Determination of the sign and magnitude of the nuclear quadrupole interaction by β-γ directional correlations

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Experiments are reported which demonstrate that the *sign* and magnitude of the quadrupole interaction of excited nuclear states can be reliably determined by means of time-differential β-γ directional correlations with radioactive sources embedded in noncubic single crystals. The 828 keV level of ^{115}In and the 247 keV level of ^{111}Cd, fed by an allowed and a unique first-forbidden β decay, respectively, have been investigated by this method. The coupling constants e^2qQ/h of these levels in Cd metal at 298 °K, are −146(5) MHz for ^{115}In and +125(4) MHz for ^{111}Cd. The positive coupling constant for ^{111}Cd in Cd coupled with a positive quadrupole moment Q of this level implies a positive electric field gradient in Cd confirming recent theoretical predictions. The negative e^2qQ for the ^{115}In level in Cd metal correspondingly indicates a negative Q for this level in conformity with its description as a member of a $K=\frac{1}{2}$ rotational band. The paper includes a discussion of the major concepts underlying the various methods available now for determination of the sign of e^2qQ. Also, a detailed account of theoretical formulas necessary to evaluate allowed and forbidden β-γ perturbed angular correlations and a brief discussion of the significance of the technique for further studies of nuclear quadrupole effects in materials are given.

[RADIOACTIVITY ^{115}Cd^m, ^{111}Ag; measured β-$\gamma(\theta)$ in Cd single crystal; deduced ^{115}In, ^{111}Cd levels e^2qQ and sign in Cd.]

I. INTRODUCTION

The interaction of the nuclear electric quadrupole moment eQ with the electric field gradient (EFG) eq in a crystalline surrounding is an effect widely studied by a number of techniques. One of the methods which, in recent years, is increasingly being applied to the measurement of the coupling constant e^2qQ is that of perturbed angular correlations (PAC) of nuclear cascade γ rays, performed in the time-differential mode on quadrupole moments of excited nuclear states. The points in favor of this technique are the relative ease with which the experiments can be carried out, its effectiveness in applications to extremely dilute foreign probe atoms in materials, and the fact that it allows convenient measurement on samples held at various temperatures as well as at high pressures. But γ-γ PAC experiments have nevertheless the drawback that they yield only the magnitude of e^2qQ and not its sign. In most cases the need to know the sign has been keenly felt. For example, in metallic systems the observed EFG arises primarily from two competing sources: the ion cores of the lattice and the conduction electrons. Since these two parts cannot be studied separately, the measurement of the sign of e^2qQ is important in determining the relative contributions of the two terms. For nuclear physics the sign of e^2qQ is necessary for obtaining the sign of Q, which is of decisive importance for comparison with nuclear models. It would therefore be very valuable to have a method with the virtues of a simple directional correlation measurement, which, in addition, is capable of yielding the sign of the quadrupole interaction (QI). It was known for a long time that, in principle, this is possible by performing a β-γ (instead of a γ-γ) directional correlation in which the consequences of parity nonconservation in nuclear β decay play a crucial role. Such an experiment was recently demonstrated for the first time in this laboratory, employing allowed as well as first-forbidden β decay.[1,2] This paper is a detailed report of these experiments.

To put in perspective the general problem of the determination of the sign of e^2qQ, we discuss in Sec. II the major physical concepts underlying the various experiments designed for this purpose. In Sec. III theoretical β-γ PAC formulas are presented for allowed and first-forbidden β decay and applied to the cases of ^{115}Cd → ^{115}In and ^{111}Ag → ^{111}Cd which were studied in the experiments. Section IV contains details of the experiments and the results. Discussions and conclusions are given in Sec. V.

II. GENERAL APPROACHES FOR DETERMINING THE SIGN OF e^2qQ

Under the action of the QI, a nuclear level of spin I splits into a set of $I+1$ (integer I) or $I+\frac{1}{2}$ (half integer I) unequally spaced m sublevels. The sublevels labeled $+m$ and $-m$ remain degenerate. Complete information on the QI is obtained when (a) one or more of the energy intervals between the split levels is measured, leading to the *magnitude* of the coupling constant e^2qQ, and (b) the level order is also determined, which leads to the *sign* of e^2qQ. The experiments designed to yield QI information (or for that matter any hyperfine interaction) fall into two broad categories: (A) "Energy" methods where full or partial information on the energies of the quadrupole split levels is directly involved in the measurement process. In these methods the m-state eigenfunctions can be considered as time-independent states. (B) "Phase" or "precession" methods in which no direct energy measurement is involved. The m-state eigenfunctions are time-dependent and information is extracted from interference effects produced by the coherent time evolution of the m-state vectors, i.e., precession of the nuclear spin and its effect on radiation emission patterns.

A. Energy methods

Consider an excited Mössbauer level, split by the QI, decaying to the ground state which is also, in general, split. The extreme energy resolution of the Mössbauer effect resolves the individual transitions between sets of m levels of the excited and ground states. The energies of these transitions are measured by means of Doppler shifting the (unsplit) γ ray from the source which serves as a primary energy standard. Thus, when there are a large number of transitions, one can determine not only the energy intervals, but the level order itself, and in many cases, the QI constants and their signs and even the ratios of the excited and ground state moments have been determined. There are ambiguities present in special cases, e.g., in ^{57}Fe where the excited state (spin $I=\frac{3}{2}$) splits symmetrically and the ground state ($I=\frac{1}{2}$, $Q=0$) is unsplit. In such instances one has to resort to the use of single crystals and the change in the absorption intensity due to the angular distribution of the two transition radiations ($\Delta m=0$ and $\Delta m=\pm 1$) to decide the sign. As will be seen below, this is equivalent to a measurement on cryogenically aligned nuclei and it can still be categorized as an energy method.

For stable nuclei, an interesting method was reported by Gregers-Hansen, Krusius, and Prickett,[3] who measured the nuclear specific heat of Re metal at very low temperatures. In this region the Schottky anomaly in the heat capacity arising from the QI can be written as[3]

$$C_Q = a(I)\left(\frac{e^2qQ}{k_B}\right)^2 T^{-2} + b(I)\left(\frac{e^2qQ}{k_B}\right)^3 T^{-3},$$

where $a(I)$ and $b(I)$ are constants depending on the nuclear spin, k_B the Boltzmann constant, and T the temperature. The presence of the second term with the odd power in e^2qQ which is sign sensitive was established by these authors at very low temperatures where it becomes significant. This is thus a measurement of the distribution of energy in the m levels which becomes sensitive to the level ordering (and hence the sign of e^2qQ) due to the nonuniform nature of the quadrupole splitting. Elegant as it is, the method is not generally useful since the QI in most cases is small and millikelvin temperatures are required to observe the effect.

