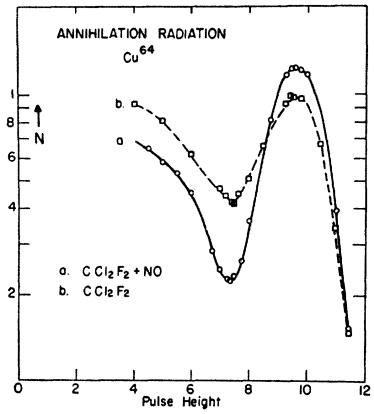

FIG. 1. Annihilation radiation spectra for $f=1$ and $f=0$.

table $2S$ state of hydrogen by an electric field.[3] In the case of positronium in a magnetic field the state with $S=1$, $M=0$ gets a small admixture of $S=0$ by virtue of the quadratic Zeeman effect. Linear Zeeman effect is absent in all states. To an accuracy more than sufficient for our purpose, the fraction of para-state admixed to the ortho state with $M=0$ is $a^2=(2\mu_0 H/E)^2$. Only the ortho-positronium atoms with $M=0$ are affected, since the probability of reorientation before annihilation is negligibly small. The pure ortho-state decays by three-photon emission at the rate $\lambda_0=7\times10^6$ sec^{-1},[4,5] while the para-state decays at the rate $\lambda_p=8\times10^9$ sec^{-1} by two-photon annihilation.[4] The effect of the admixture of para-state in a magnetic field H is therefore twofold: (1) The decay rate of one third of the ortho-positronium atoms is increased to $\lambda(H)=\lambda_0(1-a^2)+\lambda_p a^2$. (2) The probability that an ortho-positronium atom decay by three-photon annihilation, nearly unity in the absence of a field,[5] is reduced to $f(H)=\lambda_0(1-a^2)/(\lambda_0(1-a^2)+\lambda_p a^2)$. By measuring $f(H)$ *vs* H we have found a^2 and from this E. A large volume of uniform magnetic field was provided by a large electromagnet of the Research Laboratory of Electronics.[6] We are indebted to Prof. F. Bitter for the opportunity to use this magnet.

f was determined by observing in a scintillation spectrometer the annihilation radiation spectrum produced in freon. Experimental conditions were much improved over those in our earlier communications.[5] Figure 1 shows typical spectra obtained in the absence of a field in pure freon ($f\simeq1$) and freon with 3 percent nitric oxide ($f\simeq0$). Spectra obtained in pure freon with a magnetic field were intermediate between these two. In the actual experiments only points near the maximum and the minimum of a spectrum were measured with very good statistical accuracy.

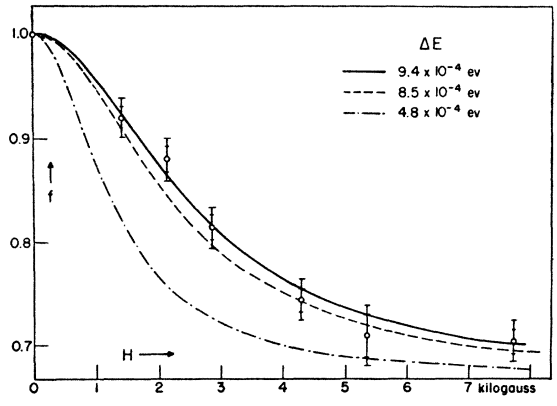

FIG. 2. Relative probability of three-photon annihilation as a function of magnetic field. Theoretical curves for different values of the fine structure of positronium.

f can be calculated from these points after some simple corrections. Figure 2 shows the results obtained. The short cross-bars on the vertical bars indicate the probable scatter of the individual points while the full vertical lengths include the uncertainty of normalization, background, etc. The fact that, for the strongest fields, f approaches $\frac{2}{3}$ of its value at zero field supports our assumption of the absence of re-orientation before annihilation. The solid line in Fig. 2 represents the best fit to the points assuming the dependence of f on H to have the theoretical functional form. It yields the value $E=9.4\times10^{-4}$ ev given above. The two dashed curves are theoretical curves calculated with and without the exchange term. Further discussion of these experiments will be presented in a paper now in preparation.

We wish to thank Dr. Henry Primakoff for valuable discussions of the theoretical aspects of the problem.

