I. 7. Hyperfine Interactions with the Presence of Both Electric Field Gradient and Magnetic Field—An Application to $^{111}$In PAC Spectroscopy

Hanada R.

Institute for Materials Research, Tohoku University

Introduction

Hyperfine interactions, the interactions between an atomic nucleus and the external crystal fields, have two origins. One is the magnetic interaction, namely the interactions between the nucleus magnetic moment and the external or internal magnetic field. The other is the electric interaction, the interaction of the nuclear quadrupole moment and the electric field gradient (EFG). When an element has a crystal structure different from the cubic, a finite EFG should take place at the atomic nuclei that is placed at the substitutional site. Such examples are Zn, Cd, Mg that have a closed packed hexagonal structure (hcp) or Sn that has a tetragonal structure. Indeed, PAC spectroscopy on them has shown precession signals of which origin is the electric.

Some elements with the hcp structure transform to ferromagnetic phase at a certain temperature. Such examples are Co and heavy rare earth elements as Gd, Tb, Dy, Ho, Er and Tm. So in their ferromagnetic phase, both the magnetic and the electric interactions are present at the probe nucleus. The purpose of the present work is to examine theoretically how this coexistence of the magnetic and electric hyperfine interactions is reflected on the PAC spectrum. Results of the calculation will be compared with the experimental results on rare earth elements and alloys in the following three papers.

In the present, we will give a general expression for the eigenvalues for I=5/2 nuclear level, for $^{111}$In, with the coexistence of the magnetic and electric terms. The solution of the equation gives the energy levels as a function of the ratio $\omega_{E}/\omega_{B}(=\gamma)$, $\eta$, the asymmetry parameter of EFG and the angles, $\alpha$, $\beta$ and $\gamma$ which specify the relative orientation between the magnetic field and the EFG. Second, we also calculate the transition probability among these sublevels for a special case of $\eta=0$. The results of the former calculation should be compared with the angular frequencies of PAC signals and the latter with the relative population of them. The whole calculation is based on the formulation by Matthias$^3$.

Matthias has calculated the energy levels for selected combinations of I, $\gamma$, $\eta$, $\alpha$ and $\beta$ without the transition probability. Böström et al have also simulated PAC spectrum based
on the theory although without any explicit expressions on the energy levels or the transition probability\(^3\).

**Formulation**

The matrix elements for the eigenvalue problem with both the electric and magnetic interactions read\(^1\):

\[
H_{mm} = -\omega_H \hbar m + \omega_E \hbar \frac{1}{2} \left( 3 \cos^2 \beta - 1 + \eta \sin^2 \beta \cos 2\alpha \right) \left( 3m^2 - I(I+1) \right),
\]

\[
H_{mm\pm 1} = \frac{3}{2} \omega_E \hbar \sin \beta \left( \cos \beta \pm \frac{\eta}{6} \right) \left( 1 \pm \cos \beta \right) e^{\pm i\gamma} e^{\pm 2\alpha m} e^{\pm \gamma/2},
\]

\[
H_{mm\pm 2} = \omega_E \hbar \frac{3}{4} \left( \sin^2 \beta + \frac{\eta}{6} \right) \left[ (1 \pm \cos \beta)^2 e^{\pm 2\alpha m} + (1 \mp \cos \beta)^2 e^{-\pm 2\alpha m} \right] e^{\pm 2\gamma/2} \left( I \pm m \pm 2 \right) \left( I \pm m \pm 1 \right) \left( I \mp m \pm 1 \right)^{1/2}.
\]

where \(\omega_H\) and \(\omega_E\) are magnetic and electric interaction frequency, respectively. Explicitly, \(\omega_H = g \mu_N H / \hbar\) and \(\omega_E = eqV_{mz} / \hbar\). \(\alpha, \beta,\) and \(\gamma\) are defined as in Fig. 1(a). The diagonalization of the hermitian matrix gives the eigenvalues for \(I=5/2\) state together with the eigenvectors \(u\).

The attenuation factor (the PAC spectrum) reads\(^3\):

\[
G_{kk}(t) = G_{N, k, k}^{N_\alpha, \alpha, \beta, \gamma}(t) = \sum_{N, m_a, m_b, n, \pi} \sum_{N, m_a, m_b, m_a, m_b} (-1)^{2\pi + m_a + m_b} \times \left( \begin{array}{cc} I & I \\ m_a' & -m_a \end{array} \right) \left( \begin{array}{cc} I & I \\ m_b' & -m_b \end{array} \right) \times u_n^{*} u_n m_a u_{n, m_b} u_{n, m_b} u_{n, m_a} \cdot u_{n, m_b}^* \cdot u_{n, m_a} \exp \left( -\frac{i}{\hbar} (E_n - E_n) t \right)
\]

So one can calculate the transition probability among sublevels (\(E_n\) and \(E_{n'}\)) by calculating the coefficient of the exponential term in eq.(2).