Consider now a radioactive nucleus imbedded in a noncubic single crystal. At room temperature the populations of the m levels split by the QI are equal and the nucleus radiates isotropically. When, however, the temperature is low enough so that k_BT is small compared to the splitting, the population $P(m)$ of the *energetically* lowest degenerate doublet of levels increases at the expense of the higher doublets with different values of $|m|$. In this condition, where $P(m)=P(-m)\neq P(|m'|)$, the nucleus is said to be *aligned*, a necessary and sufficient prerequisite for the nucleus to radiate anisotropically with respect to the axis of alignment (here the symmetry axis of the single crystal). The exact nature of the directional anisotropy of γ rays now depends on the value of $|m|$ of the lowest level which can be $|m|=I$ on the one hand and $|m|=\frac{1}{2}$ or 0 on the other, depending on the sign of e^2qQ. Thus the observation of γ-ray anisotropy of nuclei aligned cryogenically by the QI yields the sign of the QI. The key concept here is energy, since the Boltzmann distribution, with absolute temperature as a standard of energy, forces the nuclear quadrupoles into an orientation with respect to the field axis which has the lowest *energy*. The identification of this state is then provided by the γ-ray anisotropy but in principle this can be done even by performing nuclear quadrupole resonance (NQR) in the aligned state.[4] The strengths of the quadrupole resonances and their temperature dependence can reveal the nature of the nonuniform level ordering. The NQR method has not been applied so far, but the γ-ray anisotropy method has been demonstrated in a few cases.[5] Again, very low temperatures are required. Mössbauer measurements with single crystals, mentioned before, are based on this scheme. In this instance, however, no net alignment is needed to force most nuclei into a particular state, since the individual states are separately observed by means of the energy resolution of the recoilless transitions.

B. Phase methods

We now turn to precession methods in which nuclear orientation[6] is produced neither by cryogenics nor by direct observation of the m-level transitions. In the emission of cascade radiations in nuclear decay the observation of the emission direction of the first radiation selects an ensemble of nuclei in the intermediate state with definite spin orientations, a process which is equivalent to nuclear orientation. In parity conserving processes such as γ-ray emission this orientation is invariably in the form of *alignment,* because the two possible circular polarization states of the photon are not distinguished and the ensemble of nuclear states is left with $P(m)=P(-m)$. The alignment can be detected by the directional anisotropy of the coincident second γ ray of the cascade, with respect to the direction of the first. Note that this angular correlation of γ rays is present even in the absence of any interaction which produces a level splitting. Thus γ-ray anisotropies due to alignment created by previous γ-ray emission differ fundamentally from anisotropies seen in alignment produced cryogenically or in Mössbauer absorption experiments in that considerations of energy such as the strength of level splitting or the level ordering do not enter. The only criteria which dictate the observable correlation are the *angular momenta* involved, such as level spins and γ-ray multipolarities.

Let us now switch on a QI, which induces a precession of the nuclear spin about the symmetry axis of the interaction, e.g., the c axis of the crystal. If this axis coincides with the axis of alignment (i.e., the first γ-ray direction) the m-state vectors are stationary states since the interaction in this representation is diagonal. Thus the initial alignment is also stationary and the QI has no effect on the anisotropy. However, if the c axis is not collinear with γ_1, the m-state vectors are no longer stationary and they acquire a time-dependent phase factor which contains the quadrupole coupling frequency $\omega_0 \propto e^2qQ$. The coherent superposition (dictated by the lack of energy resolution of the detection process) of the m-state vectors with their complex phase factors results in an interference process (quantum beats) producing a modulation of the initial alignment with the frequency ω_0. Observation of such modulations is the basis of all precession experiments. It is worth noting that these experiments require *two* noncollinear axes, one for initial alignment and one for the interaction, unlike energy methods which need only one axis for defining the experiment.

The modulation affects the initial alignment in two ways: First, a portion of the alignment becomes oscillatory in time; and second, a portion of the initial alignment is converted into *polarization* $[P(m)\neq P(-m)]$ which is also oscillatory. The second effect is unique to the QI, in contrast to magnetic interactions which create *only* the first effect. The superposition of the phase factors resulting in polarization reflects the fact that the nuclear quadrupole precesses in opposite directions in the $+m$ and $-m$ states. The phases contributing to the oscillating *alignment* superpose in the form $(e^{in\omega_0t}+e^{-in\omega_0t})$ which oscillates like cos $n\omega_0t$, while those contributing to the oscillatory *polarization* combine in the form $(e^{in\omega_0t}-e^{-in\omega_0t})$, which oscillates like $\sin n\omega_0t$. Thus the alignment part is insensitive to the sign of the QI frequency ω_0; only the polarization part is sensitive to it. If now the directional distribution of γ_2 is measured (γ-γ PAC), a process which is sensitive only to the alignment part, the γ-γ anisotropy exhibits oscillations of the $\cos n\omega_0t$ type which yield the magnitude but not the sign of the coupling constant. Such oscillations have been observed in a number of nuclei. To derive complete QI information, including the sign, one needs a measurement sensitive to the *polarization* part, e.g., detection of the circular polarization of γ_2. It must be added here that such a measurement should be done with a single crystal so that the c axis is not distributed equally in all directions with respect to γ_1, which would cancel the net polarization to zero.

It is possible to show that quadrupole spin precession is characterized by two general effects[7]: 1. The precession mixes orientation of odd (polarization) and even (alignment) type; i.e., if initially pure polarization is present, after precession polarization as well as alignment can be observed, and *vice versa.* The latter case was discussed above. 2. The component of orientation whose odd-even nature is unchanged by the precession oscillates like $\cos n\omega_0t$ and is insensitive to the sign of ω_0. The opposite parity orientation oscillates like $\sin n\omega_0t$ and is sensitive to the sign of ω_0. Experiments to observe the sine modulations can be devised in several ways and Table I summarizes the schemes realized so far. In the first two,[8,9] polarization of the precessing level is detected either by observing the circular polarization of the deexciting γ ray or the asymmetry of the β emission in the case of unstable ground states. Measurement of the γ-ray circular polarization is tedious and inefficient. Thus, for excited nuclear

TABLE I. Experimental schemes for observation of odd parity modulations due to quadrupole spin precession.

