

Magnon-induced pseudoquadrupole interaction in ordered systems

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Temperature-dependent aspects of the Suhl-Nakamura process, contributing to the measured quadrupole interaction in ordered systems, are described. The mechanism involves a pseudoquadrupolar coupling of magnons with nuclei via the magnetic hyperfine or transferred hyperfine $I_x S_x$ interaction. Within the framework of the long-wave magnon approximation, the process is linear in the magnetization and explains recent experimental results for ^{27}Al in GdAl_2 . A simple method of utilizing the new mechanism for the sign determination of the electric field gradient q is described, and the method is applied to determine $q < 0$ at the Al sites in GdAl_2 . Various implications of the new process are briefly discussed.

I. INTRODUCTION

Among various studies,¹⁻⁴ the one case for which the magnetization dependence of the nuclear quadrupole interaction was measured with sufficient accuracy to enable critical analysis is that of ^{27}Al in ferromagnetic GdAl_2 .⁴ Unexpected at the time, the induced quadrupole interaction in GdAl_2 was found to depend *linearly* on the magnetization $M(T)$ (Fig. 1), in contrast to theories^{1,2} which predicted quadratic M dependence for a magnetically induced electric field gradient. We suggest presently a temperature-dependent mechanism of the Suhl-Nakamura-type which contributes significantly to measured quadrupole interactions in ordered systems and can account for the apparent discrepancy mentioned above. Considering the indirect spin-spin interaction in ordered systems, Suhl⁵ and Nakamura⁶ have omitted contributions of $I_x S_x$ terms in the hyperfine interaction to both the indirect interaction between spins and to the self-energy of individual nuclear spins. As will be shown presently, the $I_x S_x$ contribution to the self-energy is important because it is temperature dependent and may lead to a measurable influence on the quadrupole splitting, unlike the quadrupolelike splitting caused by the transverse components $S_{\pm} I_{\mp}$ which is constant and therefore cannot be separated experimentally from the *intrinsic* quadrupole splittings.⁷ The present mechanism is essentially a pseudoquadrupolar process, proportional in magnitude to $(A^2/\epsilon_{\text{max}})[1 - M(T)/M_0]$, where A is the magnetic hyperfine constant, ϵ_{max} is the energy of a zone-boundary magnon, and $M_0 \equiv M(T=0)$. Evaluated numerically, the new mechanism appears to be of significance for many actual magnetic systems. As will be shown, the suggested mechanism provides a straightforward method to establish the sign of the electric field gradients q , and, in principle, can also reflect various properties of spin-wave systems.

II. THEORETICAL-MODEL CALCULATION

Consider the simple model of an axially symmetric hyperfine coupling between a nucleus I and a surrounding of neighboring electronic spin S within a magnetically ordered solid. The Hamiltonian of the system, including the magnon energy, has the form

$$\mathcal{H} = A_{\parallel} S_x I_x + \frac{1}{2} A_{\perp} (I_+ S_- + I_- S_+) + P_1 (I_x^2 - \frac{1}{3} \mathbf{I}^2) + \mathcal{H}_m, \quad (1)$$

where the first two terms describe the magnetic hyperfine interaction, the third is the intrinsic quadrupole interaction and $\mathcal{H}_m = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} n_{\mathbf{k}}$, where $\epsilon_{\mathbf{k}}$ is the energy of a magnon with a wave vector \mathbf{k} and $n_{\mathbf{k}}$ is the number of magnons in the \mathbf{k} th state. The first step in the model calculation is to reexpress the electronic spin operators (S_x , S_{\pm}) in terms of magnon operators. Choosing the origin at the site of the electronic spin and neglecting

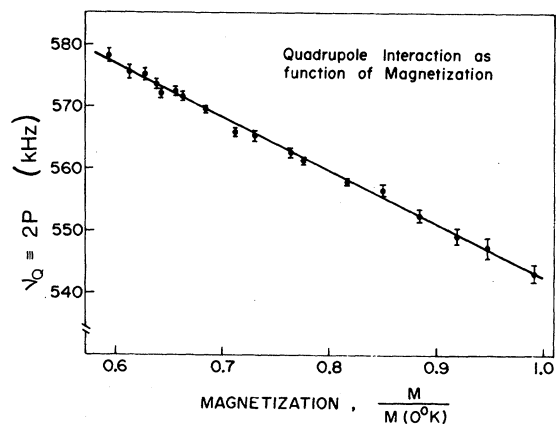


FIG. 1. Measured quadrupole splitting $\nu_Q = 2P$, of ^{27}Al in ferromagnetic GdAl_2 as function of the magnetization $M(T)$ (from Ref. 4).

