Effects of K-Capture Dealignment and Ionic Couplings on Low-Temperature Nuclear Alignment*

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The angular distributions of γ rays emitted from traces of Co^{58} aligned in single crystals of (x Ni, $(1-x)$ Zn, Co^{58})$SiF_6 \cdot 6H_2O$ have been studied with various nickel fractions (x) between 0 and 0.19. The nuclear alignment is found to be strongly influenced by the presence of paramagnetic nickel ions in the nickel-bearing crystals. Crystals of 0% nickel show evidence of a small dealignment of the 10^{-11} sec intermediate state which just precedes emission of the 810-keV γ ray. This is due to the highly ionized and optically excited electronic shell which is present after the K capture.

INTRODUCTION

THE technique of nuclear alignment of radioactive elements in paramagnetic crystals has been extensively used to study such nuclear properties as magnetic moments and spins, as well as γ-rays multipolarities. It has also been pointed out that such techniques are potentially useful for the study of crystalline properties when the properties of the nuclear processes studied are sufficiently well known. As illustration, the present experiment, begun to study a particular nuclear problem, concluded as a study of the ionic interactions of the paramagnetic crystal used and their effects on nuclear alignment studies.

The experiment was initiated to study the allowed β decay of Co^{58} to the first excited state of Fe^{58}. The decay scheme is shown in Fig. 1. This decay may proceed either through the Gamow-Teller interaction, in which the spins of the β particle and the neutrino combine to carry away one unit of angular momentum, or through the Fermi interaction, in which no angular momentum is carried away. The parameter λ, which is defined as the fraction of Fermi β decays, was originally studied by Griffiing and Wheatley.[1] Their experiments required a higher degree of precision than these because they used Co^{57} aligned in (0.23 Cu, 0.66 Zn)$K_2(SO_4)_2 \cdot 6H_2O$ which has two nonequivalent cobalt lattice sites, each with an uncertain alignment axis direction. They obtained $\lambda = 0.11 \pm 0.04$. Later, Boehm and Wapstra,[2] who studied the circular polarization of the γ ray following β decay, obtained $\lambda < 0.012$. This number was not obtained directly, but relied on the non-parity-conserving theory of β decay. Later, and simultaneously with part of these experiments, Dagley *et al.*[3] using Co^{58} aligned in (Ni,Zn)$SiF_6 \cdot 6H_2O$ obtained $\lambda = -0.003 \pm 0.005$. This result is much more accurate than the older Griffing and Wheatley result due to the choice of a salt. All the cobalt lattice sites in (Ni,Zn)$SiF_6 \cdot 6H_2O$ are equivalent.

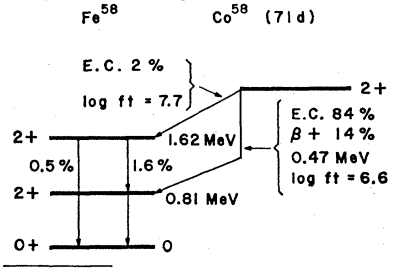

FIG. 1. Decay scheme of Co^{58}. E.C. is electron capture.

However, this report demonstrates that even these results can be improved in accuracy if an even better salt is chosen, namely, $ZnSiF_6 \cdot 6H_2O$. Because Co^{58} is present only in trace amounts, there is no appreciable interaction between magnetic ions to give an unpredictable initial alignment to the Co^{58} nucleus as is the case when Ni ions are included.

The physical effect of interest in this experiment is the deorientation of the initially oriented Co^{58} nuclei as a result of the β decay. Since the Gamow-Teller decays contribute to such a deorientation while the Fermi decays do not, this is a sensitive measure of their ratio.

The deorientation can be studied by observing the angular distribution of the succeeding γ rays about the axis of nuclear alignment. The computation of λ from this result then depends only on a knowledge of the spins and multipolarities involved, and the initial alignment, provided only that no other external influence acts to reorient the nucleus during the lifetime of the intermediate state.

