interpretation of damage studies will be possible.

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EFFECT OF EXCHANGE SCATTERING ON NUCLEAR SPIN RELAXATION IN MAGNETIC MATERIALS*

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There has recently been a growing body of evidence that three-magnon processes play an important role in relaxing the nuclear magnetization in ordered magnetic insulators. In particular, (1) Fromhold and Narath[1] have measured the Cr^{53} T_1 in metamagnetic $CrCl_3$ and find temperature and field dependences characteristic of the three-magnon process but about an order of magnitude shorter than the theoretical prediction. (2) Welsh and Portis[2] have observed a T^5 dependence for the relaxation rate of Mn^{55} in antiferromagnetic $CsMnF_3$. This is the predicted[3] temperature dependence for the three-magnon process in antiferromagnets at temperatures high compared to the effective spin-wave gap temperature. (3) Kaplan and Jaccarino[4] have investigated T_1 of Mn^{55} in antiferromagnetic MnF_2 and find strong evidence for intrinsic spin-wave relaxation processes in the region $k_BT \gtrsim \Delta$ (Δ is energy gap in the spin-wave spectrum).

The necessity for considering the three-magnon process for nuclear relaxation relative to lower order spin-wave mechanisms can be summarized as follows: (1) The direct process, i.e., emission of single spin waves of arbitrarily well-defined energies, is not allowed by energy conservation because the nuclear resonance frequency is invariably much smaller than the minimum spin-wave energy (Δ). (2) The Raman process,[5] i.e., scattering of a thermal magnon with a simultaneous nuclear spin flip, has a vanishing matrix element in the situations of interest here where we have an isotropic hyperfine interaction, $A\vec{I}\cdot\vec{S}$, and the electronic spins and nuclear are quantized along the same axis. The selection rule against this process is a result of angular-momentum conservation. Thus the lowest order spin-wave process which can relax the

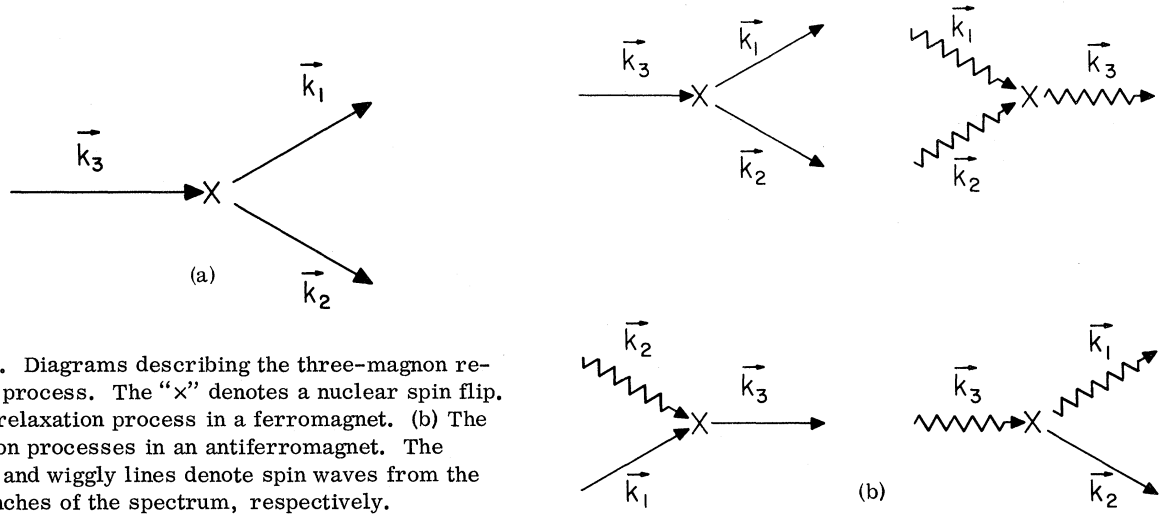

FIG. 1. Diagrams describing the three-magnon relaxation process. The "×" denotes a nuclear spin flip. (a) The relaxation process in a ferromagnet. (b) The relaxation processes in an antiferromagnet. The straight and wiggly lines denote spin waves from the two branches of the spectrum, respectively.

nuclear spin and conserve both energy and angular momentum is the three-magnon process. In addition, of course, there are various types of impurity relaxation mechanisms which under certain conditions may supersede the intrinsic processes. We are further restricting ourselves to those systems where phonon mechanisms are not competitive. This is most likely when dealing with S-state electronic systems.