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Nuclear Electric Quadrupole Interaction in Crystals with Nonaxially Symmetric Fields*

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POUND[1] has reported the theory and some experiments on the splitting of nuclear magnetic resonance absorption lines in single crystals by the interaction of the nuclear electric quadrupole moment Q with the electric field gradient at the site of the nuclei in question. His work on the influence of the crystal orientation on the line splitting is restricted primarily to the special case of crystals in which the field gradient tensor has an axis of symmetry. Becker and Kruger[2] report further experiments on rotating a crystal with an axially symmetric field. Schuster and Pake,[3] and Hatton, Rollin, and Seymour[4] report some additional work on crystals, but they do not discuss the dependence of the line splitting on the crystal orientation.

We have extended Pound's theory[1] to cover the dependence of the line splitting on the crystal orientation for the general case of crystals in which the traceless symmetric field gradient tensor does not necessarily have an axis of symmetry, but is given in terms of its principal axes by:

$$\partial E_x/\partial x=eq(1-\eta)/2\ \ \partial E_y/\partial y=eq(1+\eta/2\ \ \partial E_z/\partial z=-eq$$

with

$$eq\equiv\Sigma_j e_j(3\cos^2\theta_j-1)r_j^{-3}\ \eta\equiv\left(\frac{\partial E_x}{\partial x}-\frac{\partial E_y}{\partial y}\right)\Big/\frac{\partial E_z}{\partial z}$$

in the notation of Pound's paper.[1] (The z axis is here chosen as the one with the largest absolute value of the field gradient component, and x with the smallest, so that $0\leq\eta\leq1$). We have also experimentally verified this theory in the case of the Li^7 lines obtained from a single crystal of $LiAl(SiO_3)_2$ (spodumene).

We choose three mutually perpendicular right-handed axes in the crystal which are easily recognized externally, and label them X, Y, Z (in the monoclinic spodumene crystal the b axis was chosen as X, the c axis as Y, and the direction perpendicular to b and c as Z. The crystallographic a axis lies between our Y and $-Z$ axes.). Let X have the direction cosines λ_i, Y have μ_i, and Z have ν_i, with respect to the right-handed axes x, y, z in which the field gradient tensor is diagonal at the site of the nuclei in question. (Although in spodumene there are four Li atoms per unit cell the existence of known symmetry centers and of twofold screw axes in the b direction guarantees that the principal axes of the field gradient tensor have the same orientation at the four sites

with some one of the principal axes parallel to the b direction.) Let X, Y, Z in turn be made perpendicular to the uniform external magnetic field H_0, and in each case let the crystal be rotated about this axis normal to H_0. We have found that first-order perturbation theory gives for the frequency difference $2\Delta\nu$ between the two lines arising from the transitions between Zeeman levels m ($>\frac{1}{2}$) and $m-1$ and the levels $-(m-1)$ and $-m$, the expression:

$$2\Delta\nu = A + B\cos 2\theta + C\sin 2\theta, \tag{1}$$

where for the case of the rotation about the X axis

$$\begin{aligned} A_X &= 2k[\tfrac{1}{2} - \tfrac{3}{2}\lambda_3^2 + \tfrac{1}{2}\eta(\lambda_2^2 - \lambda_1^2)] \\ B_X &= 2k[\tfrac{3}{2}(\psi_3^2 - \nu_3^2) + \tfrac{1}{2}\eta(\mu_1^2 - \nu_1^2 - \mu_2^2 + \nu_2^2)] \\ C_X &= 2k[-3\mu_3\nu_3 + \eta(\mu_2\nu_2 - \mu_1\nu_1)] \\ k &= [e^2qQ/h][3(2m-1)/8I(2I-1)]. \end{aligned} \tag{2}$$

Here m is the largest magnetic quantum number of the four values involved in the two transitions. θ_X is measured from the position in which Y coincides with H_0. Expressions for the Y and Z rotations are obtained by cyclic permutation from the above. Pound's results[1] refer to the Y rotation in the special case $\eta=0$, $\lambda_1=\mu_2=\nu_3=1$, the other direction cosines being zero, so that $A_Y=k$, $B_Y=3k$, $C_Y=0$, and Eq. (1) reduces to

$$2\Delta\nu = 2k(3\cos^2\theta - 1).$$

It may be easily shown that of the nine constants A, B, C for the X, Y, Z rotations only five are independent, *viz.* the three C's and any one pair A, B, the other two pairs being related to the first one by the four identities:

$$A_X + A_Y + A_Z = 0, \quad A_Y - B_Y = A_Z + B_Z, \quad A_Z - B_Z = A_X + B_X,$$