third- and higher-order terms in the expansion for S_{\pm} , the Holstein-Primakof transformation yields

$$S_z = S - \frac{1}{N} \sum_{\vec{k}, \vec{k}'} b_{\vec{k}}^{\dagger} b_{\vec{k}'}; \quad S_+ = \left(\frac{2S}{N}\right)^{1/2} \sum_{\vec{k}} b_{\vec{k}}^{\dagger};$$

$$S_- = \left(\frac{2S}{N}\right)^{1/2} \sum_{\vec{k}} b_{\vec{k}}.$$

Replacing in Eq. (1), one obtains

$$\mathcal{H}_{\text{eff}} = \langle \cdots n_{\vec{k}} \cdots | \mathcal{H}_{\text{hf}} | \cdots n_{\vec{k}} \cdots \rangle + \sum_{n_{\vec{k}}} \frac{\langle \cdots n_{\vec{k}} \cdots | V | \cdots n_{\vec{k}} \cdots \rangle \langle \cdots n_{\vec{k}} \cdots | V | \cdots n_{\vec{k}} \cdots \rangle}{E \cdots n_{\vec{k}} \cdots - E \cdots n_{\vec{k}} \cdots}, \quad (3)$$

where \mathcal{H}_{hf} is the hyperfine part of Eq. (2), $|\cdots n_{\vec{k}} \cdots\rangle$ is a wave function with a magnon state $n_{\vec{k}}$, and the primed summation means exclusion of $n_{\vec{k}}$ states for which $E \cdots n_{\vec{k}} \cdots = E \cdots n_{\vec{k}} \cdots$. A straightforward calculation results in

$$\mathcal{H}_{\text{eff}} = \mathcal{H}_{\text{Zeeman}} + \mathcal{H}_{\mathcal{Q}} + v_1 + v_2 + v_3 = A_{\parallel} \langle S \rangle I_z + P_i (I_z^2 - \frac{1}{3} \tilde{I}^2) + \left(A_{\perp}^2 \frac{S}{2N} \sum_{\vec{k}} \frac{2n_{\vec{k}} + 1}{\epsilon_{\vec{k}}} \right) I_z$$

$$+ \left(A_{\perp}^2 \frac{S}{2N} \sum_{\vec{k}} \epsilon_{\vec{k}}^{-1} \right) (I_z^2 - \tilde{I}^2) + \left(\frac{1}{2} A_{\parallel}^2 \frac{1}{N^2} \sum_{\vec{k} \neq \vec{k}'} \frac{n_{\vec{k}} - n_{\vec{k}'}}{\epsilon_{\vec{k}} - \epsilon_{\vec{k}'}} \right) I_z^2, \quad (4)$$

where $\langle S \rangle = S - (1/N) \sum_{\vec{k}} n_{\vec{k}} \equiv S(1 - \Delta M/M_0)$. The last three terms (referred to as v_1 , v_2 , v_3 , respectively), resulting from the perturbation part of Eq. (3), will now be examined in detail. The term v_1 , which is linear in I_z , contributes a small second-order temperature-dependent correction to $\mathcal{H}_{\text{Zeeman}}$. Formally, such a correction may even be included in $\mathcal{H}_{\text{Zeeman}}$ by replacing A_{\parallel} with $A_{\parallel}(T)$. The second-order term v_2 is temperature independent, and being quadratic in I_z may be formally included in $\mathcal{H}_{\mathcal{Q}}$ by replacing P_i with a corrected constant P . Such a procedure is possible since one can always pick out the traceless operator $I_z^2 - \frac{1}{3} \tilde{I}^2$ from an operator $I_z^2 + \alpha \tilde{I}^2$, and neglect the rest which depends only on \tilde{I}^2 . The term v_2 was first derived by Suhl⁵ and Nakamura⁶ and was discussed for MnF_2 by Yasuoka, Ngwe, and Jaccarino.⁷ In the framework of the approximation employed in the present paper, one can show that $v_2 = \frac{1}{2} a (A_{\perp}/A_{\parallel})^2 (I_z^2 - \tilde{I}^2)$, where the constant a is defined in Eq. (8).¹⁰ Finally, we examine v_3 which results from the $I_z S_z$ operator in Eq. (1). Although again quadratic in I_z , the coefficient of v_3 is temperature dependent. Therefore, using the same procedure as above and noting that

$$\sum_{\vec{k} \neq \vec{k}'} \frac{n_{\vec{k}} - n_{\vec{k}'}}{\epsilon_{\vec{k}} - \epsilon_{\vec{k}'}} = 2 \sum_{\vec{k} \neq \vec{k}'} \frac{n_{\vec{k}}}{\epsilon_{\vec{k}} - \epsilon_{\vec{k}'}} ,$$

one may write

$$\mathcal{H} = \mathcal{H}_{\mathcal{C}_m} + A_{\parallel} S I_z + P_i (I_z^2 - \frac{1}{3} \tilde{I}^2)$$

$$- \frac{1}{N} A_{\parallel} I_z \sum_{\vec{k}, \vec{k}'} b_{\vec{k}}^{\dagger} b_{\vec{k}'}$$

$$+ A_{\perp} \left(\frac{S}{2N} \right)^{1/2} \left(I_+ \sum_{\vec{k}} b_{\vec{k}} + I_- \sum_{\vec{k}} b_{\vec{k}}^{\dagger} \right). \quad (2)$$