Initial orientation	Produced by	Post-precession orientation	Detected by	Ref.
Alignment (even)	γ decay; Observation of γ_1 direction.	Polarization (odd)	Coincident γ_2 circular polarization	8
Alignment (even)	Recoil after (d,p) reaction	Polarization (odd)	β asymmetry	9
Polarization (odd)	Coulomb excitation sideways projectile scattering	Alignment (even)	Coincident γ_2 directional anistropy	11 12
Polarization (odd)	β decay; observation of β direction	Alignment (even)	Coincident γ directional anistropy	1 2

states, methods 3 and 4, where alignment has to be detected, are more attractive since this can be done simply by measuring the directional anisotropy of γ rays. These experiments require the production of polarization by the first radiation which can be achieved in many ways. For example, method 3 employs the fact that in a nuclear reaction process like Coulomb excitation polarization of the excited nucleus results if the projectile is scattered sideways.[10-12] The excited nucleus can be simultaneously implanted with the aid of the recoil energy of the reaction into a noncubic single crystal where it is subject to the quadrupole precession. The resulting alignment can be detected by the anisotropy of the deexcitation γ ray observed in coincidence with the scattered projectiles. Alternatively, the present work makes use of the fact that because of parity nonconversation in nuclear β decay, the observation of the β-particle direction selects an ensemble of polarized nuclei. This leads directly to the realization that β-γ directional correlations perturbed by the nuclear quadrupole coupling in a noncubic single crystal are capable of yielding complete QI information. The enormous simplicity brought about by the fact that only a *directional* correlation is required certainly sets this apart from most of the methods discussed above as a reliable and unambiguous laboratory tool for the study of the quadrupole interaction.

III. THEORY

The theoretical basis for β-γ directional correlations perturbed by the nuclear quadrupole interaction was first derived by Harris[13] for the case of allowed β decay. We give here an outline of this theory in the more generally accepted notation of Frauenfelder and Steffen[14] and include the case of first-forbidden β decay for application to our experiments on $^{111}Ag \rightarrow {}^{111}Cd$. We start with the general expression for the angular correlation function for a β-γ cascade connecting three nuclear levels with the spin sequence $I_1 \xrightarrow{\beta} I \xrightarrow{\gamma} I_2$, including the perturbation due to the QI in the intermediate state:

$$W(\vec{k}_1,\vec{k}_2,t)=\sum_{\substack{k_1,k_2\\N_1,N_2}}\frac{(-1)^{k_1+k_2}A_{k_1}(\beta)A_{k_2}(\gamma)}{[(2k_1+1)(2k_2+1)]^{1/2}}G_{k_1k_2}^{N_1N_2}(t)Y_{k_1}^{N_1*}(\theta_1,\varphi_1)Y_{k_2}^{N_2}(\theta_2,\varphi_2)\,. \quad (1)$$

The angles θ_1,φ_1 and θ_2,φ_2 are the polar and azimuthal angles of the β and γ emission directions, respectively, with respect to the axis of quantization z (see Fig. 1). The Y_k's are spherical harmonics defining the angular part of the PAC function. The angular correlation coefficients $A_{k_2}(\gamma)$ have the usual definition:

$$A_{k_2}(\gamma)=[F_{k_2}(LLI_2I)+2\delta F_{k_2}(LL'I_2I)+\delta^2F_{k_2}(L'L'I_2I)]/(1+\delta^2)\,, \quad (2)$$

where L and L' are the contributing multipoles whose amplitudes are mixed in the ratio δ. To estimate the magnitude and sign of the observed correlation it is necessary that δ and its sign be known reliably. This information is, in general, available for most γ transitions of interest. $A_{k_1}(\beta)$ is discussed below in detail. The indices k_1, k_2, N_1, and N_2 are all integers, with N taking all integer values $-k \leq N \leq +k$. Since only the direction of the γ ray is measured, averaging over both circular polarization states of the photon results in k_2 being restricted to the even integers 0, 2, 4, etc. k_1 takes all values 0, 1, 2, 3..., since the odd terms arise due to the nuclear polarization after β decay. As we shall see, they eventually contribute to the γ-ray directional correlation due to the quadrupole precession. The number of terms in (1) is determined by the limits on k_1 and k_2. The properties of F coefficients (Eq. 2) imply that $k_{2\,\max}^{(\mathrm{even})} \leq$ minimum of the numbers $2I$, $2L'$. The limit on k_1 depends on the type of β decay (see below).

The form of Eq. (1) as given here differs from that found in Ref. 14 by the phase factor $(-1)^{k_1+k_2}$. The factor $(-1)^{k_2}$ should normally be present in the general angular correlation formula, in the framework of Ref. 14, and in addition $(-1)^{k_1}$ is required in order to be consistent with the definitions of β-particle parameters used in Ref. 14. The $(-1)^{k_1+k_2}$ factor was irrelevant for most previously performed experiments, since these were not sensitive to terms with (k_1+k_2) odd. Experiments discussed here, however, aim to observe just these terms, and therefore the angular correlation expression has to be derived in a completely consistent scheme where the phases are carefully accounted for.

The effect of the perturbing field is contained in $G(t)$, the so-called perturbation factor. For an axially symmetric EFG whose symmetry axis (e.g., the c axis of a hexagonal crystal) is along the z axis, the QI is diagonal in this representation and results in $N_1 = N_2$. With this, it can be shown that

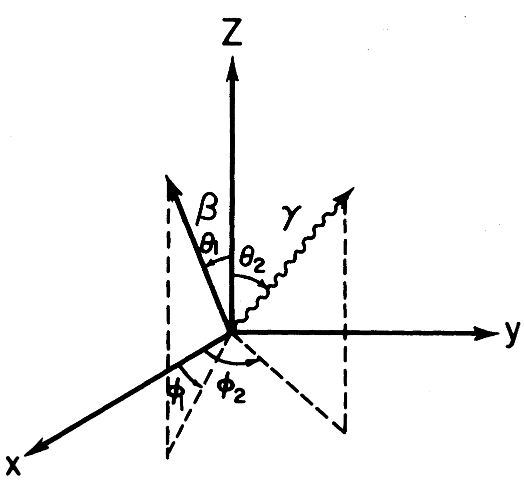

FIG. 1. Definition of polar and azimuthal angles of emission of the β particle and the γ ray with respect to a coordinate system whose z axis serves as the quantization axis.