$A_X - B_X = A_Y + B_Y$ (from which it also follows that $B_X + B_Y + B_Z = 0$). Thus by performing the three rotations one obtains experimental values for five independent quantities of the nine A, B, C. The five independent relations of the form (2) involving these five quantities, taken together with the six orthonormality conditions for the direction cosines, may be solved for the eleven unknowns consisting of $|k|$, η and the nine direction cosines. This gives an experimental determination of the orientation within the crystal of the principal axes of the field gradient tensor, of the asymmetry parameter η, and of the magnitude of the quadrupole coupling constant $|e^2qQ/h|$.

In some types of crystals it may be known from their symmetry properties that one of the principal axes of the field gradient tensor must be parallel to one of the crystallographic axes. (In the case of spodumene the b axis must be parallel to some one of the principal axis.) If, say, the X axis of rotation could be made to coincide exactly with this principal axis then the above algebra would be considerably simplified, for then $C_Y=C_Z=0$, and the X rotation alone would give the complete information.

The three rotations were performed for a natural single crystal of spodumene of approximate dimensions $10\times10\times8$ mm in a recording oscillating detector spectrometer designed and built by Collins[5] which is similar to the one described by Pound and Knight.[6] The frequency differences $2\Delta\nu$ between the two outer components ($m=I=\frac{3}{2}$) of the Li^7 line was measured in a magnetic field of approximately $H_0=7190$ gauss corresponding to the unperturbed resonance frequency $\nu_0=\mu H_0/Ih=11.90$ Mc/sec. (A spot check was made for one particular crystal orientation which gave a large $\Delta\nu$ to verify that in accordance with Eqs. (1) and (2) the observed $\Delta\nu$ is actually independent of H_0 which was varied from approximately 6500 to 8000 gauss.) Figure 1 is a reproduction of several examples of recorded absorption derivative curves. Figure 2 gives the $2\Delta\nu$ curve for the X rotation.

The other two rotations also gave similar curves of the general form of Eq. (1). The experimental values of the constants (in kilocycles/sec) are:

$A_X=$ 34.0	$B_X=-$ 4.7	$C_X=$41.6
$A_Y=-$14.8	$B_Y=$ 53.0	$C_Y=$ 5.4
$A_Z=-$19.2	$B_Z=-$47.8	$C_Z=$ 1.9

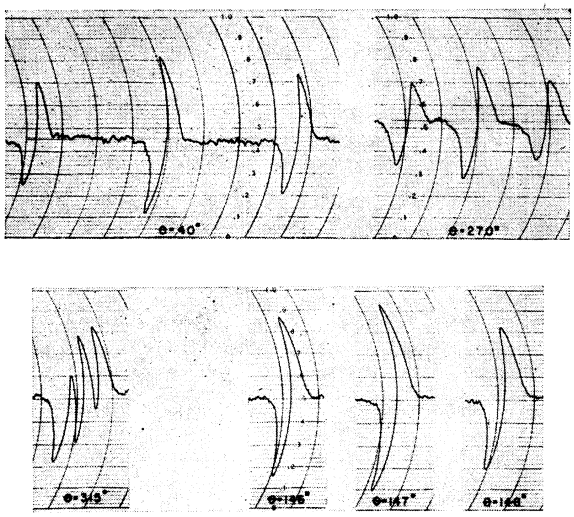

FIG. 1. Examples of recorded absorption derivative curves. The first three show selected splittings for the X rotation. The last three demonstrate the accuracy with which the position of zero splitting at $\theta_Y=147°$ was determined for the Y rotation. A change of θ_Y by $\pm1°$ produces a marked decrease in the maximum of the derivative curve. The sweep rate in all cases is 10 kc/sec per division.

The above deviations of C_Y and C_Z from zero correspond to errors of less than 3° in aligning the rotation axes. The relative signs for the above nine quantities are definitely given by the experiment, but not the absolute signs. The above experimental values lead to the following values computed from the general formulas:

$\eta=$ 0.793	$\lvert k\rvert=18.9$ kc/sec	$\lvert e^2qQ/h\rvert=$ 75.6 kc/sec
$\lambda_1=-0.074$	$\lambda_2=$ 0.997	$\lambda_3=$ 0.020
$\mu_1=$ 0.746	$\mu_2=$ 0.042	$\mu_3=$ 0.665
$\nu_1=$ 0.662	$\nu_2=$ 0.065	$\nu_3=-0.747$

Only the relative signs of the above direction cosines are important. Since $\lambda_2\approx1$ it appears that the y principal axis is parallel to the b direction. The deviation of λ_2 from unity, and of λ_1, λ_3, μ_2, and ν_2 from zero is a measure of the accuracy of aligning the rotation axes.