**Calculated results.**

**Eigenvalues**

The diagonalization of the matrix elements results in a characteristic equation shown in Fig. 1(b) of which solution gives the eigenvalues in terms of \(\gamma=(\omega_H/\omega_E), \eta, \alpha, \) and \(\beta\) (independent on \(\gamma\) when polycrystal). Note that this is a general solution and with it one can calculate the eigenvalues for \(I=5/2\) level with any combination of the parameters \(\gamma, \eta, \alpha,\) and \(\beta\) when both the magnetic and electric interactions are present. Fig. 2(a) shows an example of such a calculation for the case of \(\beta=90\) and \(\eta=0\) (independent on \(\alpha\) when \(\eta=0\)) with \(y\) varying
between 0 and 20. At $y=0$, namely, for pure quadrupole interaction only, we have three levels 10,-2 and -8, the transitions among them give the well known quadrupole frequencies of $\omega_1(=6\omega_b)$, $2\omega_1$, and $3\omega_1$. However, each sublevel is doubly degenerated and hence split into two with the application of the magnetic field thus giving 6 magnetic sublevels as expected for $I=5/2$ spin. Without the quadrupole interaction, namely for the pure magnetic interaction, the energy differences between the neighboring sublevels are the same. So the transitions between two neighboring levels (the total number of 5 for $n'-n=1$) give only one frequency, $\omega_{LH}$. Larmor frequency. The 1st harmonic of it, namely, $2\omega_{LH}$ is also possible for $n'-n=2$. So we have the well known two frequencies of $\omega_{LH}$ and $2\omega_{LH}$ for the pure magnetic interactions.

With the presence of the quadrupole interaction, however, the differences become dependent on the magnetic quantum number as shown in Fig. 2(a). So we should have 5 different frequencies in the PAC spectrum when both the magnetic and the electric interaction are present.

**Transition Probability and PAC Fourier Spectrum**

The transition probability can be obtained by calculating the coefficient of the exponential term of eq.(2). This gives the relative population of each transition. The eigenvectors $u$'s are obtained during the process of the diagonalization of the Hamiltonian matrix of eq.(1)

The relative populations of each transition are shown by Fig. 2(b) for $(n-n'=0)$, Fig. 2(c) for $(n-n'=1)$ and Fig. 2(d) for $(n-n'=2)$ transition. So if these results are applied to the PAC spectrum, for instance, for the $n-n'=1$ transition, we should have 5 different frequencies derived from the results in Fig. 1(a), each of which has the different populations given by Fig. 1(c). If these are combined, one can simulate a PAC Fourier spectrum for the $n-n'=1$ case when both the magnetic and the electric interactions coexist. Such an example is shown in Fig. 3 for the $n-n'=1$ case. These should be compared with the component near the basic frequency $1\omega_{LH}$ when both the magnetic and EFG with $\eta=0$ are present. Although not shown here, the same applies to the unperturbed term $(n-n'=0)$ with those in Fig. 2(b) or the 1st harmonic $2\omega_{LH}(n-n'=2)$ with Fig. 2(d)

_A Limiting Case of $y>>20$_

A limiting case of large $y$, namely, the case of much larger magnetic field than the EFG will be discussed in the next paper on Tb. There, the PAC spectrum was measured at RT (paramagnetic with only the EFG to give $\omega_b$) and at 77K (ferromagnetic with both the magnetic hyperfine field to give $\omega_{LH}$ and $\omega_b$) where $\omega_{LH}>>\omega_b$ was found. In the limiting case of large $y$, one can apply a simplified treatment of which relation to the present exact calculation will also be discussed.
Eigenvalues for Variable Parameters including the \( \eta = 0 \) case

Fig. 4(a) through Fig. 4(d) show the energy level of \( I = 5/2 \) spin as a function of \( y \) for several set of \( \alpha, \beta, \eta \). The population of each transition can also be calculated by eq. (4), though the presentation of them is beyond the scope of the present paper. Also it is possible to simulate the PAC Fourier spectra as in Fig. 3 for the general case of \( \eta = 0 \).

Application to Experimental Results

The present results apply not only for the analysis of PAC spectrum but also for those of NMR/ON or \( \beta \)-NMR on specimens with the EFG. These methods require to apply the external magnetic field and hence the analysis as in the present is inevitable. In these external magnetic field experiments, though, single crystals must be used to define the angles relation as shown in Fig. 1(a). For the case of the internal field as the hyperfine field, the magnetization axis and the EFG axis have a definite angle relation in each grain of the polycrystal and hence the present analysis can apply even to the experimental results on polycrystals.