$$G_{k_1k_2}^{NN}(t) = \sum_{n\geq 0} S_{nN}^{k_1k_2} \times \begin{cases} \cos n\omega_0 t & \text{for } (k_1+k_2) \text{ even} \\ (-i)\sin n\omega_0 t & \text{for } (k_1+k_2) \text{ odd}, \end{cases} \tag{3}$$

where ω_0 is the basic QI frequency defined by

$$\begin{aligned} \omega_0 &= 3\omega_Q(\text{integer } I), \\ &= 6\omega_Q(\text{half-integer } I), \end{aligned} \tag{4}$$

and

$$\omega_Q = e^2qQ/4I(2I-1)\hbar,$$

where we define the EFG

$$eq = \frac{\partial^2 V}{\partial z^2},$$

V being the electrostatic potential. The coefficients $S_{nN}^{k_1k_2}$ have the general properties

$$S_{n0}^{k_1k_2} = \delta_{n0}\delta_{k_1k_2}, \tag{5a}$$

$$S_{nN}^{k_1k_2} = (-1)^{k_1+k_2} S_{n-N}^{k_1k_2}, \tag{5b}$$

$$S_{nN}^{k_1k_2} = S_{nN}^{k_2k_1}, \tag{5c}$$

and have been listed in Ref. (13) with various values of k_1, k_2, and N for the permissible values of n for each I.

The main feature of quadrupole precession effects in a β-γ correlation is exhibited in Eq. (3). This shows that if odd values of k_1 are allowed by the nature of the first radiation (equivalent to nonzero residual nuclear polarization), then, in general, one admits terms in the expansion of (1) with (k_1+k_2) odd (k_2 is always even since a γ-ray circular polarization is not measured). These terms modulate the unperturbed correlation in the form of a *sine* oscillation which is sensitive to the *sign* of the precession frequency ω_0. One sees that at $t=0$ (or in the field-free case), the odd terms are zero, and since

$$\begin{aligned} G_{k_1k_2}^{NN} &= \sum_{n\geq 0} S_{nN}^{k_1k_2} \quad (k_1+k_2) \text{ even} \\ &= \delta_{k_1k_2}, \end{aligned} \tag{6}$$

one recovers the full unperturbed correlation. The polarization sensitive odd terms are thus introduced into the correlation only at later times as the quadrupole spin precession progresses. We now consider the $A_{k_1}(\beta)$ factors and the form of $W(t)$ for the specific cascades used in this work.

a. Allowed β decay. β transitions between levels with spin difference $\Delta I = 0, 1$ and with no parity change are characterized as allowed transitions. In the "allowed" approximation of β-decay theory, the leptons do not carry any orbital angular momentum and in general, e.g. for a $\Delta I = 0$ $(I \rightarrow I)$ transition, there are two components, one with electron and neutrino having parallel spins ($J=1$) and one with antiparallel spins ($J=0$). The $J=1$ component is termed Gamow-Teller (G-T) and $J=0$ is termed Fermi (F). Thus, as in multipole mixing occurring in γ transitions, for a $\Delta I=0$ allowed β transition, there is G-T and F mixing with a ratio $y=$G-T/F which must be known to calculate $A_{k_1}(\beta)$. For a pure G-T transition ($\Delta I=1$), only the $J=1$ component contributes. The present case of $^{115}\mathrm{Cd} \xrightarrow{\beta^-} {}^{115}\mathrm{In}$ is such a transition.

The value of k_1, in allowed G-T transitions, is limited to the values 0 and 1 only.[14] Since k_2 takes only the values 0, 2 etc., and $k_1=k_2$ for the field-free case (Eq. 6), only the term $k_1=0$ and $k_2=0$ contribute to $W(t)$, leading to the well-known result that the allowed β-γ unperturbed correlation is *isotropic*. In the presence of the QI, by (3), terms with (k_1+k_2) odd contribute, therefore the terms $k_1=1$ and $k_2=2,4$ appear and the β-γ *perturbed* angular correlation is *anisotropic*, the anisotropy being induced solely through the agency of the quadrupole precession.

The correlation $W(t)$ is especially simple if the γ ray involved is of dipole ($L=1$) character, resulting only in the term $k_1=1$, $k_2=2$ appearing in $W(t)$ which reduces (1) to

$$W(t)=\frac{1}{4\pi}-\frac{A_1(\beta)A_2(\gamma)}{\sqrt{15}}G_{12}^{11}(t)Y_1^{1*}(\theta_1\varphi_1)Y_2^1(\theta_2\varphi_2)\,. \tag{7}$$

Using Eq. (3) and (5b), normalizing by 4π, and expanding the spherical harmonics,

$$W(t)=1-\tfrac{1}{2}\sqrt{3}\,A_1(\beta)A_2(\gamma)\sin\theta_1\sin 2\theta_2\sin(\varphi_2-\varphi_1)\times\Big[\sum_n S_{n1}^{12}\sin n\omega_0 t\Big]. \tag{8}$$

γ rays with multipole order higher than dipole result in an additional term in (8) with $k_1=1$ and $k_2=4$, but for the application to the present work Eq. (8) is sufficient. $A_1(\beta)$ is defined for a pure G-T

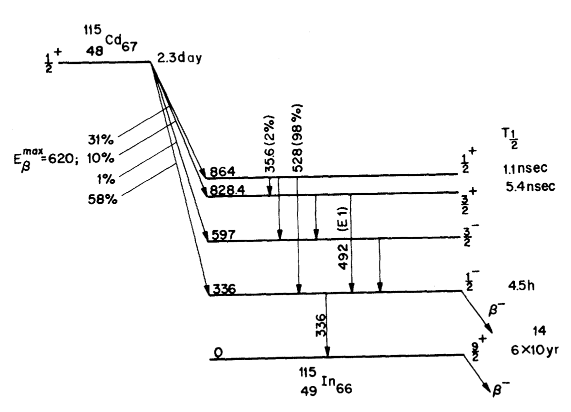

FIG. 2. Decay scheme of $^{115}\mathrm{Cd}\rightarrow{}^{115}\mathrm{In}$.

decay ($J=1$) as

$$A_1(\beta^{\mp})=\pm\tfrac{2}{3}F_1(11I_1I)v/c\,, \tag{9}$$

reflecting the fact that the residual nuclear polarization is proportional to the velocity v of the $\beta^{\mp}$ particle observed in coincidence with the γ ray.