The 810 keV γ intensity is a function of polar angle (θ) with the alignment axis case and can be written from equations given by Cox *et al.*[4] as

$$W(\theta) = 1 - (5/7) f_2 (1+\lambda) P_2(\cos\theta) + (80/9) f_4 (1 - 5\lambda/2) P_4(\cos\theta), \quad (1)$$

where f_2 and f_4 are parameters specifying the original alignment of the Co^{58} nuclei whose computation is discussed in the theory section.

In this experiment, Co^{58} nuclei were aligned in single

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[1] D. Griffing and J. Wheatley, Phys. Rev. **104**, 389 (1956).
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crystals of $(Zn,Ni)SiF_6 \cdot 6H_2O$ with nickel concentrations varying from 4 to 19%. Values of λ were obtained by fitting the observed γ-ray angular distributions to curves obtained from Eq. (1). The values of λ obtained by this process ranged from -0.12 to $+0.02$ as a function of temperature and nickel concentration. This result can be interpreted as a breakdown of the simple spin Hamiltonian theory (used to predict the initial alignment) due to the interactions of Co ions with neighboring paramagnetic Ni ions.

To eliminate such interaction effects further studies were made of Co^{58} nuclei aligned in nonparamagnetic surroundings. The results of these experiments, discussed in detail below, indicate the presence of other deorienting effects during the lifetime of the intermediate state since negative values of λ are still obtained. It is probable that these effects must be attributed to high magnetic fields generated as aftereffects of K capture and the following Auger cascade.

Because of the difficulty in accounting for ionic interaction effects on the nuclear alignment and the apparent presence of other perturbing influences, it is possible to conclude only that $\lambda \simeq 0$. The experiment provides an interesting light on the nature of ionic interactions in $(Zn,Ni)SiF_6 \cdot 6H_2O$ and illustrates the strong effects of such interactions on the results of nuclear alignment experiments in this substance.

EXPERIMENTAL TECHNIQUE

The experiments used single crystals grown from a water solution of $ZnSiF_6$ and $NiSiF_6 \cdot 6H_2O$ plus a trace of $Co^{58}Cl_2$. The crystals were usually perfect hexagonal columns of about 2.5 g. Each crystal was mounted on a pedestal supported by a thin glass rod from the bottom of the glass sample chamber. A $KCr(SO_4)_2 \cdot 12H_2O$ guard salt was also mounted on this rod to keep the heat leak small. The demagnetizations were made from a magnetic field of 20 000 G at a temperature of 1°K. The final temperature of the salt varied from 0.006 to 0.02°K depending on the percentage of nickel in the crystal.

An array of four NaI(Tl) scintillation crystals was used to measure the angular distribution of 810 keV γ rays. One counter was placed along the axis of nuclear alignment and another at 90° to this axis. The second two counters were placed at 45° positions, either in the plane of the 0° and 90° counters or in the plane determined by the vertical axis and the alignment axis, and were used to ensure that the counters were correctly placed with respect to the alignment axis.

In a typical run counts in each detector were recorded for 100-sec intervals over a period of about $\frac{1}{2}$ h after demagnetization. Transfer gas was then put into the sample chamber to bring the crystal to 1°K and several further counts were taken. The normalized counting rate $W(\theta)$ was obtained by dividing each count by the average warm counting rate. Corrections were made for the effect of the weak competing Co^{58} γ rays, the finite solid angle of the counters, the measured counter noise, and the Co^{60} contamination in the sample. These corrections were generally less than 0.02 in λ. The data from the 45° counters were found to be consistent with the 0° and 90° data within the limits of error, i.e., $\pm 2°$ misalignment. A χ^2 test for each crystal showed the data to be statistically significant and internally consistent.

