate varies slowly with angle. Therefore, by using Eq. (5) and assuming a definite value for λ, β is determined from the $\varphi \simeq 90°$ data, since in this region one had $\varphi \simeq \theta$. Given this value for β, α is determined from the $\varphi \simeq 0°$ data. Given these values for β and α, the intermediate angle θ_i is determined from the experimental data and the theoretical expression given in Eq. (5).

The values obtained for α and θ_i are functions of the parameter λ so that by using the values of normalized

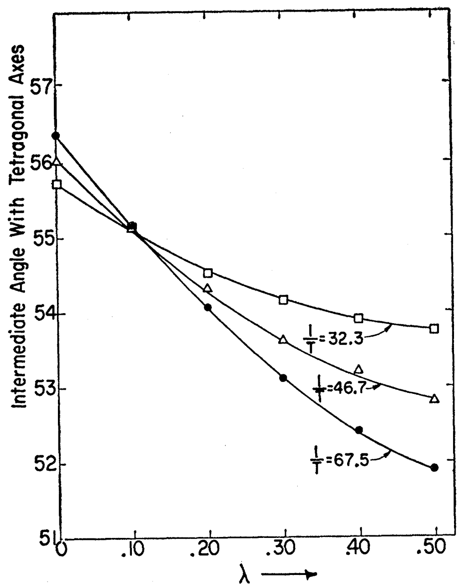

FIG. 7. Intermediate counting angle *versus* λ for three values of absolute temperature.

counting rate corresponding to one value of susceptibility, curves of both θ_i and α *versus* λ may be plotted. Now regardless of the value of the susceptibility the values for θ_i, α and λ must be the same. Thus sets of parametric curves of both θ_i and α *versus* λ may be plotted with susceptibility as a parameter. Their intersection corresponds to that set of coordinates (θ_i,λ), (α,λ) which fits the data at all temperatures. The curves of θ_i *versus* λ are shown in Fig. 7 for three values of susceptibility. The values of $1/T$ given for each curve correspond to the value of β derived for $\lambda=0.11$. The curves of α *versus* λ are not shown, but the intersection yields the same value of λ as well as determining α.

The statistical error in the raw data results in angular errors of the order of 0.2–0.3°. The fact that there is a common intersection to within the errors expected from the raw data shows that the data are internally consistent. The method of treatment is iterative so that the propagation of errors is very complex. The error assigned to λ is ± 0.04.

Once these three parameters have been established for the γ_1 data, a relation is established between the magnetic susceptibility and the parameter β describing the theoretical curve. In order to determine the true normalized counting rate for γ_3 as a function of the magnetic susceptibility, the sum line must be subtracted as discussed in Sec. III-d. In order to effect this subtraction, the experimental data were divided into bins as described in the first paragraph of this section. The ordinates and abcissas were averaged, and then at each value of the magnetic susceptibility the value of $f_{0.8}$ and f_t were taken from the experimental data and the corresponding value of $f_{1.6}$ determined from Eq. (15). The latter data are presented in Fig. 6 along with the γ_1 data. Sample statistical errors are indicated on one point for each angle. The data at 86.5° were obtained with greatest precision for both gamma-ray energies.

Since the two unknown angles α and θ_i and the relation between the susceptibility and the theoretical parameter β were determined from the γ_1 data, the only problem in treating the γ_3 data is the determination of λ, if one assumes the $2-2-0$ cascade. This assumption will be discussed in Sec. V. By using the γ_1 results, curves of normalized counting rate for γ_3 *versus* β were plotted. Then theoretical curves of normalized counting rate *versus* β were calculated for various values of λ. Next the sum of the squares of the deviations of the experimental points from each of the various theoretical curves was calculated. These data are presented in Fig. 8. The vertical axis is the reciprocal of the sum of the squares of the deviations so that the peak value of each curve is proportional to its statistical weight, and the value of λ at this peak corresponds to the best fit.

