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# Ferromagnetic Relaxation. II. The Role of Four-Magnon Processes in Relaxing the Magnetization in Ferromagnetic Insulators*

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The role of four-magnon processes in relaxing the uniform precession, in relaxing magnons of nonzero wave vector in parallel pumping experiments, and in establishing thermal equilibrium is considered. It is shown that the effect of the exchange-induced four-magnon process should be observable in parallel pumping experiments, but it is found that the four-magnon processes arising from dipole, cubic anisotropy, and exchange coupling are not very effective in relaxing the uniform precession. Both three- and four-magnon processes are important in the later stages of the relaxation scheme by which thermal equilibrium is established.

## INTRODUCTION

IN the first paper[1] of this series, a scheme was proposed for the relaxation process accompanying excitation of uniform precession magnons in a ferromagnetic resonance experiment, with particular reference to highly pure yttrium iron garnet (YIG). The linewidth and relaxation time of the total $z$ component of magnetization calculated in I were in reasonable agreement with experiment. There remain the problems of explaining the direct relaxation of the uniform precession to modes other than the degenerate spectrum and of considering the later stages of relaxation scheme.

Kasuya and LeCraw[2] have recently estimated relaxation frequencies for a number of processes which directly relax the uniform precession. They suggest that the relaxation frequency calculated for the three-magnon process and the two-magnon–one-phonon process, both arising from the uniaxial anisotropy Hamiltonian, could explain the experimental results. The ferrimagnetic nature of YIG is considered in their estimates.

In an earlier paper[3] Kasuya gave a qualitative treatment of four-magnon dipole and pseudodipole processes. Morkowski,[4] using the Dyson treatment of spin waves, calculated the relaxation frequency for the uniform precession for the four-magnon pseudodipole process. He found that the relaxation frequency varies as $T^4$ rather than $T^2$ as found by Kasuya. However, there is a slight error in one of Morkowski's final integrations and, indeed, the correct temperature dependence of his result is $T^2$. Several workers[5] have considered the role of four-magnon processes in establishing thermal equilibrium in the spin system.

In the present paper we first report the results of calculations of the relaxation frequency of the uniform precession directly by four-magnon processes. It is found that four-magnon processes are not very effective in relaxing the uniform precession. We then consider the relaxation of magnons of nonzero wave vector by four-magnon exchange processes. It is shown that the effect of the four-magnon process should be experimentally observable in parallel pumping experiments.[6–8] The final stages of the relaxation processes which establish thermal equilibrium are then discussed very briefly.

## DIRECT RELAXATION OF UNIFORM PRECESSION BY FOUR-MAGNON PROCESSES

In modulation experiments[9] and in parallel pumping experiments[10] by LeCraw, Fletcher, and Spencer, the direct relaxation of the uniform precession to modes other than the degenerate spectrum was measured. In reference 2 the following properties of this direct-relaxation frequency $1/T_0$ are reported: (1) The magnitude of $1/T_0$ is $4\times10^6$ sec$^{-1}$ at room temperature and a frequency of 9.34 kMc/sec. (2) In the range 150°K to 400°K, $1/T_0$ is proportional to $T^n$ with $1\leqq n\leqq 2$, the larger values of $n$ corresponding to the higher temperatures. (3) At room temperature $1/T_0$ is proportional to frequency for $\nu\geqq 3$ kMc/sec and is inversely proportional to the saturation magnetization. (4) Finally, the modulation experiments indicate that the $z$ component of the total magnetization is relaxed in the direct processes.

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[1] M. Sparks, R. Loudon, and C. Kittel, Phys. Rev. **122**, 791 (1961), hereafter referred to as I.
[2] T. Kasuya and R. C. LeCraw, Phys. Rev. Letters **6**, 223 (1961).
[3] Tadao Kasuya, Progr. Theoret. Phys. (Kyoto) **12**, 802 (1954).
[4] Janusz Morkowski, Acta Phys. Polon. **19**, 3 (1960).
[5] See the review paper by A. I. Akhiezer, V. G. Bar'yakhtar, and M. I. Kaganov, Soviet Phys.—Uspekhi **3**, 661 (1961).
[6] F. R. Morgenthaler, J. Appl. Phys. **31**, 95S (1960) and Doctoral Thesis Proposal, Massachusetts Institute of Technology (1959), unpublished.
[7] M. I. Kaganov and V. M. Tuskernik, Soviet Phys.—JETP **37**, 823 (1959); **37**, 582 (1960).
[8] E. Schlömann, J. J. Green, and U. Milano; J. Appl. Phys. **31**, 386S (1960).
[9] R. C. Fletcher, R. C. LeCraw, and E. G. Spencer, Phys. Rev. **117**, 955 (1960).
[10] These parallel pumping experiments are discussed briefly in reference 2 and will be discussed in detail in a paper in preparation by R. C. LeCraw.