Consider the β^- decay of ^{115}Cd to the 828 keV level of ^{115}In (see Fig. 2 for decay scheme)[15] with a spin of $I=\frac{3}{2}^+$, decaying by the emission of the 492 keV γ ray which is $E1$. The spin sequence of this β-γ cascade is

$$\tfrac{1}{2}^+ \xrightarrow{\beta^-(\mathrm{G-T})} \tfrac{3}{2}^+ \xrightarrow{\gamma(E1)} \tfrac{1}{2}^-\,.$$

From the tables of F coefficients (Ref. 14), Eqs. (9) and (2), $A_1(\beta)=-\frac{1}{3}\sqrt{5}v/c$ and $A_2(\gamma)=\frac{1}{2}$. For $I=\frac{3}{2}$, only S_{n1}^{12} is needed with $n=1$; $S_{11}^{12}=(\frac{3}{5})^{1/2}$. Thus, we have

$$W(k_1,k_2,t)=1+\tfrac{1}{4}(v/c)\sin\theta_1\sin 2\theta_2\times\sin(\varphi_2-\varphi_1)\sin\omega_0 t\,. \tag{10}$$

The effect is maximized if we choose the geometry

$$\theta_1=\tfrac{1}{2}\pi,\quad \theta_2=\tfrac{1}{4}\pi,\quad\text{and}\quad(\varphi_2-\varphi_1)=\varphi=\pm\tfrac{1}{2}\pi\,, \tag{11}$$

which yields the very simple result

$$W(\pm,t)=1\pm\tfrac{1}{4}(v/c)\sin\omega_0 t\;(\varphi=\pm\tfrac{1}{2}\pi)\,. \tag{12}$$

Experimentally, the β-γ time-differential coincidence spectrum has the form

$$C(\pm,t)=W(\pm,t)e^{-t/\tau}+K\,, \tag{13}$$

where τ is the mean life of the interacting level and K accounts for random coincidences. It is convenient to form the ratio $A(t)$, the normalized reflection asymmetry of the coincidences at $\varphi=\pm\frac{1}{2}\pi$, defined by

$$A(t)=\frac{2[C(-)-C(+)]}{[C(-)+C(+)-2K]}\,. \tag{14}$$

Insertion of (13) and (12) gives

$$A(t)=-(v/2c)\sin\omega_0 t\,. \tag{15}$$

The reflection asymmetry, in this case, is thus a pure sinusoidal oscillation, the polarity of which yields the sign of ω_0.

b. Forbidden β-decay. β transitions with $\Delta I=2$, 1, or 0 with change of parity are classified as first-forbidden transitions. In general there are three components with total angular momentum $J=0$, 1, or 2. In the cases where all these components are allowed, e.g., a $\Delta I=0$, $I\rightarrow I$ transition, there are interferences between these components with not just one but several mixing ratios. In addition, within each component there could be further interferences from several matrix elements contributing to that component. Thus, as a rule, it is impossible to predict $A_{k_1}(\beta)$ for forbidden transitions which makes them rather unattractive as a tool to study quadrupole effects. However, as in allowed β decay, the type of transition with maximum allowed spin change $\Delta I=2$, called a *unique* first-forbidden transition, offers a case in which only the component $J=2$ can contribute and, since $\Delta I=2$, only the tensor type matrix element drives this transition. No interferences are then possible. This is thus an example of a "pure" forbidden β decay analagous to "pure" G-T allowed decay and pure γ transitions. $A_{k_1}(\beta)$ can therefore be written down as usual in terms of the F coefficients.

The possible values of k_1 for the unique first-forbidden (UFF) case[14] are 0, 1, 2, and 3, and if the γ transition of the cascade has quadrupole character, k_2 ranges through 0, 2, and 4. This is thus considerably more complicated, allowing as many as five odd and two even oscillatory terms in the expansion of (1). Notice also, that (k_1+k_2) even terms are present which represent an anisotropic unperturbed correlation in contrast to the pure G-T case discussed above which was isotropic. The $A_{k_1}(\beta)$ factors analogous to Eq. (9) are[14]:

$$A_0(\beta^\mp)=1\,,$$
$$A_2(\beta^\mp)=f_2(W)F_2(22I_1I)\,, \tag{16a}$$

$$A_1(\beta^\mp)=\pm f_1(W)F_1(22I_1I)v/c\,,$$
$$A_3(\beta^\mp)=\pm f_3(W)F_3(22I_1I)v/c\,, \tag{16b}$$

where f_1, f_2, and f_3 are functions of the relativistic energy W of the β particle [not to be confused with the correlation function $W(t)$] and can be obtained from Ref. (14).

Owing to the fact that several terms are present in the expansion of (1), care must be given to including the effect of finite solid angle of the detectors of the radiation which affects the different terms to different extents. This could, in principle, have significant effects in terms of the frequency composition of the observed time-differential correlation. Geometrical effects are accounted for by insertion of the factor $[Q_{k_1}(\beta)Q_{k_2}(\gamma)]$ in (1), where

$$Q_{k_1}(\beta)=\int_0^\alpha P_{k_1}(\cos\theta)d(\cos\theta)\bigg/\int_0^\alpha d(\cos\theta)\,. \tag{17}$$

Here α is the half-angle subtended by the detector at the source and $P_k(\cos\theta)$ is the Legendre polynomial of order k. This expression is useful for the $Q_{k_1}(\beta)$ part, while for the γ part the $Q_{k_2}(\gamma)$ are found, e.g., in Ref. (14) for NaI(T1) detectors. With these factors one can now write out the complete expression of Eq. (1) for the experimental geometry of Eq. (11):

$$\begin{aligned}W(\pm,t)=1&-\tfrac{1}{8}Q_2(\beta)A_2(\beta)Q_2(\gamma)A_2(\gamma)-\tfrac{3}{8}Q_2(\beta)A_2(\beta)Q_2(\gamma)A_2(\gamma)\sum S^{22}_{n2}\cos n\omega_0t\\&-\tfrac{15}{32}\sqrt{\tfrac{5}{3}}\,Q_2(\beta)A_2(\beta)Q_4(\gamma)A_4(\gamma)\sum S^{24}_{n2}\cos n\omega_0t\mp\tfrac{1}{2}\sqrt{3}\,Q_1(\beta)A_1(\beta)Q_2(\gamma)A_2(\gamma)\sum S^{12}_{n1}\sin n\omega_0t\\&\mp\tfrac{1}{16}\sqrt{10}\,Q_1(\beta)A_1(\beta)Q_4(\gamma)A_4(\gamma)\sum S^{14}_{n1}\sin n\omega_0t\pm\tfrac{3}{4}(1/\sqrt{2})Q_3(\beta)A_3(\beta)Q_2(\gamma)A_2(\gamma)\sum S^{32}_{n1}\sin n\omega_0t\\&\pm\tfrac{1}{32}\sqrt{15}\,Q_3(\beta)A_3(\beta)Q_4(\gamma)A_4(\gamma)\sum S^{34}_{n1}\sin n\omega_0t\pm\tfrac{35}{32}(1/\sqrt{7})Q_3(\beta)A_3(\beta)Q_4(\gamma)A_4(\gamma)\sum S^{34}_{n3}\sin n\omega_0t\,,\end{aligned} \tag{18}$$

where $\pm$ refers to $\varphi=\pm\frac{1}{2}\pi$.