(b) Magnetic Susceptibility

The susceptibility as a function of $1/T$ was determined by using the γ_1 data from the curve of susceptibility *versus* normalized counting rate at 86.5°. From a given value of normalized counting rate, β may be calculated by applying Eq. (5). Equation (2) then yields $1/T$ if A is known. The parameter A was measured for stable Co^{59}.[18] The value of A for Co^{58} is given by

$$A_{58}=\frac{(\mu/I)_{58}}{(\mu/I)_{59}}A_{59}. \tag{16}$$

Since both $(\mu/I)_{58}$ and $(\mu/I)_{59}$ have been measured,[14,26] the experimental value of A_{59} determines A_{58}. Thus the relation between the susceptibility and $1/T$ is determined. The Curie law part of the susceptibility occurs in the high-temperature region.

According to Eq. (11), the ratio $\chi(T)/[\chi_0(T)F(T)_{\text{hfs}}]$ represents the contribution of the interactions to the susceptibility, and should fit a parabola in $1/T$. The constants Δ and ϵ in Eq. (11) were determined by curve fitting to be $\Delta=-(8.0\pm2.0)\times10^{-3}$ K° and $\epsilon=+(4.4\pm0.5)\times10^{-5}$ K°^{2}. This procedure yields a semiempirical determination of the relation between the absolute temperature and either the magnetic susceptibility or the magnetic temperature, which is defined by the relation

$$T^*=c/\chi(T^*). \tag{17}$$

V. RESULTS AND DISCUSSION

(a) Radioactive Decay

The curves shown in Fig. 7 relating to γ_1 give the values $\theta_i=55.0\pm0.3°$ and $\lambda_{0.8}=0.11\pm0.04$. The cor-

[26] W. G. Proctor and F. C. Yu, Phys. Rev. **77**, 716 (1950); W. G. Proctor and F. C. Yu, Phys. Rev. **81**, 20 (1951).

responding curves of α *versus* λ (not shown) give the values $\alpha=37.7\pm0.3°$ and $\lambda_{0.8}=0.12\pm0.04$. These results are all for the beta transition to the first excited state in Fe^{58}.

The character of the beta decay to the 0.805-Mev level has been determined by Grace.[27] His result indicates $\lambda_{0.8}=0.1$, in agreement with the value of 0.11 determined in this experiment.

Another independent value for $\lambda_{0.8}$ may be obtained as follows. The ratio of the magnetic moments of Co^{58} and Co^{60} have been determined[28] as a function of the assumed nature of the two decay schemes. For a complete Gamow-Teller decay in Co^{58} and for a $5-4-2-0$ cascade in Co^{60}, the ratio of magnetic moments was determined to be 1.10 ± 0.03. Jeffries *et al.*[14] have determined the absolute values of the magnetic moments of Co^{58} and Co^{60} by means of a refined paramagnetic resonance technique. The ratio of magnetic moments as determined from their absolute values is 1.065 ±0.016. The results of Jeffries may be combined with the results of the $Co^{58}-Co^{60}$ ratio experiment above to determine an independent value for $\lambda_{0.8}$. This method gives $\lambda_{0.8}=0.08\pm0.06$ in agreement with the new results.