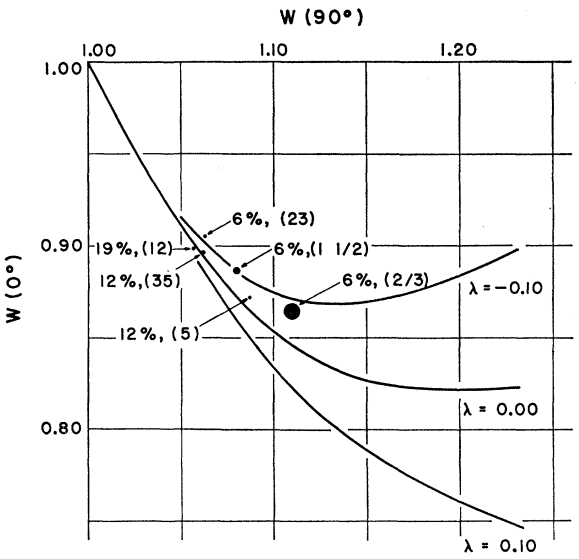

FIG. 2. $W(0°)$ vs $W(90°)$ for 6, 12, and 19% Ni uniform crystals. The number in parenthesis associated with each point represents the average *time in minutes* for the sample to warm up to that temperature. The size of the point represents both statistical counting error and uncertainties associated with shifts in the electronics, etc. Theoretical curves are for no interactions or K-capture dealignment.

The results were plotted on a graph as in Figs. 2 and 4 and compared to curves plotted from Eq. (1) using values of f_2 and f_4 derived on the basis of simple spin Hamiltonian theory as discussed in the theory section. Values of λ as a function of temperature and concentration were, thus, obtained.

EXPERIMENTAL RESULTS

The first set of data was taken using crystals which were uniform in nickel and Co^{58} concentration. Three separate studies were made of crystals containing 6, 12, and 19% molar nickel concentrations, respectively, as determined by chemical analysis. These crystals contained approximately 15 μC of Co^{58} and were grown from saturated water solutions with molar Ni-Zn ratios of 0.043, 0.085, and 0.17, respectively.

The results of these experiments, obtained in 11, 4, and 4 demagnetizations, respectively, are shown in Fig. 2. The points shown are obtained by averaging results from all runs for the same crystal which fall in the same apparent temperature ranges. The ranges chosen were $\beta<1.00$; $1.00<\beta<1.3$; $1.3<\beta<1.6$; $1.6<\beta<2$; and $2<\beta$ ($\beta=A/2kT$, where AS_zI_z gives the hyperfine interaction energy due to the z component of the nuclear spin).

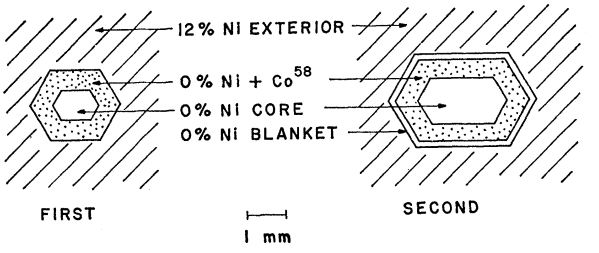

FIG. 3. Structure of 0% crystals. Cross section perpendicular to the axis of alignment.

The results obtained from these three experiments indicated that the observed value of λ depended strongly on Ni concentration and it was felt that an attempt should be made to study the behavior of Co^{58} nuclei in an environment free of Ni ions. For this purpose two separate layered crystals were grown. Figure 3 shows the construction of these crystals. The first weighed 2.3 g with 4 μC of radioactivity, the second 1.4 g with 1.8 μC of radioactivity. The results of these experiments, grouped in temperature and averaged as previously discussed, are shown in Fig. 4. It must be noted that the impurities present in the Co^{58} sample as obtained from Oak Ridge could be as concentrated as $\frac{3}{4}$ molar percent when grown in a thin layer. These impurities include paramagnetic Ni, Fe, Co, etc., whose interaction with Co is not well known and could possibly affect the alignment parameters somewhat in these cases. We believe, however, that the results should closely represent the condition of zero concentration.

