Theory of the antiferromagnetic-resonance linewidth of MnF_2

R. M. White and R. Freedman
Xerox Palo Alto Research Center, Palo Alto, California 94304

Roy B. Woolsey
Technology for Communications International, Mountain View, California 94040

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The temperature-dependent contribution to the linewidth of the antiferromagnetic resonance in MnF_2 at low temperatures is shown to arise from four-magnon scattering processes. The scattering amplitudes associated with the exchange and anisotropy interactions are comparable but of opposite sign, leading to an "anisotropy-narrowed" linewidth. As the temperature increases the anisotropy decreases and the exchange scattering becomes dominant. The theory provides a quantitative fit to high-field experimental data up to $T_N/4$. Above this temperature six-magnon scattering appears to dominate.

Magnetic resonance is characterized by a uniform precession of magnetic moments. In an ordered magnetic material this corresponds to a long-wavelength spin wave. The lifetime associated with this excitation is determined by the manner in which the energy is transferred into other modes of the system. The classic ferromagnetic system is yttrium iron garnet (YIG). Its relaxation has been exhaustively studied[1] and it has been established that the energy of the uniform precession is first transferred to short-wavelength spin waves and subsequently to phonons. In this paper we show that this sequence occurs in the antiferromagnet MnF_2 and argue that it should be a general feature of antiferromagnets.

Manganese fluoride is the classic antiferromagnet with a Néel temperature of 68 °K. MnF_2 owes its unique position to several facts. First of all, the manganese ion is in the divalent state which is an orbital singlet ($g = 2.001$). This removes hard-to-calculate spin-orbit contributions to the anisotropy and magnetoelastic energies and makes MnF_2 attractive from a theoretical point of view. On the experimental side, both Mn^{55} and F^{19} possess nuclear moments so that nuclear magnetic resonance can be used to probe both local environments. Furthermore, the neutron scattering cross sections are such as to make elastic and inelastic scattering possible. And, finally, large single crystals are relatively easy to grow. As a result of these advantages, the basic magnetic properties of MnF_2 have been thoroughly studied. In particular, the magnetic excitation spectrum is well known as a function of temperature and applied field.[2]

One of the disadvantages of MnF_2 arises from its tetragonal structure. As a result the magnetic dipolar interaction gives rise to a relatively large anisotropy, which drives the antiferromagnetic-resonance (AFMR) frequency up into the far infrared. For studying relaxation phenomenon, this is a difficult region in which to work: tuneable electromagnetic sources are not available and neutron scattering does not have high enough resolution. However, in the presence of a strong (80 kOe) magnetic field one of the AFMR modes is driven down into the millimeter-wave region where it can be studied.

The first AFMR study on MnF_2 was carried out by Johnson and Nethercot.[3] They observed that the linewidth broadened as the temperature approached the Néel point. This could be understood in terms of fluctuations in the exchange field. They were not able, however, to explain the residual low-temperature linewidth. There have been estimates made of various mechanisms that might be responsible for this linewidth. Genkin and Fain[4] obtained a linewidth of 12 Oe at 6 °K associated with four-magnon exchange scattering in zero applied field. Similarily, Upadhyaya and Sinha[5] estimated the linewidth associated with a one-phonon two-magnon process to be 10 Oe at 10 °K. However, Kotthaus and Jaccarino[6] have recently identified this linewidth as arising from two-magnon imperfection scattering. When this contribution is subtracted from the measured linewidth it leaves a small thermally induced relaxation rate which is only of the order of 10^6 sec^{-1} at 4 °K but increases rapidly with temperature. The fact that this linewidth is smaller than that predicted by the magnon-phonon mechanism suggests that the magnon-phonon coupling constant is smaller than originally thought. On the other hand, the Genkin-Fain estimate of the four-magnon exchange mechanism is two orders of magnitude larger than the data of Kotthaus and Jaccarino. However, the anisotropy interaction provides an additional four-magnon scattering amplitude which must also be considered. As we shall see below the anisotropy scattering amplitude interferes with that from the exchange to give a much smaller linewidth.