Experiments on the frequency splitting of the Li^6 and Al^{27} lines in spodumene, and second-order perturbation calculations ap-

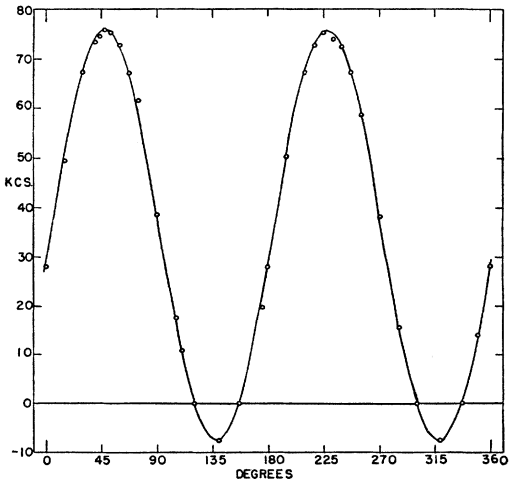

FIG. 2. Frequency difference between the two outer components of the Li^7 line for the X rotation about the b axis of spodumene. The circles are the experimental points. The solid curve represents $2\Delta\nu=33.95-41.85\cos^2(\theta_X+41°45')$. $\theta_X=0$ when the c axis of spodumene coincides with the magnetic field H_0.

plicable to larger values of e^2qQ/h in asymmetric fields are now in progress.

A complete account of this work is being submitted to the *Canadian Journal of Physics.*

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‡ Holder of a National Research Council Bursary. Now with the Dominion Gulf Company, Box 281, Larder Lake, Ontario.
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Angular Correlation of the Gamma-Rays of Cs^{134}*

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A PROGRAM has been initiated for the study of the correlations of cascade radiations[1] in the decay of radioisotopes. We make use of the proportional properties of scintillation crystals and of the high efficiency of NaI(Tl) as a γ-ray detector. We reduce the accidental counting rates considerably by using fast amplifiers and a fast coincidence circuit for timing, and slower, linear amplifiers in parallel, pulse-height selection channels. There are no heavy metals in the vicinity of the detecting crystal and no collimation is used. We rely only upon pulse-height selection to eliminate false coincidences due to scattered radiation.

Each photomultiplier output goes to two parallel channels. One consists of a fast amplifier and leads to a "fast" coincidence circuit whose resolving time is 30 μsec. The other channel is a linear amplifier and single channel pulse-height selector and leads

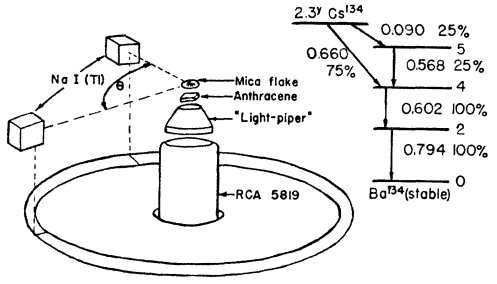

FIG. 1. Decay scheme of Cs^{134} and schematic sketch of detector system (β-ray detector exploded).

to a "pulse-height" coincidence circuit, whose resolving time is $\frac{3}{4}$ μsec. The pulse-height selectors are set to accept pulses corresponding to γ-ray energies appropriate to the problem. For instance, in experiments on Co^{60} and Cs^{134}, whose γ-rays are all of nearly the same energy, the pulse-height selectors are set to the peak of the Compton distribution. The "fast" and "pulse-height" coincidence signals are fed into still another coincidence circuit. An output signal in this last stage then indicates that there has been a coincidence of two signals of proper pulse height. All critical voltages are regulated, and the battery supplying the photomultipliers is monitored continuously.

The system was checked by measuring the angular correlation of the γ-rays of Co^{60}. The ratio $W(\pi)/W(\frac{1}{2}\pi)$ was found to be

TABLE I. Angular correlation of γ-rays coincident with 660-kev β-ray of Cs^{134}. Spins 4, 2, 0, and quadrupole radiations.