Considering now the magnon terms as a perturbation V , one obtains an effective nuclear spin Hamiltonian \mathcal{H}_{eff} valid up to second-order perturbation theory,^{8,9} of the form

$$v_3 = \mathcal{H}_{\mathcal{Q}}^{\text{pseudo}} = P^{\text{pseudo}} (I_z^2 - \frac{1}{3} \tilde{I}^2),$$

$$P^{\text{pseudo}} = A_{\parallel}^2 \frac{1}{N^2} \sum_{\vec{k} \neq \vec{k}'} \frac{n_{\vec{k}}}{\epsilon_{\vec{k}} - \epsilon_{\vec{k}'}} \quad (5)$$

from which an effective quadrupole Hamiltonian of the form

$$\mathcal{H}_{\mathcal{Q}}^{\text{eff}} = (P + P^{\text{pseudo}}) (I_z^2 - \frac{1}{3} \tilde{I}^2) \quad (6)$$

is obtained. This Hamiltonian, together with $\mathcal{H}_{\text{Zeeman}}$, determines the NMR frequency of nuclei in magnetic systems.

In order to calculate P^{pseudo} , we shall first calculate the sum over \vec{k}' in Eq. (5). Taking into account only the "acoustic" mode in the isotropic long-wave approximation, with $\epsilon = Dk^2$ as the dispersion law, the summation over k' is transformed to an integral by

$$\frac{1}{N} \sum_{\vec{k}'} (\epsilon_{\vec{k}} - \epsilon_{\vec{k}'})^{-1} \rightarrow - \frac{V}{2\pi^2 ND} P \int_0^{k_{\text{max}}} \frac{k'^2}{k'^2 - k^2} dk',$$

where the principal value of the integral is to be considered. In this approximation, one obtains

$$\frac{1}{N} \sum_{\vec{k}'} (\epsilon_{\vec{k}} - \epsilon_{\vec{k}'})^{-1} = \frac{V}{2\pi^2 ND}$$

$$\times \left(k_{\text{max}} + \frac{1}{2} k \ln \left| \frac{k_{\text{max}} - k}{k_{\text{max}} + k} \right| \right). \quad (7)$$

While Eq. (7) has a logarithmic divergence at $k = k_{\text{max}}$, it is easily seen that the sum which enters

in the definition of P^{pseudo} [Eq. (5)] does not contain any divergence. Indeed, at thermal equilibrium we find for $\vec{k} \approx \vec{k}'$ that

$$(n_{\vec{k}} - n_{\vec{k}'})(\epsilon_{\vec{k}} - \epsilon_{\vec{k}'})^{-1} = -\beta e^{\beta \epsilon_{\vec{k}}} (e^{\beta \epsilon_{\vec{k}}} - 1)^{-2},$$

where $\beta = 1/k_B T$. In fact, the divergence in Eq. (7) is the consequence of introducing the principal-value integration which is not valid for $k' = k_{\text{max}}$. The divergence can be avoided formally by the cut-off procedure which excludes the point $k = k_{\text{max}}$ yielding for that point the size-dependent term $\ln(L/a_0)$, where L is the sample size and a_0 is the lattice constant. Turning to the second summation in Eq. (5), we note that for the low-temperature range, say for $T \approx 0.3T_c$, the contribution from the summation is negligible for $\vec{k} \approx \vec{k}_{\text{max}}$ because in this region $n_k \ll 1$ and therefore $n_k \ln(L/a_0) \ll 1$. For the rest of the range, i.e., for $\vec{k} \ll \vec{k}_{\text{max}}$ the logarithmic term in Eq. (7) is small compared with the first one. As a result, the sum over k' becomes independent of k for low temperatures and the second summation, over k , yields simply $(1/N) \sum_{\vec{k}} n_{\vec{k}} = S [M_0 - M(T)] / M_0$. From Eqs. (5) and (7) one obtains, finally,

$$P^{\text{pseudo}} = -4\pi S \frac{A_{\parallel}^2}{\epsilon_{\text{max}}} \left(1 - \frac{M(T)}{M_0} \right) \\ = -a \left(1 - \frac{M(T)}{M_0} \right); \quad a > 0 \quad (8)$$

after using $\epsilon_{\text{max}} = Dk_{\text{max}}^2$ and $k_{\text{max}}^3 = (2\pi)^3(N/V)$. It is worthwhile to note at this point that the linearity of P^{pseudo} with M , which is evident from Eq. (8), does not depend strongly on the particular dispersion law used in the derivation of Eq. (8). Thus, while more realistic dispersion laws may affect the slope a appreciably, one may still expect a linear M dependence to be dominant in the spin-wave region.