The above equation is sufficiently general to be applied to any β-γ cascade that might be encountered, since the expansion has been carried out to $k_1(\max)=3$ and $k_2(\max)=4$. It is unlikely that these maxima will be exceeded for practical cases. We can now consider the unique first-forbidden β decay of ^{111}Ag (see decay scheme[16] in Fig. 3) to the well-known 247 keV level of ^{111}Cd. The spin sequence of this (β-γ) cascade is

$$\tfrac{1}{2}^-\xrightarrow{\beta^-(\mathrm{UFF})}\tfrac{5}{2}^+\xrightarrow{\gamma(E2)}\tfrac{1}{2}^+\,,$$

for which Eq. (18) applies in its entirety. The relevant values of $S^{k_1k_2}_{nN}$ for $I=\frac{5}{2}$, Q_k for the geometry used in this work, and $A_{k_1}(\beta)$ are given in Tables II and III. The values of $A_{k_1}(\beta)$ contain the $f(W)$ factors, but to obtain an idea of the form of the expected correlation these factors have been evaluated at the end point of the β spectrum where $f_1(W_0)=\frac{3}{5}$, $f_2(W_0)=\frac{7}{5}$, and $f_3(W_0)=\frac{3}{5}$. These data reduce Eq. (18) to the simple approximate form

$$\begin{aligned}W(\pm,t)\approx 1&\mp0.18\frac{v}{c}\sin2\omega_0t\mp0.08\frac{v}{c}\sin3\omega_0t\\&-0.10\cos3\omega_0t\quad\text{for }\varphi=\pm\tfrac{1}{2}\pi\,,\end{aligned} \tag{19}$$

where small terms with amplitudes of only a few percent have been neglected. The amplitudes in (19) are maximum limits. In practice the energy dependence of the $f(W)$ factors will reduce these coefficients, but not enough to affect their signs. We can then write down, as before, $A(t)$, defined in (14), for this case:

$$A(t)\approx\frac{0.36(v/c)\sin2\omega_0t+0.16(v/c)\sin2\omega_0t}{1-0.1\cos3\omega_0t}\,. \tag{20}$$

The reflection asymmetry is thus dominantly the

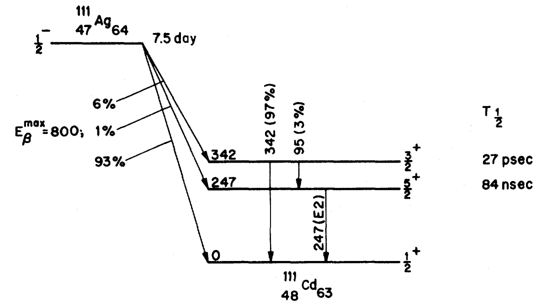

FIG. 3. Decay scheme of $^{111}Ag \rightarrow {}^{111}Cd$.

additive superposition of two positive sine oscillations. The sign of ω_0 is therefore determined directly by the initial polarity of the observed $A(t)$ spectrum.

IV. EXPERIMENTAL DETAILS AND RESULTS

A. ^{115}In in Cd metal

The source of ^{115}Cd ($T_{1/2} = 54$ h) was prepared by neutron irradiation of natural Cd for periods of 10–15 min at a flux of $\sim 10^{13}\ n/\mathrm{cm}^2\,\mathrm{sec}$. A single crystal of Cd metal in the form a disk (50 mg/cm^2 thick, 10 mm diam) with the c axis of the crystal in the plane of the disk, was employed. The irradiated source was used in the experiments after 1 day, without any further treatment. The geometry of (11) was realized in practice, as illustrated in Fig. (4) with the plane of the crystal set facing the β detector, and with the γ detector seeing the crystal edgewise. The crystal was oriented so that the c axis was 45° to the γ detector. The reflection asymmetry was measured by recording the β-γ delayed coincidence spectrum with the β detector at the two mirror image positions across the $[\hat{c}, \gamma]$ plane, which corresponds to $\varphi = \pm \frac{1}{2}\pi$ (see Fig. 4). Since the interest in the present experiments was the sign measurement, it was estimated that the reduction in modulation amplitude due to multiple and backscattering of the electrons in the source as well as in the airspace between the source and the β detector was within tolerable levels as far as clear observation of the modulations was concerned. Thus no efforts were made to enclose the electron flight path in vacuum or to use extremely thin source crystals.

TABLE II. Values of $S_{nN}^{k_1k_2}$ for $I = \frac{5}{2}$.

S	n 1	2	3
S_{n2}^{22}	0.643	⋯	0.357
S_{n2}^{24}	−0.479	⋯	0.479
S_{n1}^{12}	0.361	0.452	⋯
S_{n1}^{14}	−0.571	0.286	⋯
S_{n1}^{32}	−0.138	0.690	⋯
S_{n1}^{34}	0.218	0.436	⋯
S_{n3}^{34}	⋯	⋯	0.745

TABLE III. Angular correlation parameters for the β-γ cascade $\frac{1}{2}^- \xrightarrow{\beta\,\mathrm{UFF}} \frac{5}{2}^+ \xrightarrow{\gamma E2} \frac{1}{2}^+$. The geometrical factors Q_k correspond to the actual experiment and are typical values.

A, Q	k 1	2	3	4
$A_k(\beta)$	−0.410	−0.748	+0.657	0
$A_k(\gamma)$	0	−0.535	0	−0.617
$Q_k(\beta)$	0.95	0.85	0.71	⋯
$Q_k(\gamma)$	⋯	0.90	⋯	0.71