The Hamiltonian for MnF_2 consists of exchange,

magnetic dipole, and Zeeman terms. The dipolar interactions introduce ellipticity into the spin-wave modes which can be important under certain circumstances. However, for the case we are considering here it is sufficient to represent the dipolar interaction by a phenomenological anisotropy term,

$$\mathcal{H}_{an} = -\frac{K}{2}\left(\sum_i (S_i^z)^2 + \sum_j (S_j^z)^2\right), \tag{1}$$

where i and j refer to the two sublattices. The phenomenological constant K has the value of 0.41 °K at 4 °K and decreases according to $K(T)/K(0) = [M(T)/M(0)]^3$. The exchange Hamiltonian has the isotropic Heisenberg form

$$\mathcal{H}_{ex} = -2|J_1|\sum_i \sum_{\vec{\delta}} \vec{S}_i \cdot \vec{S}_{i+\vec{\delta}} + 2J_2 \sum_i \sum_{\vec{\delta}'} \vec{S}_i \cdot \vec{S}_{i+\vec{\delta}'}, \tag{2}$$

where[2] $|J_1| = 0.32$ °K and $J_2 = 1.76$ °K. The sum over $\vec{\delta}'$ is a sum over the eight next nearest neighbors on the opposite sublattice while that over $\vec{\delta}$ is over the two nearest neighbors on the same sublattice.

Spin-deviation operators are introduced in the usual manner,

$$\begin{aligned} S_i^+ &= \sqrt{2S}\, f_i a_i, \quad & S_j^+ &= \sqrt{2S}\, b_j^\dagger f_j, \\ S_i^- &= \sqrt{2S}\, a_i^\dagger f_i, \quad & S_j^- &= \sqrt{2S}\, f_j b_j, \\ S_i^z &= S - a_i^\dagger a_i, \quad & S_j^z &= -S + b_j^\dagger b_j, \end{aligned} \tag{3}$$

where

$$f_i = (1 - a_i^\dagger a_i/2S)^{1/2}.$$

These, in turn, are related to spin wave operators by

$$\begin{aligned} a_i &= \frac{1}{\sqrt{N}} \sum_{\vec{k}} a_{\vec{k}} e^{i\vec{k}\cdot\vec{r}_i}, \\ b_j &= \frac{1}{\sqrt{N}} \sum_{\vec{k}} b_{\vec{k}} e^{-i\vec{k}\cdot\vec{r}_j}, \end{aligned} \tag{4}$$

where N is the number of Mn ions per sublattice per unit volume. Due to the intersublattice exchange these spin waves are not the normal modes of the system. That is, when the Hamiltonian is expanded to second order in these operators, we observe coupling between $a_{\vec{k}}$ and $b_{\vec{k}}^\dagger$. The normal modes of the system may be obtained by diagonalizing these quadratic terms. The resulting transformation is

$$\begin{aligned} a_{\vec{k}} &= u_{\vec{k}} \alpha_{\vec{k}} - v_{\vec{k}} \beta_{\vec{k}}^\dagger, \\ b_{\vec{k}}^\dagger &= -v_{\vec{k}} \alpha_{\vec{k}} + u_{\vec{k}} \beta_{\vec{k}}^\dagger, \end{aligned} \tag{5}$$

where

$$u_{\vec{k}} = \left(\frac{A + \omega_{\vec{k}}}{2\omega_{\vec{k}}}\right)^{1/2}, \quad v_{\vec{k}} = \left(\frac{A - \omega_{\vec{k}}}{2\omega_{\vec{k}}}\right)^{1/2}; \tag{6}$$

and

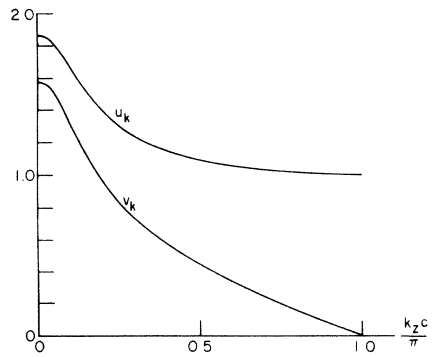

FIG. 1. Wave-vector dependence of the normal-mode transformation coefficients for $\vec{k} \| [001]$.

$$A = \frac{2z_2 J_2 S}{\hbar} + \frac{KS}{\hbar} = \omega_{ex} + \omega_{an}, \tag{7}$$

$$\omega_{\vec{k}} = [(2\omega_{ex} + \omega_{an})\omega_{an} + \omega_{ex}^2(1 - \gamma_{\vec{k}}^2)]^{1/2}, \tag{8}$$

with

$$\gamma_{\vec{k}} = \frac{1}{8}\sum_{i=1}^{8} e^{i\vec{k}\cdot\vec{\delta}_i}. \tag{9}$$