	$W(\pi/2)$	$W(3\pi/4)$	$W(\pi)$
Theoretical value	1	1.073	1.167
Theoretical value corrected for solid angle	1	1.063	1.139
Experimental value	1	1.085±0.03	1.109±0.03

TABLE II. Theoretical and experimental "over-all" correlation ratios for Cs^{134}.

Spin of 3rd state	Multipole character of 1st γ-ray	$W(\pi/2)$	$W(3\pi/4)$	$W(\pi)$
5	Dipole	1	1.036	1.074
5	Quadrupole	1	1.027	1.037
6	Quadrupole	1	1.063	1.139
Experimental values		1	1.046±0.01	1.106±0.01

1.17±0.05, in good agreement with the results of other experiments.[2]

With a slight modification of this system we have investigated the γ-ray cascade following the β-decay of the 2.3-year isomer of Cs^{134}. The decay scheme is shown in Fig. 1.[3] The source was deposited on a mica flake mounted upon a chip of anthracene (about 1 cm×1 cm×1.5 mm) as shown in Fig. 1. The output of the β-detector photomultiplier was amplified by one of the fast amplifiers and fed into the "fast" coincidence circuit. The 90-kev β-ray was entirely suppressed since it gave pulses too small to actuate the fast coincidence circuit. The anthracene was quite insensitive to γ-rays and the overall efficiency for the 660-kev β-ray was about $\frac{1}{3}$. Thus we required a coincidence of two γ-rays with the further condition that one of them be in coincidence with a 660-kev β-ray.

Some 4000 events were counted at each of three positions, (90°, 135°, 180°). The angular correlation of the γ-rays coincident with the 660-kev β-ray is in good agreement with the theoretical correlation function for transitions involving spins 4, 2, 0, and quadrupole radiations. The results are shown in Table I.

At the same time we measured the "over-all" correlation of all the γ-rays; that is, accepting any of the three γ-rays. The results are shown in Table II and are in agreement with the other observations.[2]

Assuming the spins 4, 2, 0, and quadrupole radiations, theoretical correlation functions were calculated[4] on the assumptions that the third excited state of Ba^{134} had spin 5 (dipole and quadrupole radiation) and spin 6 (quadrupole radiation.) Using the β-decay branching ratio of 3:1 and correcting for solid angle, we derive the results shown in Table II.

The spins of the excited states of Ba^{134} may then be assigned as indicated in Fig. 1. This assignment is in agreement with the character of the radiations as deduced from interval conversion coefficients.[5]

Work is in progress on other triple correlations.

We are indebted to Dr. M. E. Rose of the Oak Ridge National Laboratory for the calculation of the angular correlations of the first and third γ-rays with the second unobserved one.

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Anomalous Resistance of Noble Metals Containing Paramagnetic Ions

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EXPERIMENTS performed by Gerritsen and Linde[1] on diluted alloys of Mn in Cu, Ag, and Au reveal anomalies which are similar but more pronounced than those observed in

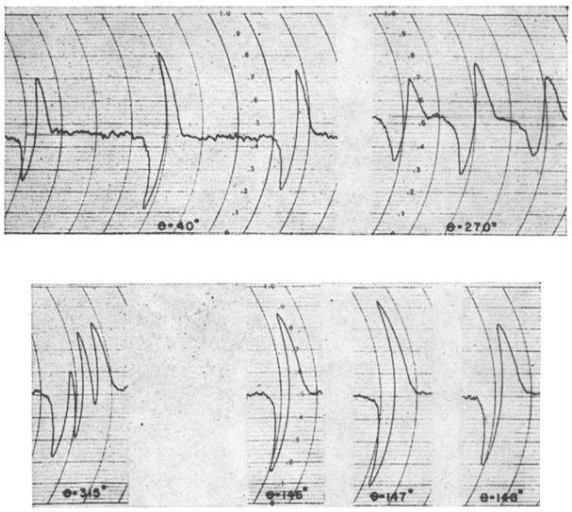

FIG. 1. Examples of recorded absorption derivative curves. The first three show selected splittings for the X rotation. The last three demonstrate the accuracy with which the position of zero splitting at $\theta_Y = 147°$ was determined for the Y rotation. A change of θ_Y by $\pm 1°$ produces a marked decrease in the maximum of the derivative curve. The sweep rate in all cases is 10 kc/sec per division.