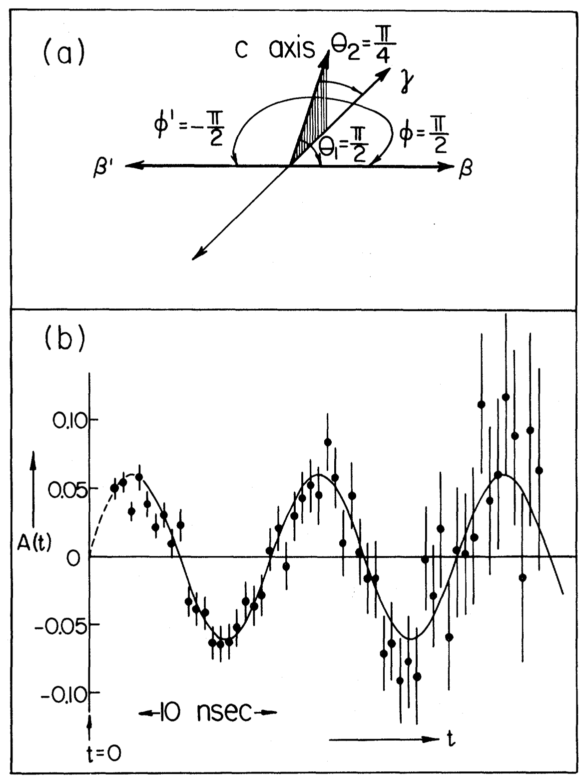

FIG. 4. (a) Experimental configuration used for β-γ perturbed directional correlation experiment in the case of $^{115}Cd \rightarrow {}^{115}In$. Note that the position of the β detector β' is the mirror reflection of the position β through the plane containing the c axis of the Cd crystal and the γ detector. (b) Time dependence of the reflection asymmetry $A(t)$ of β-γ delayed coincidences between the angles $\varphi = \pm\frac{1}{2}\pi$ of the β counter, displaying the nuclear quadrupole spin precession of the 828 keV level of ^{115}In in Cd metal.

As seen in Fig. 2 the β decay of ^{115}Cd to the 828 keV level of ^{115}In has a 10% branching and an end-point energy of $E_{\beta_{max}} = 620$ keV. An anthracene crystal was used to detect β rays between 250–400 keV, the gate being chosen to avoid too low energies (which have small polarization) and on the higher energy side, to minimize chance coincidences by inclusion of the more intense and more energetic β branch to the 335 keV level. The average $\langle v/c \rangle$ in the gate was about 0.7. The 828 keV level deexcites ($T_{1/2} = 5.5$ nsec) mainly by the emission of a 492 keV γ ray which was detected by a NaI(Tl) crystal. The γ-ray gate was chosen to minimize the unavoidable admixture of a 528 keV γ ray from the 860 keV level ($T_{1/2} = 1.0$ nsec) which produces coincidences with the β gate chosen. These coincidences, however, are not serious since the admixture decays much faster than the 5.5 nsec half-life of the 828 keV level and the interference is significant only near $t = 0$ of the observed time spectrum. More subtle is the unavoidable interference of the β-(35γ)-492γ coincidences proceeding through the 860 keV level via the 35 keV unobserved γ ray which are indistinguishable in time structure from the (β-492γ) coincidences. They also have, in principle, a different correlation. Fortunately this admixture is very small owing to the minimal branching of the 35 keV γ ray at the 860 keV level.

The time spectra of β-γ coincidences for the two positions of the β detector were recorded separately using standard slow-fast coincidence counting electronics. The ratio $A(t)$ computed from this data is shown in the lower half of Fig. (4). The solid line is a least squares fit to the data points using (15), the points close to $t = 0$ being omitted due to interference from the (β-528 keV γ) coincidences. Comparison of the observed positive sine form of $A(t)$ with (15) clearly shows that ω_0 is *negative* and the result of the fit gives $e^2qQ/h = -146(5)$ MHz, the magnitude being in excellent agreement with recent γ-γ PAC results[17] for this case. The amplitude of the observed $A(t)$ is much less than predicted by (15), the reduction coming from less than maximum polarization due to integrated observation of β particles over a range of energies, multiple and backscattering of β rays in the source, and finite solid angles of the detectors. The most significant of these is believed to be backscattering in the relatively thick source used in the experiment. This is in principle no serious limitation if the determination of sign and the magnitude of e^2qQ is the main object of a time-differential experiment of this kind.

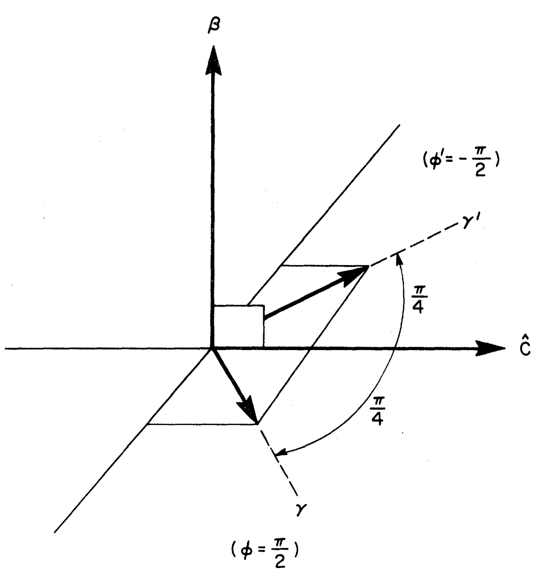

FIG. 5. Experimental configuration used for the β-γ perturbed directional correlation experiment in the case of ^{111}Ag-^{111}Cd.

B. ^{111}Cd in Cd metal

The parent ^{111}Ag activity was produced by the reaction ^{110}Pd$(n,\gamma)^{111}$Pd, ^{111}Pd$^m \xrightarrow{\beta} {}^{111}$Ag. The silver activity was separated carrier-free from neutron irradiated natural Pd in the form of Ag(NO_3) in aqueous solution. This was deposited by evaporation on a single crystal disk of Cd metal (150 mg/cm^2 thick, 10 mm in diameter) cut with the c axis in the plane of the disk. The activity was diffused into one side of the disk by annealing in H_2 at ~300° for 6 h. After the annealing, residual surface activity was removed by etching and chemical polishing.

Since the activity is restricted to a thin layer on only one side of the Cd crystal, the β detector had to be fixed facing the source side, and the previous arrangement where the β detector alternately faced both sides of the crystal was unsuitable. The detector arrangement was reconstructed as shown in Fig. 5 to realize the geometry defined by (11). Thus the γ-ray detector was movable in the plane of the crystal containing the c axis and the $[\hat{c},\gamma]$ plane perpendicular to β. The two positions $\varphi = \pm\frac{1}{2}\pi$ then correspond to the γ detector at $\mp\frac{1}{4}\pi$ with respect to the c axis.

The unique first-forbidden β group to the 247 keV level ($T_{1/2} = 84$ nsec) in ^{111}Cd is relatively weak (1% branching) and has an end point of ~800 keV. The energy gate on the β detector was set between 250–750 keV and the γ detector was gated on the 247 keV γ ray. The admixtures due to β-(95γ)-247γ coincidences through the 340 keV level were unimportant owing to the very weak branching of 95 keV transition at the 340 keV level.