Because of their importance to our subsequent discussion we have plotted these transformation coefficients in Fig. 1. These values are relatively insensitive to the variation of ω_{an} with temperature. Under this transformation the quadratic part of the total Hamiltonian takes the form

$$\mathcal{H}^{(2)} = \sum_{\vec{k}} (\hbar\Omega_{\vec{k}}^{(+)} \alpha_{\vec{k}}^\dagger \alpha_{\vec{k}} + \hbar\Omega_{\vec{k}}^{(-)} \beta_{\vec{k}}^\dagger \beta_{\vec{k}}), \tag{10}$$

where

$$\Omega_{\vec{k}}^{(\pm)} = \omega_{\vec{k}} \pm \gamma H_0 \tag{11}$$

and $\gamma = g\mu_B/\hbar$. It is interesting to note that the transformation in which the minus signs associated with the $v_{\vec{k}}$'s in Eq. (5) are replaced by plus signs also diagonalizes the Hamiltonian with the same eigenvalues. The choice of this phase is determined by noting that the minus sign gives the same mode configuration for the uniform precession found by Keffer and Kittel[7] from the classical equation of motion. Therefore by the correspondence principle this is the sign that must be used in Eq. (5). One also arrives at the same conclusion from an approach based on the quantum-mechanical equations of motion. As we shall see below this phase is important in determining the relaxation of the uniform precession.

The temperature dependence of spin-wave frequencies and spin-wave rates depend on nonlinearities, that is, terms in the spin-wave expansion of the Hamiltonian that are higher than second order. Due to the antiferromagnetic symmetry there are no third-order terms in this expansion. Therefore

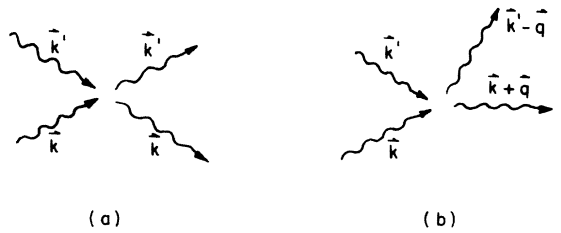

FIG. 2. Schematic representation of the two general classes of four-magnon processes: (a) those contributing to renormalization of the spin-wave frequencies, and (b) those contributing to relaxation.

the lowest-order nonlinear processes in antiferromagnets arise from four-magnon terms. Two most important classes of such terms are illustrated in Fig. 2. The contributions of the terms shown in Fig. 2(a) to the temperature dependence of the spin-wave frequences have already been computed.[8] This gives very good agreement with the spin-wave dispersion relation obtained from inelastic neutron scattering. An additional test of this renormalization is the calculation of the dependence of the spin-flop transition in MnF_2. If one characterizes this transition as the point at which the spin-wave frequency goes to zero then the resulting phase boundary has the form indicated by the solid line[9] in Fig. 3. These successful applications of nonlinear spin-wave theory give us confidence that such an approach may also describe relaxation phenomena. In fact, the relaxation of zone-boundary magnons in MnF_2 has already been investigated.[10]

Since the relaxation measurements of Kotthaus and Jaccarino were carried out in a very large magnetic field the high-frequency modes should have little effect on the results for low temperatures ($T<20\,^{\circ}\mathrm{K}$). We therefore consider only those four-magnon terms in the Hamiltonian involving the low-frequency modes. The four-magnon terms arising from the exchange are

$$\mathcal{H}_{ex}^{\alpha-\alpha}=\sum_{\substack{\vec{k}_1,\vec{k}_2\\ \vec{k}_3,\vec{k}_4}} C^{(ex)}_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4}\alpha^{\dagger}_{\vec{k}_1}\alpha^{\dagger}_{\vec{k}_2}\alpha_{\vec{k}_3}\alpha_{\vec{k}_4}\Delta(\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4)\,, \tag{12}$$

where

$$C^{(ex)}_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4}=\frac{J_2 z_2}{2N}(u_{\vec{k}_1}v_{\vec{k}_2}v_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_1}+u_{\vec{k}_1}u_{\vec{k}_2}u_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_4}+u_{\vec{k}_1}v_{\vec{k}_2}u_{\vec{k}_3}u_{\vec{k}_4}\gamma_{\vec{k}_2}+v_{\vec{k}_1}v_{\vec{k}_2}u_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_3}-4u_{\vec{k}_1}v_{\vec{k}_2}u_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_2-\vec{k}_4})$$