III. EXPERIMENTAL IMPLICATIONS AND APPLICATIONS

Some implications of Eq. (8) will now be discussed. Substituting in Eq. (6), it is seen that P^{pseudo} vanishes at $T=0$ when the spins are fully aligned. At this limit a measurement of P yields, apart from the correction mentioned earlier due to v_2 [Eq. (4)], the real quadrupole interaction

$P_i = (3e^2Qq)/[4I(2I-1)]$. As the temperature is raised, the onset of magnetic excitations results in negative contribution added on to P . From Eq. (8) it is evident that in the spin-wave region P^{pseudo} changes *linearly* with the magnetization, with a positive slope, a , which becomes more significant the stronger is the hyperfine coupling and the lower is T_c for the system in question. For AS values in actual magnetic systems with $T_c \approx 300^\circ\text{K}$, Eq. (8) predicts typical slope values in the range $5 \text{ kHz} \lesssim a \lesssim 5 \text{ MHz}$. It is therefore feasible to test Eq. (8) experimentally. As pointed out already earlier, in the only documented study available at present a linear M dependence was indeed observed for $\mathcal{H}C_{\text{eff}}^{\text{eff}}$ of ^{27}Al in GdAl_2 (Fig. 1), with a slope $|M_0(\partial P^{\text{eff}}/\partial M)| = 40 \text{ kHz}$.¹¹ On the other hand, using the parameters appropriate for GdAl_2 [$S = \frac{7}{2}$, $A_{\parallel} \approx 20 \text{ MHz}$, $T_c = 174^\circ\text{K}$, $\epsilon_{\text{max}} \approx k_B T_c / (S+1)$] the prediction of Eq. (8) is $a = 22 \text{ kHz}$. In view of the approximation in deriving Eq. (8), the agreement between the two results is quite reasonable. Lacking any other explanation for the experimental results of Ref. 4, we therefore propose these results as the first experimental verification of the temperature-dependent P^{pseudo} mechanism.

The P^{pseudo} process can be used to determine directly the sign of the product Qq . To see this, note that when $|A_{\parallel}| \gg |P|$, the quadrupole splitting will have the form [see Eqs. (6) and (8)] $\Delta E_Q = |P - a[1 - M(T)/M_0]|$. Since a is always positive, $d(\Delta E_Q)/dM > 0$ means $P > 0$ and therefore positive Qq product, whereas $d(\Delta E_Q)/dM < 0$ implies $Qq < 0$. Applying the criterion to ^{27}Al in GdAl_2 , the observed negative slope implies a negative intrinsic electric field gradient, $q < 0$, at the Al sites of GdAl_2 .

For another implication of Eqs. (6) and (8), note that even in systems with an essentially vanishing Qq product, it is quite feasible to observe a quadrupolarlike splitting of an NMR line upon the introduction of magnetic excitations into the system. Several such systems are presently being investigated in our laboratory. Finally, we note that at least, in principle, an experimental study of P^{pseudo} [Eq. (5)] may reflect both the dispersion relations and the thermodynamic properties of actual magnon systems.

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¹⁰Comparing ν_2 with the ν_3 term [Eq. (8)] we note that the $I_z S_z$ contribution is small compared with the transverse contribution only when $M(T)/M_0 \sim 1$, whereas for $M(T)/M_0 \sim \frac{1}{2}$, the two terms are already comparable. Qualitatively, this may imply that the $I_z S_z$ contribution to the indirect spin-spin interaction, which leads to terms of the form

$$I_z^i I_z^j A^2 \frac{1}{N^2} \sum_{kk'} \frac{n_k - n_{k'}}{\epsilon_{k'} - \epsilon_k} \cos(\vec{k} - \vec{k}') (\vec{r}_i - \vec{r}_j),$$

may be of importance to NMR linewidths at finite temperatures.

¹¹The vertical axis of Fig. 3 in Ref. 4 represents values of $3e^2 Qq/[2I(2I-1)]$ and not $3e^2 Qq/[4I(2I-1)]$ as printed in the caption.