The time spectrum of $A(t)$ observed for ^{111}Cd in Cd metal is shown in Fig. 6. The solid line is a fit to the equation

$$A(t) = a\sin 2\omega_0 t + b\sin 3\omega_0 t \tag{21}$$

[see Eq. (20); the effect of the denominator is small and does not change the curve in any essential manner], where a and b are *positive*. The data clearly show that $A(t)$ is a superposition of two positive sine waves, thus showing unambiguously that ω_0 is positive. The result of the fit is $e^2qQ/h = (10/3\pi)\omega_0 = +125(4)$ MHz. The agreement of the magnitude with earlier γ-γ PAC results for this case[18] demonstrates that the diffusion process has resulted in the ^{111}Ag parent atoms being located at substitutional lattice sites.

V. DISCUSSION

The foregoing experiments clearly demonstrate the feasibility and utility of the β-γ PAC technique as a simple and reliable tool for the study of quadrupole interactions in nuclear excited states. The information on the sign of the QI which is derived directly is a unique feature of these experiments. The measurement on the 828 keV level in ^{115}In is of interest to nuclear physics. This level has many intriguing properties. To account for them, the interesting proposal[15] has been made that the level is a member of a rotational band with $K=\frac{1}{2}$ which coexists with other spherical shell-model levels in the single-particle nucleus $^{115}_{49}$In. This proposal predicts the quadrupole moment of this level to be about 0.6 b, which has been verified.[17] It also requires Q to be negative. Our measurement shows that e^2qQ for this level in Cd metal is

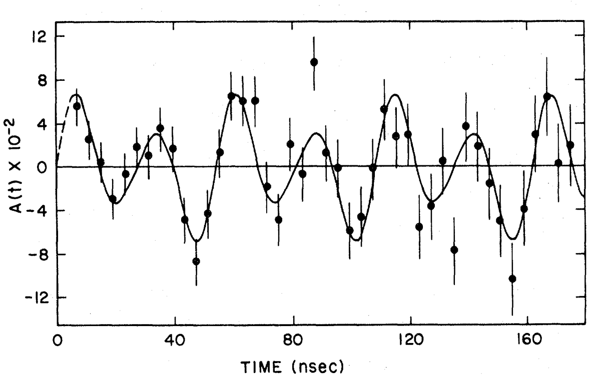

FIG. 6. Time dependence of the reflection asymmetry $A(t)$ of β-γ delayed coincidence between the positions γ and γ' of the γ detector, displaying the nuclear quadrupole spin precession of the 247 keV level of ^{111}Cd in Cd metal.

negative. Anticipating the result that the EFG for Cd in Cd metal is positive (see below), the EFG for In in Cd is very likely positive. Thus the Q of the 828 keV level is indeed very likely to be negative as required for the rotational picture. A result with the opposite sign would have completely ruled out the rotational model for this level.

The result that e^2qQ for Cd in Cd is positive is important. Recent studies of the QI in Cd and its variation with temperature[19] and pressure[20] have disclosed the major role of the conduction electrons in producing quadrupole effects in this metal. Theoretical calculations of the electronic part of the EFG in metals have been rare; but stimulated by these results, Mohapatra, Singal, and Das[21] have recently attempted to calculate in detail the different contributions to the effective EFG in Cd from a pseudopotential approach, and have shown that their theory is in reasonable agreement with the experimentally observed pressure dependence of the QI. An important aspect of this calculation is the prediction that the EFG due to the conduction electrons is greater than, and opposite in sign to, that due to the ion cores of the Cd lattice, producing a resultant EFG which is positive. Confirmation of this sign would provide definitive support to this theoretical approach for understanding quadrupole effects in metals. To obtain the sign of the EFG the sign of the quadrupole moment is needed. There is considerable evidence that Q of the 247 keV level of ^{111}Cd is positive. The systematic occurrence of low-lying $\frac{5}{2}^+$ states in odd Cd isotopes of mass 105–115 has been observed and these are generally thought to be $d_{5/2}$ neutron levels. The $\frac{5}{2}^+$ ground states of ^{105}Cd, ^{107}Cd, and ^{109}Cd are known to have $Q>0$.[22] Indirect experimental evidence from the work of Behrend and Budnick[8] also supports this. These authors measured $e^2qQ<0$ for the same ^{111}Cd level in an In metal lattice. The EFG for In in In is known to be negative.[23] If we make the plausible assumption that replacement of the In ion with a Cd ion with only s-type valence electrons does not change the sign of the EFG, a positive sign is indicated for the Q of the 247 keV level of ^{111}Cd. Accepting this, the present measurement shows that the EFG in Cd metal is positive as predicted by Mohapatra, Singal, and Das.[21] Our experimental result thus provides considerable encouragement for further tests of the pseudopotential approach to the theory of the EFG in Cd metal on a quantitative basis.

The ^{111}Cd measurement is interesting from another aspect. The 247 keV level of this nucleus has been one of the most extensively utilized angular correlation probes for QI studies in a number of systems. The coupling constants measured in all these instances are only magnitudes. Successful application of the β-γ PAC method to this particularly popular case is therefore valuable, opening as it does the possibility of deriving the signs in most of these cases, thereby making these experimental data more incisive for theoretical interpretation. As an example, we have reported elsewhere[24] β-γ measurements of the sign of the EFG at ^{111}Cd impurities in several noncubic metals. Such information is becoming available in a systematic way for the first time.

Potential new applications of the β-γ PAC technique are already under discussion in the literature. Allowed β-γ PAC is especially interesting. The observation of anisotropy is an unmistakable signature of the presence of a quadrupole coupling of the intermediate state; even an infinitely large magnetic interaction can result only in isotropy. This remarkable property can be exploited to selectively search for and identify small electric field gradients simultaneously present with large magnetic fields. It may be particularly valuable to apply this to field gradients at impurities in cubic ferromagnets, which arise as a result of asphericity induced in the conduction electron charge distribution by the spin-orbit coupling.[25] Another interesting application is the attempt of Rots, Namavar, and Coussement[26] to isolate quadrupole interaction induced by magnetostrictive strains at an iodine impurity in ferromagnetic nickel. Denisenko and Khamiovich[27] have proposed the detection of nuclear acoustic resonance by observing the induced β-γ correlation in a cubic crystal at the resonant frequency. Finally, quite apart from their application to QI studies, these experiments point to the use of the phenomenon of nuclear quadrupole precession as a general method for the detection of polarization in nuclear reaction and decay processes, thereby providing a useful new addition to the few methods presently available for this purpose.

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