$$+\frac{|J_1|z_1}{4N}(\gamma_{\vec{k}_1}+\gamma_{\vec{k}_2}+\gamma_{\vec{k}_3}+\gamma_{\vec{k}_4}-4\gamma_{\vec{k}_2-\vec{k}_4})(u_{\vec{k}_1}u_{\vec{k}_2}u_{\vec{k}_3}u_{\vec{k}_4}+v_{\vec{k}_1}v_{\vec{k}_2}v_{\vec{k}_3}v_{\vec{k}_4})\,. \tag{13}$$

Similarily, the anisotropy interaction gives the terms

$$\mathcal{H}_{an}^{(\alpha-\alpha)}=\sum_{\substack{\vec{k}_1,\vec{k}_2\\ \vec{k}_3,\vec{k}_4}} C^{(an)}_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4}\alpha^{\dagger}_{\vec{k}_1}\alpha^{\dagger}_{\vec{k}_2}\alpha_{\vec{k}_3}\alpha_{\vec{k}_4}\times\Delta(\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4) \tag{14}$$

where

$$C^{(an)}_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4}=-\frac{K}{2N}(u_{\vec{k}_1}u_{\vec{k}_2}u_{\vec{k}_3}u_{\vec{k}_4}+v_{\vec{k}_1}v_{\vec{k}_2}v_{\vec{k}_3}v_{\vec{k}_4})\,. \tag{15}$$

Notice that the exchange contribution involves a complicated combination of the transformation coefficients u and v, the majority of the terms containing an *odd* number of v's. The anisotropy contribution on the other hand is dominated by terms involving products of four u's. The fact that v is less than u and is accompanied by a minus sign as we discussed above leads to the result that the four-magnon scattering amplitudes associated with the anisotropy are comparable to but opposite in sign to those associated with the exchange. Consequently both must be retained in the calculation.

The general expression for the relaxation rate associated with the four-magnon scattering illustrated in Fig. 2(b) is derived in a manner similar to that for the three-magnon process.[11] The result is

$$\eta_{\vec{k}_1}=\frac{\pi}{\hbar^2}[\exp(\hbar\Omega_{\vec{k}_1}/k_BT)-1]\sum_{\vec{k}_2\vec{k}_3\vec{k}_4}|C|^2\exp(\hbar\Omega_{\vec{k}_2}/k_BT)\times n_{\vec{k}_2}n_{\vec{k}_3}n_{\vec{k}_4}\Delta(\vec{k}_2-\vec{k}_3-\vec{k}_4)\delta(\Omega_{\vec{k}_1}+\Omega_{\vec{k}_2}-\Omega_{\vec{k}_3}-\Omega_{\vec{k}_4})\,, \tag{16}$$

where

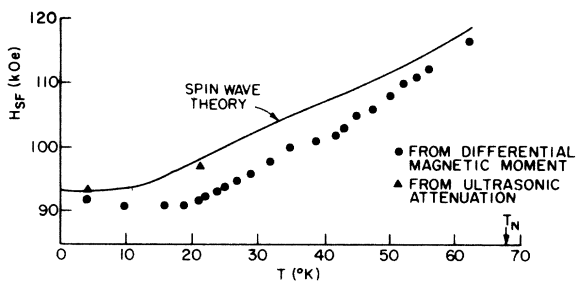

FIG. 3. Comparison of theory (solid line) and experimental data for the spin-flop field in MnH_2 (after Timbie, Ref. 9).

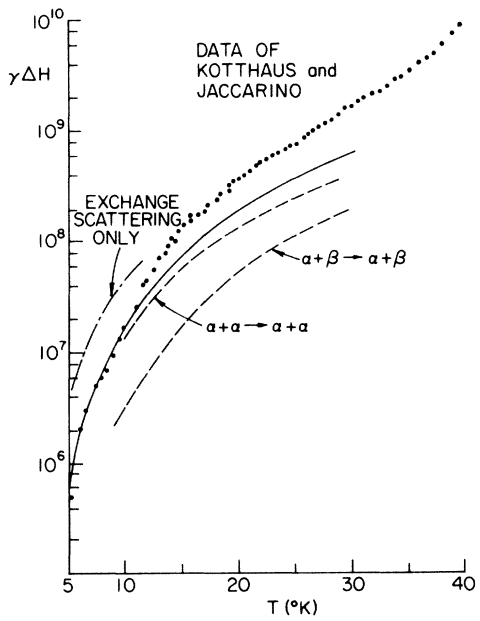

FIG. 4. Comparison of theory (solid line) and experimental data for the linewidth of the antiferromagnetic resonance in MnF_2.