References


\[
\lambda^4 + \left( -168 - \frac{56}{4} \right) \lambda^3 + \left( 56 \% - 56 \% \right) \lambda^2 + \left( \frac{256}{16} \right) \lambda + \left( 9 \right)
\]

\[
+ \left( 4704 \% - 972 \% - 52 \% \right) \lambda^4 + \left( 12 \% \right) \lambda^3 + \left( 10 \% \right) \lambda^2 + \left( 1 \% \right) \lambda + \left( 1 \% \right)
\]

\[
+ 324 \% \lambda^4 \cos(\beta)^2 \lambda^4 + \left( -4 \% \right) \lambda^3 + \left( -4 \% \right) \lambda^2 + \left( -1 \% \right) \lambda + \left( -1 \% \right)
\]

\[
- 960 \% \lambda^4 + \left( -16 \% \right) \lambda^3 + \left( -16 \% \right) \lambda^2 + \left( -16 \% \right) \lambda + \left( -16 \% \right)
\]

\[
- 160 \% \lambda^4 \cos(\beta)^2 \lambda^4 + \left( -4 \% \right) \lambda^3 + \left( -4 \% \right) \lambda^2 + \left( -1 \% \right) \lambda + \left( -1 \% \right)
\]

\[
- 4000 \% \lambda^4 + \left( 5 \% \right) \lambda^3 + \left( 5 \% \right) \lambda^2 + \left( 5 \% \right) \lambda + \left( 5 \% \right)
\]

\[
- 650 \% \lambda^4 \cos(\beta)^2 \lambda^4 + \left( 25 \% \right) \lambda^3 + \left( 25 \% \right) \lambda^2 + \left( 1 \% \right) \lambda + \left( 1 \% \right)
\]

\[
+ \left( 100 \% \right) \lambda^4 \cos(\beta)^2 \lambda^4 + \left( -1 \% \right) \lambda^3 + \left( -1 \% \right) \lambda^2 + \left( -1 \% \right) \lambda + \left( -1 \% \right)
\]

\[
- 4000 \% \lambda^4 + \left( -5 \% \right) \lambda^3 + \left( -5 \% \right) \lambda^2 + \left( -5 \% \right) \lambda + \left( -5 \% \right)
\]

\[
- 900 \% \lambda^4 + \left( -225 \% \right) \lambda^3 + \left( -225 \% \right) \lambda^2 + \left( 3 \% \right) \lambda + \left( 3 \% \right)
\]

\[
- 600 \% \lambda^4 \cos(\beta)^2 \lambda^4 + \left( 1 \% \right) \lambda^3 + \left( 1 \% \right) \lambda^2 + \left( 1 \% \right) \lambda + \left( 1 \% \right)
\]

Fig. 1. (a) The angular relations used in the present calculation according to Ref. (1).

Fig. 1. (b) The characteristic equation to give the eigenvalues for \( I = 5/2 \) spin with \( y = \omega y / \omega z \), \( \alpha \), \( \beta \) and \( \eta \) as the parameters.
Fig. 2. (a) Eigenvalues for \( I=5/2 \) spin with \( \beta=90^\circ \) and \( \eta=0 \) as a function of \( \gamma(=\omega_{H}/\omega_{E}) \).

Fig. 2. (b)-(d) the transition probabilities as a function of \( \gamma(=\omega_{H}/\omega_{E}) \) for \( I=5/2 \) spin with \( \beta=90^\circ \) and \( \eta = 0 \). (b) \( n-n'=0 \), (c) \( n-n'=\pm 1 \) and (d) \( n-n'=\pm 2 \).

Fig. 3. A simulated Fourier spectra for \( I=5/2 \) spin with \( \eta=0 \) as \( \gamma(=\omega_{H}/\omega_{E}) \) the parameter. Horizontal axis: the basic frequency \( (n-n'=\pm 1) \). Vertical axis: the relative populations (a) \( \beta=90^\circ \), (b) \( \beta=60^\circ \), (c) \( \beta=45^\circ \) and (d) \( \beta=30^\circ \).
Fig. 4. The eigenvalues for $I = 5/2$ spin. (a) $\eta = 0$, $\beta$ dependence. (b)(c) $\eta = 1$, $\alpha$ dependence with a fixed $\beta$. (d) $\eta = 0.5$, $\alpha$ dependence with a fixed $\beta$. 