THEORY

The angular distribution of γ rays from aligned Co^{58} nuclei is given in Eq. (1). In this formula, f_2 and f_4 and λ are to be determined. The parameters f_2 and f_4 specify the orientation of the parent nucleus and depend on crystalline properties, while the parameter λ depends on nuclear properties. In terms of expectation values of powers of the z projection of the nuclear spin, I_z, f_2, and f_4 can be defined as

$$f_2=\frac{1}{4}(\langle I_z^2\rangle-2),$$
$$f_4=\frac{1}{16}\left(\langle I_z^4\rangle-\frac{31}{7}\langle I_z^2\rangle+\frac{72}{35}\right). \quad (2)$$

One can calculate f_2 and f_4 as a function of the temperatures in a straightforward way if the hyperfine levels of the nucleus in the crystal are known. The assumption is usually made that only the lowest electronic level of the crystal is populated for temperatures of interest ($T<1$°K) and that there is no significant interaction between different ions in the crystal. The hyperfine levels are then well predicted by the spin Hamiltonian which for Co^{58} in $ZnSiF_6\cdot 6H_2O$ has the form,

$$\mathcal{H}/k=g_{\parallel}\beta H_z S_z+g_{\perp}\beta(H_x S_x+H_y S_y)+AS_zI_z+B(S_xI_x+S_yI_y). \quad (3)$$

Here S is the equivalent electron spin, which is $\frac{1}{2}$, and I is the nuclear spin. $S_{x,y,z}$ and $I_{x,y,z}$ are quantum-mechanical operators, A and B describe the hyperfine interaction, and $H_{x,y,z}$ is any external magnetic field. Here β is the Bohr magneton.

The adequacy of this theory has been checked for dilute paramagnetic crystals by paramagnetic resonance and values of the parameters have been obtained.[5] Using these values of $A=0.0403$°K, $B=0.0103$°K, $g_{\parallel}\beta=3.91\times10^{-4}$°K/G, and $g_{\perp}\beta=2.31\times10^{-4}$°K/G; diagonalizing the energy matrix to obtain the eigenstates and their eigenvalues; and then assuming a Boltzman distribution across these levels, the parameters f_2 and f_4 can be calculated as a function of $\beta=A/2kT$. The results of this calculation used in Eq. (1) yield the solid curves shown in Figs. 2 and 4.

In the actual crystals used for our experiments, the assumption made above that there is no interaction between neighboring ions is not clearly justified since the density of paramagnetic ions is relatively high. The Hamiltonian of Eq. (3) can be modified to include these interactions. In the absence of external fields it becomes

$$\mathcal{H}=A(S_{oz}I_{oz})+B(S_{ox}I_{ox}+S_{oy}I_{oy})+\sum_{jk}P(S_j,S_k)+\sum_{i\neq0}D(S_i)+\sum_{i\neq0}Q(S_i,I_i). \quad (4)$$

The subscript "o" is now taken to designate a particular Co atom and the subscript "i" any neighboring

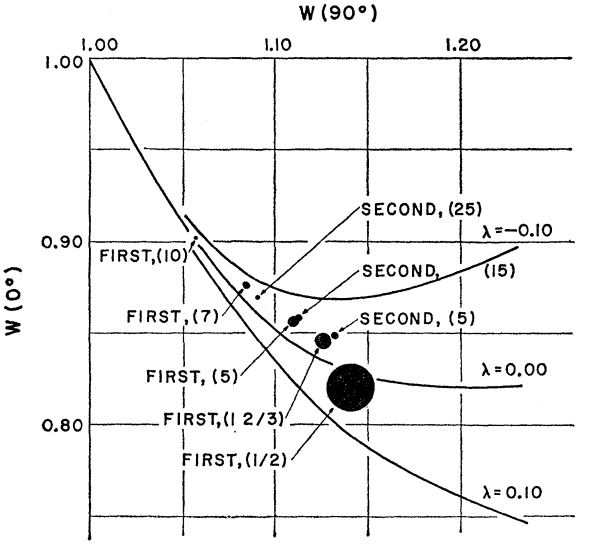

FIG. 4. $W(0°)$ vs $W(90°)$ for 0% Ni layered crystals. The number in parenthesis associated with each point represents the average *time in minutes* for the sample to warm up to that temperature. Theoretical curves are for no interactions or K-capture dealignment.

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paramagnetic nickel ion. The significant additional term is the P term describing the interaction of the Co ion with all neighboring ions. This interaction could be either a magnetic dipole–dipole interaction or an exchange interaction.