$$C = C_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4} + C_{\vec{k}_2\vec{k}_1\vec{k}_3\vec{k}_4} + C_{\vec{k}_1\vec{k}_2\vec{k}_4\vec{k}_3} + C_{\vec{k}_2\vec{k}_1\vec{k}_4\vec{k}_3} \ . \quad (17)$$

This expression was evaluated numerically for $\vec{k}_1 = 0$. The sums over the six coordinates associated with the two independent wave vectors in Eq. (16) were carried out by dividing the range from $k = 0$ to $k = k_{max}$ into approximately 50 points and weighing the contribution of each to the sum appropriately. This corresponds to $(50)^6$ operations which took about one hour and a half to run on a CDC 6600. The δ function was handled by testing if $|\Omega_{\vec{k}_1} + \Omega_{\vec{k}_2} - \Omega_{\vec{k}_3} - \Omega_{\vec{k}_4}|$ were less than some value $\frac{1}{2}\Delta\Omega$. If so, the contribution of that term was weighted by $1/\Delta\Omega$. A range of mesh sizes and $\Delta\Omega$'s were found that made the answer independent of these quantities. The resulting relaxation rate as a function of temperature is indicated by the solid line in Fig. 4. The points are the data of Kotthaus and Jaccarino.[6] The point to keep in mind is that there are *no* adjustable parameters in this relaxation calculation. Therefore the excellent fit to the experimental data in the low-temperature region where spin-wave theory is valid indicates that four-magnon scattering is the dominant relaxation mechanism.

In order to demonstrate the relative roles of the exchange and anisotropy the dot-dashed curve in Fig. 4 shows the relaxation rate associated with exchange scattering only. We notice that without the interference from the anisotropy scattering amplitude the linewidth is an order of magnitude larger than what is observed.

We notice that the experimental points break away from our theoretical curve about 15° indicating that another relaxation channel may becoming important. Since the energy of the high-field branch is at 22 °K we might suspect that *inter*mode scattering is now becoming important. The Hamiltonian for the confluence of an α and a β mode giving rise to a new α and a new β mode is

$$\mathcal{H}_{ex}^{\alpha-\beta} = \sum_{\substack{\vec{k}_1,\vec{k}_2\\ \vec{k}_3,\vec{k}_4}} D^{(ex)}_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4} \alpha^\dagger_{\vec{k}_1}\beta^\dagger_{\vec{k}_2}\alpha_{\vec{k}_3}\beta_{\vec{k}_4} \times \Delta(\vec{k}_1 - \vec{k}_2 - \vec{k}_3 + \vec{k}_4) \quad (18)$$

where

$$D^{(ex)}_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4} = \frac{J_2 z_2}{N}\left(u_{\vec{k}_1}u_{\vec{k}_2}u_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_2} + u_{\vec{k}_1}v_{\vec{k}_2}v_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_3} + u_{\vec{k}_1}u_{\vec{k}_2}v_{\vec{k}_3}u_{\vec{k}_4}\gamma_{\vec{k}_1} + v_{\vec{k}_1}u_{\vec{k}_2}v_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_4} + v_{\vec{k}_1}v_{\vec{k}_2}v_{\vec{k}_3}u_{\vec{k}_4}\gamma_{\vec{k}_2} + v_{\vec{k}_1}u_{\vec{k}_2}u_{\vec{k}_3}u_{\vec{k}_4}\gamma_{\vec{k}_3} + v_{\vec{k}_1}v_{\vec{k}_2}u_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_1} + u_{\vec{k}_1}v_{\vec{k}_2}u_{\vec{k}_3}u_{\vec{k}_4}\gamma_{\vec{k}_4} - 2u_{\vec{k}_1}u_{\vec{k}_2}u_{\vec{k}_3}u_{\vec{k}_4}\gamma_{\vec{k}_2-\vec{k}_4} - 2u_{\vec{k}_1}v_{\vec{k}_2}v_{\vec{k}_3}u_{\vec{k}_4}\gamma_{\vec{k}_3-\vec{k}_4} - 2v_{\vec{k}_1}u_{\vec{k}_2}u_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_2-\vec{k}_1} - 2v_{\vec{k}_1}v_{\vec{k}_2}v_{\vec{k}_3}v_{\vec{k}_4}\gamma_{\vec{k}_3-\vec{k}_1}\right) . \quad (19)$$