At the lowest temperature reached, 0.012°K, the normalized counting rate for γ_3 was observed to be 1.23 at 86.5° to the alignment axes. At $T=0$, the normalized counting rate at 90° to an alignment axis is 0.75 for an $L=1$ gamma ray in a $2-\xrightarrow{\beta}1-\xrightarrow{\gamma}0$ cascade, 1.25 for an $L=2$ gamma ray in a $2-\xrightarrow{\beta}2-\xrightarrow{\gamma}0$ cascade, and 1.44 for an $L=3$ gamma ray in a $2-\xrightarrow{\beta}3-\xrightarrow{\gamma}0$ cascade. Since the observed counting rate for γ_3 is higher than that for γ_1 in the known $2-\xrightarrow{\beta}2-\xrightarrow{\gamma}0$ cascade, it is not possible that γ_3 be part of a $2-\xrightarrow{\beta}1-\xrightarrow{\gamma}0$ cascade. Moreover, the counting rate at the lowest temperatures is substantially less for γ_3 than that theoretically expected for a pure $L=3$ gamma ray in a $2-\xrightarrow{\beta}3-\xrightarrow{\gamma}0$ cascade. A large admixture of $L=2$ and a change of the decay scheme to include a heretofore unobserved spin 1 level would be necessary for γ_3 to be partly $L=3$. Thus we conclude that γ_3 is $L=2$ and part of a $2-\xrightarrow{\beta}2-\xrightarrow{\gamma}0$ cascade, in agreement with the results of Frauenfelder and co-workers. Consequently, the difference in the angular distributions for γ_1 and γ_3 must result from different values for λ. The results presented in Fig. 8 give $\lambda_{1.6}$ for each of the three counting angles. The weighted average of these three values is $\lambda_{1.6}=0.45$ ±0.11.

The value of K, Eq. (6), calculated from the experimental data,[17] ranges from about 0.5 to 1.4, depending on the case. If K is taken to be unity, then in the beta decay to the 0.805-Mev level in Fe^{58} the ratio of nuclear matrix elements $|\int 1|^2/|\int\sigma|^2$ is about $\frac{1}{8}$, whereas in the decay to the 1.6-Mev level this ratio is about unity. If

[27] M. A. Grace (private communication to J. C. Wheatley).
[28] Wheatley, Griffing, and Hill, Phys. Rev. **99**, 334 (1955).

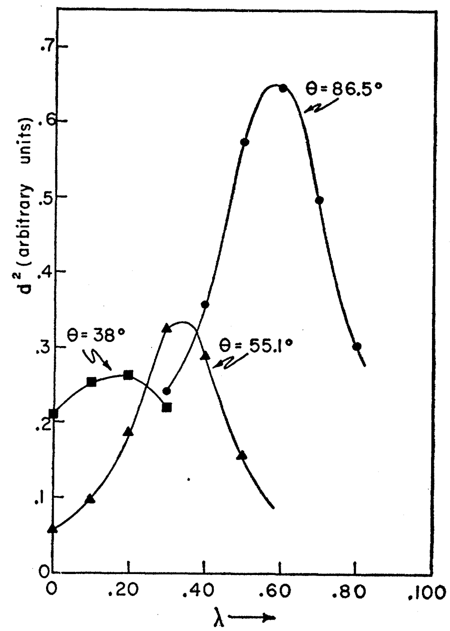

FIG. 8. Reciprocal of the sum of the squares of the deviations of the experimental curves shown in Fig. 6 from theoretical curves calculated for various values of λ.

K is not unity, then the ratio of nuclear matrix elements above is changed proportionately.

The values of λ were obtained by using orientation parameters f_k determined both for the approximate case with $B=0$, and for the expected case with $A/B=4$. The values of λ obtained as described in Sec. IV-a were the same for both cases to within the assigned error. Thus λ is independent of the exact validity of $A/B=4$. Moreover, λ is determined through internal consistency arguments without ever invoking any particular magnetic temperature-absolute temperature relation and therefore is independent of this relationship.

(b) Magnetic Susceptibility

The curve of $1/T^*$ *versus* $1/T$ determined as discussed in Sec. IV-b is given in Fig. 9, where the smooth curve represents the semiempirical formula and the circles the experimental points. The treatment of the γ_1 data to determine the $T-T^*$ relation, Fig. 9, depends on the assumed parabolic dependence of the contribution to the susceptibility of the interactions. The fact that the parabola represented the data very well in the range of temperature from $1/T=30$ to $1/T=85$ supports this assumption. The errors assigned Δ and ϵ take into account all the sources of error which were considered and are therefore not statistical errors. The effect of the magnetic dipole-dipole and exchange interactions on the paramagnetic susceptibility was estimated in Sec. II-c by using the theory of Daniels[18] in conjunction with the experimental data of Benzie *et al.*[20] The