These experimental results show that these terms are large enough to influence the experimental results a measurable amount. Very crude theoretical estimates of the sizes of these terms show that even in the dilute (6% Ni) salts it is reasonable for these terms to influence the measured λ an appreciable amount. These estimates are based on the magnitude of the interaction terms obtained from the final temperature reached after demagnetizing (from 1°K and 20 kG in our case).

However, while it is not surprising to get unreal negative measured values for λ in salts containing magnetic Ni ions, it is surprising that λ is negative in the 0% Ni crystals. This leads us to the problem of K-capture dealignment.

There is very little known about the short-lived state which follows K capture. In the case of Fe it is expected from x-ray linewidths that the K shell is vacant for about 4×10^{-16} sec.[6] The theory of Fermi[7] leads us to expect hyperfine levels spread over 0.02 eV (assuming the nuclear g factor is 0.4 in the intermediate state as predicted by the vibrational model). On the basis of any picture of the intermediate state, this is much too weak a coupling to cause significant nuclear precession in 4×10^{-16} sec.

However, the excited ion will undergo an Auger cascade which lasts for about 10^{-14} sec and leaves the ion highly ionized and optically excited. It takes the order of 2×10^{-11} sec for the γ ray to be emitted, and it is during this time that we expect nuclear dealignment to take place.

The details of the Auger cascade are not well enough known to permit a theoretical calculation predicting the effect of the nuclear precession on the alignment. However, the theory of Steenberg,[8] and the theory of Tolhoek *et al.*[9] indicate that dealignment effects will influence the measurement of λ by the order of ±0.04 when

$$(A'\tau/2\hbar)^2\approx0.02. \tag{5}$$

Here A' is the coefficient of the assumed $\mathbf{I}\cdot\mathbf{S}$ hyperfine coupling in the intermediate state and τ is the lifetime of the state ($\approx2\times10^{-11}$ sec). If the coupling constant A' in the ionized and excited intermediate state is only twice that of the Co^{58} ground state, the condition of Eq. (5) is satisfied and we can expect errors in the measurement of λ of ±0.04.

This equation is approximate. The right-hand side will certainly be influenced by the details of the excited electronic state following the ejection of three (on the average) Auger electrons. Weidenmüller[10] gives a rigorous theoretical treatment of this problem in the special case when the dealignment of the intermediate state is due to the short-lived hole in the K shell. While his treatment is not exact in our case (our dealignment is due to excitations and holes in the optical shells following the Auger cascade), it is in agreement with Eq. (5).

DISCUSSION

The results we have obtained indicate that interactions between neighboring ions in paramagnetic $(Zn,Ni)SiF_6\cdot6H_2O$ cause significant deviations from predictions of the simple spin Hamiltonian theory for the alignment parameters of Co in this salt. Estimates of the magnitude of the interactions from thermodynamic data show that they are indeed large enough to be significant. The thermodynamic estimates of the interaction together with a simple model allow predictions of the magnitude of the deviations in alignment which are in qualitative agreement with experimental observations.

It does not seem reasonable that the negative values of λ found in the 0% crystals are due to ionic interactions since the density of paramagnetic ions should be very low in the neighborhood of the Co ions. Effects of temperature inhomogeneities likewise do not seem to account for these results.[11] A possible explanation of these measurements is further deorientation of the intermediate state caused by the high fields resulting from K capture and the subsequent Auger cascade. This hypothesis could be checked by measuring the angular distribution of those γ rays following β decay only.

These experiments demonstrate that ionic interactions can influence the results of a nuclear alignment experiment even when the salt is dilute in paramagnetic ions (6%). Secondly, these experiments point out how such effects can be eliminated as a source of error. Thirdly, they suggest that the intermediate state of the Co^{58} nucleus undergoes an appreciable precession in its 10^{-11} sec lifetime due to the very powerful forces exerted on it by the ionized electronic shell following K capture.

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[7] E. Fermi, Z. Physik **60**, 320 (1930).
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[11] The heat leak into the salt was only 0.2 ergs/sec based on warm-up times and specific-heat measurements on the 15% nickel salt by N. Kurti (private communication). The thermal conductivity of this single crystal will not support an important temperature gradient with this small heat flow. Note also that there is no general tendency for the measured λ's to become more negative as the heating continues following demagnetization.