The corresponding relaxation rate is given by Eq. (16) where the symmetrized coefficient C is now replaced by the expression given in Eq. (19). Since two input and two output modes are not identical this coefficient is *not* symmetrized as was done in Eq. (17). On the other hand Eq. (16) must be multiplied by a factor of 2 to account for the fact that the two output magnons are not identical. The result of this calculation is indicated by the appropriate dashed line in Fig. 4. We see that the sum of the two processes does improve the agreement with the experimental data somewhat. But there is still a significant departure at high temperatures. Another feature neglected in this calculation is the wave-vector dependence of the anisotropy in MnF_2 arises primarily from the magnetic dipole-dipole interaction, this should be used in place of the single-ion interaction given in Eq. (1). This has several consequences. First of all, it introduces quadratic terms of the form $a_{\vec{k}}a_{-\vec{k}}$ which require a 4×4 transformation instead of the 2×2 transformation of Eq. (5). This lifts some of the degeneracy of the spin-wave spectrum. Secondly, the four-magnon terms associated with the dipole-dipole interaction have a different coefficient than that given for the single-ion anisotropy in Eq. (15).

The wave-vector dependence enters both the quadratic and quartic terms through expressions of the form

$$\sum_i \frac{1}{R_i^3}\left(1-\frac{3Z_i^2}{R_i^2}\right)e^{i\vec{k}\cdot\vec{R}_i}\ .$$

The dipolar sum converges when R_i is of the order of five lattice constants. Therefore when the wave vectors start to become of the order of $\frac{1}{10}k_{BZ}$ we might expect to see effects from the "softening" of the anisotropy constant. This would tend to reduce the interference and enhance the relaxation rate. Although this might account for some of the observed increase in ΔH above 20 °K it is clear from Fig. 4 that even the curve associated with four-magnon exchange scattering only will eventually fall below the data.

When our numerical results are plotted on a logarithmic scale, they display a T^4 dependence, while the experimental data begin to show a stronger dependence at temperatures above 15 °K. The reason that the four-magnon process is not as effective at these temperatures has to do with the fact that the large field at which these experiments were carried out has pushed the uniform precession frequency to a relatively low value. Therefore the four-magnon process indicated in Fig. 2(b) looks like a three-magnon process for which energy and momentum cannot be satisfied with an antiferromagnetic dispersion relation. We verified this conclusion by computing the relaxation rate for a field of 1 kOe and found the four-magnon process to continue to be important at higher temperatures.

We would like to thank Madeline Rodoni for her assistance in carrying out the computations and Professor Jaccarino and Dr. Kotthaus for initiating our interest in this problem.

[1]See, for example, M. Sparks, *Ferromagnetic-Relaxation Theory* (McGraw-Hill, New York, 1964).
[2]A. Okazaki, K. C. Turberfield, and R. W. H. Stevenson, Phys. Lett. 8, 9 (1964).
[3]F. M. Johnson and A. H. Nethercot, Phys. Rev. 114, 705 (1959).
[4]V. N. Genkin and V. M. Fain, Zh. Eskp. Teor. Fiz. 41, 1522 (1961) [Sov. Phys.—JETP 14, 1086 (1962)].
[5]U. N. Upadhyaya and K. P. Sinha, Phys. Rev. 130, 939 (1963).
[6]J. P. Kotthaus and V. Jaccarino, Phys. Lett. A 42, 361 (1973).
[7]F. Keffer and C. Kittel, Phys. Rev. 85, 329 (1952).
[8]G. G. Low, *Inelastic Scattering of Neutrons*, Vol. I, (International Atomic Energy Agency, Vienna, 1965).
[9]J. P. Timbie, Ph.D. dissertation (Stanford University, Stanford, California, 1971) (unpublished).
[10]R. B. Woolsey and R. M. White, Int. J. Magn. 2, 51 (1972).
[11]Reference 1, p. 91.