The three-magnon nuclear relaxation mechanism was first considered by Oguchi and Kefer[6] who found a $T^{7/2}$ temperature dependence for $k_BT \gg \Delta$ in the ferromagnetic case. The calculation proceeds by expanding the transverse part of the hyperfine interaction in spin-wave creation ($b_k^\dagger$) and destruction (b_k) operators[7] keeping only the three-operator terms,

$$AI^+S^- \rightarrow -AI^+[(2S)^{1/2}/4S]b_1^\dagger b_2^\dagger b_3, \qquad (1)$$

where the subscripts refer to three distinct wave vectors. This process is diagrammatically represented in Fig. 1(a). The relaxation rate is then simply calculated by time-dependent perturbation theory using (1) as the perturbation Hamiltonian. It should be pointed out that there is assumed to be no correlation between nuclear spins and thus no wave-vector conservation is necessary. Furthermore, (1) is only the lowest order term in a power series expansion in $(2S)^{-1}$. To be more precise, $(4S)^{-1}$ in (1) should be replaced by $1-(1-1/2S)^{1/2}$. While such corrections may be important for $S \leq 1$, we shall restrict ourselves here to processes of lowest order in $(2S)^{-1}$. We have extended[3] this treatment to antiferromagnets where there are now four distinct three-magnon processes [illustrated in Fig. 1(b)] involving combinations of spin waves from the two branches of the spectrum and find the aforementioned T^5 dependence on the high-temperature regime. Of course, any processes such as these which involve thermal excitations have rates which fall exponentially to zero for $k_BT < \Delta$.

The purpose of this note is to point out that such calculations may severely underestimate the three-magnon process, i.e., there exist other mechanisms involving identical initial and final states in the scattering process which interfere constructively with the previously discussed matrix elements and thus may enhance the corresponding rates, essentially preserving their temperature and field dependences. In particular, we consider the following second-order process, Fig. 2: (1) A nu-

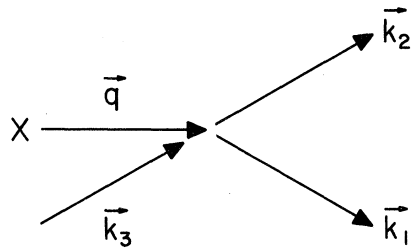

FIG. 2. The second-order process, where the vertex at the right corresponds to the dynamical exchange scattering, Eq. (2). Conservation of wave vector in the exchange scatter requires $\vec{q}=\vec{k}_1+\vec{k}_2-\vec{k}_3$.

clear spin flips creating a single virtual magnon, the matrix element again arising from AI^+S^-; (2) this virtual spin wave is then scattered by a thermal magnon via the dynamical spin-wave interaction[8] which in a ferromagnet is given in the long-wavelength limit by

$$\mathcal{H}_{ex}=\frac{Jza^2}{12}\sum_{k_1,k_2,k_3,k_4} b_1^{\dagger}b_2^{\dagger}b_3b_4 \times(\vec{k}_1\cdot\vec{k}_2+\vec{k}_3\cdot\vec{k}_4)\delta(\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4), \quad (2)$$

where J is the nearest-neighbor exchange energy, z the number of nearest neighbors, and a is the lattice constant. The effective matrix element for this second-order process which interferes with (1) is then

$$\mathcal{H}_{\text{eff}}=-\frac{(2S)^{1/2}}{6}\frac{AI^+Jza^2}{E(\vec{k}_1+\vec{k}_2-\vec{k}_3)}b_1^{\dagger}b_2^{\dagger}b_3 \times[\vec{k}_1\cdot\vec{k}_2+\vec{k}_3\cdot(\vec{k}_1+\vec{k}_2-\vec{k}_3)], \quad (3)$$

where $E(\vec{k})=\frac{1}{3}Jza^2k^2$ is the energy of a spin wave of wave vector $\vec{k}$. An investigation of (3) shows that it is essentially independent of J and of the same order as (1) in A and $(2S)^{-1}$; in fact, the effect of (3) is approximately to multiply the matrix element (1) by a factor of two and thus enhance the relaxation rate by a factor of four. This type of effect can, of course, also occur in any ordered magnetic system such as antiferromagnets or metamagnets where there exist more than one branch to the spin-wave spectrum. In these cases, several combinations of exchange and hyperfine effects can give rise to matrix elements which interfere with each of the processes diagrammed in Fig. 1(b) and thus even stronger enhancements of the relaxation rate may be expected in such systems.

It has been further pointed out by Holstein[9] that repeated exchange scatterings would give rise to more interference terms and would thus seem to enhance the rate further. In fact, the matrix element for each repeated exchange scattering is of lower order than the preceding one by $(2S)^{-1}$. Thus we have a power series in $(2S)^{-1}$ which presumably is rapidly converging for $S>1$ but may severely modify the result for $S=\frac{1}{2}$.

In a further more comprehensive publication we shall give a detailed investigation of these repeated exchange scatters together with a complete study of the enhancement in antiferromagnetic systems.

In conclusion, we should mention that the effect discussed here was already predicted in 1961, in a general way, by Winter[10] who pointed out that the direct process may obtain if the power spectrum of the spin wave is sufficiently lifetime broadened to include transverse spin fluctuations at the nuclear resonance frequency. It is evident that our exchange-scattering process is a concrete example of such an effect.

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