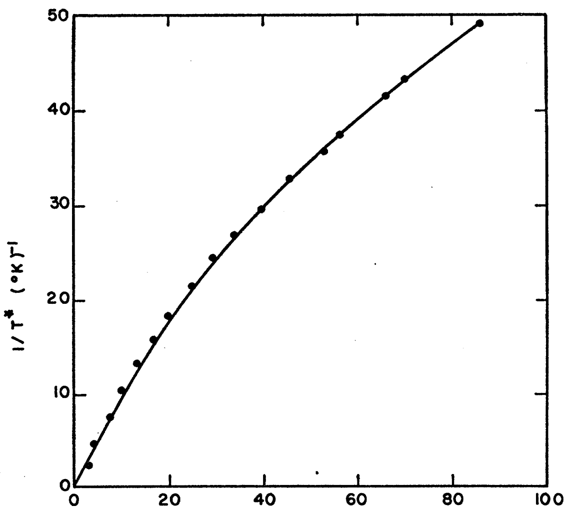

FIG. 9. Relation between $1/T$ and $1/T^*$. The solid curve fits a semiempirical formula. The circles are experimental points.

numerical results of these calculations are

$$\Delta_{max} = -8.8\times 10^{-3}\ \text{K}^\circ \quad \text{(antiferromagnetic)}$$
$$= +7.5\times 10^{-3}\ \text{K}^\circ \quad \text{(ferromagnetic)},$$
$$\epsilon = +4.4\times 10^{-5}(\text{K}^\circ)^2 \quad \text{(either)}.$$

These results are to be compared with the experimental results of the present work, given by

$$\Delta = -(8.0\pm 2.0)\times 10^{-3}\ \text{K}^\circ,$$
$$\epsilon = +(4.4\pm 0.5)\times 10^{-5}(\text{K}^\circ)^2.$$

The calculated sign of ϵ is necessarily positive, but the calculated sign of Δ is ambiguous depending on whether the exchange interaction is antiferromagnetic or ferromagnetic. A comparison of the theoretical and experimental values shows that the experimental results are consistent with a large antiferromagnetic exchange interaction and a small magnetic dipole-dipole interaction between paramagnetic copper ions.

The paramagnetic susceptibility of concentrated Cu-K_2 Tutton salt has been measured in the temperature range above 0.07°K by DeKlerk,[29] Garrett,[30] and Cooke *et al.*[31] The absolute values of Δ obtained by these investigators disagree in magnitude but are all positive and therefore consistent with a ferromagnetic rather than an antiferromagnetic exchange interaction.

It is felt that it is completely unreasonable to assume that the measured A and B constants in the spin Hamiltonian are sufficiently incorrect (they would have to be increased by about a factor of 4) that an error in the hfs correction alone would account for the negative sign of Δ obtained in this work. A set of nineteen single crystals were used instead of an ellipsoid. It is conceivable that this had some effect on the low-temperature susceptibility.

None of the sources of error which have been considered will produce a correction which would change the sign of Δ from minus to plus. Therefore, it must be concluded either that in fact in these crystals the exchange interactions are antiferromagnetic or that there is some unknown factor which causes a systematic error in the conversion, via the theory, of the values of experimental normalized counting rate to the values of $1/T$. In view of the above, there is still some question as to the validity of using the radioactive counting rate as a thermometer.

It is interesting to note that in similar experiments by Poppema *et al.*[5] using Co^{60} in dilute cobalt ammonium Tutton salt crystals, a relationship between T and T^* was obtained which was unexpected and unexplained. T^* should have been greater than T, whereas in fact it was found to be less. In the experiments of Bleaney *et al.*[12] using copper Tutton salts, the contribution of interactions also appears to make T less than T^*, contrary to expectations.

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[30] C. G. B. Garrett, Proc. Roy. Soc. (London) **A203**, 375 (1950).
[31] Cooke, Meyer, and Wolf, Proc. Roy. Soc. (London) **A233** 536 (1956).