For the direct relaxation of the uniform precession, the four-magnon processes which we treat are, first, the three-in and one-out process which arises from the dipole interaction; second, the two-in and two-out dipole process; and third and fourth, the two-in and two-out and three-in and one-out processes, respectively arising from the cubic anisotropy Hamiltonian. The three-in and one-out process relaxes $M_z$, the $z$ component of magnetization, as required to explain (4) above. The two-in and two-out process does not relax $M_z$ directly; however, if the output magnons from this process relax very rapidly by a process which relaxes $M_z$, and this is probably the case, the net effect of the two processes is a relaxation of $M_z$ in a time determined by the two-in and two-out process. The exchange interaction cannot relax the uniform precession, since the exchange Hamiltonian commutes with the total magnetization or with $M_z$, and at least one of these changes in any process which relaxes the uniform precession.

### DIPOLE RELAXATION

We consider first the three-in and one-out process. On transforming the dipole Hamiltonian to magnon creation and destruction operators[11] $a^\dagger$ and $a$ by the standard method, we find for the three-in and one-out term

$$\mathcal{H}_{3-1}=\sum_{\mu\nu\sigma}\sum_{\lambda}{}' C_{\lambda\mu\nu\sigma}a_\lambda^\dagger a_\mu a_\nu^\dagger a_\sigma^\dagger+\text{c.c.}, \tag{1}$$

where c.c. denotes Hermitian conjugate, the prime on the summation signifies omit $k_\lambda=0$, and

$$C_{\lambda\mu\nu\sigma}=-(2\pi\mu^2/V)(k_\lambda^+/k_\lambda)^2\Delta(\mathbf{k}_\lambda+\mathbf{k}_\nu+\mathbf{k}_\sigma-\mathbf{k}_\mu).$$

The notation is the same as that used in I.

In a rather long calculation, using the standard transition probability method, we find for the contribution $1/T_{3-1}$ to $1/T_0$

$$1/T_{3-1}=(28\mu^4/15\pi\hbar D^3)(k_BT)^2, \tag{2}$$

where $\mu$ is the Bohr magneton and $D$ defines the high-frequency dispersion relation $\hbar\omega=Dk^2$. Taking[12] $D=9\times10^{-29}$ erg cm$^2$ and $T=300$°K, we get

$$1/T_{3-1}=1.0\times10^3\ \text{sec}^{-1}. \tag{3}$$

Similarly, we find that the relaxation frequency for the two-in and two-out dipole process has the same dependence on temperature, frequency, and saturation magnetization and is of the same order of magnitude as (2). Thus the total contribution of the four-magnon dipole processes to the relaxation frequency is of the order of two times the value of $1/T_{3-1}$ given by (2). This value is three orders of magnitude smaller than the observed value of relaxation frequency and does not have the observed dependence on frequency or on saturation magnetization. Thus, we conclude that the four-magnon dipole processes are not effective in relaxing the uniform precession.

### ANISOTROPY RELAXATION

We now consider the contribution to four-magnon relaxation of the uniform mode arising from the magnetocrystalline anisotropy energy. Since the average environment of the $Fe^{3+}$ ions in YIG has cubic symmetry, there is a term in the spin Hamiltonian of the form

$$\mathcal{H}_A=K\sum_i(S_{\eta i}^4+S_{\zeta i}^4+S_{\xi i}^4),$$

where $\eta$, $\zeta$, and $\xi$ are the cubic crystal axes. The anisotropy constant $K$ is determined from Rodrigue *et al.*[13] to be about $1.8\times10^{-3}$ cm$^{-1}$ for YIG when treated as a ferromagnet. Kasuya and LeCraw[2] consider the more detailed uniaxial surroundings of the $Fe^{3+}$ ions by using a two-sublattice model. Then, each sublattice has uniaxial terms in its Hamiltonian of the form $ES_{\eta i}^2$ which tend to cant the individual spins. From this larger interaction, $E\approx10^{-1}$ cm, arises the three-magnon terms calculated by Kasuya and LeCraw. When describing YIG by one sublattice, these uniaxial terms average to a constant.

In the usual way, we expand $\mathcal{H}_A$ in creation and annihilation operators. If we let $\alpha_\eta$, $\alpha_\zeta$, $\alpha_\xi$ be the direction cosines of the magnetization with respect to the crystal axes, the four-magnon part of $\mathcal{H}_A$ which relaxes $S^2$ is

$$\mathcal{H}_{2-2}=\frac{KS^2}{2N}[45(\alpha^4+\alpha_\zeta^4+\alpha_\xi^4)-27]\times\sum_{\lambda,\mu,\nu,\sigma}a_\lambda^\dagger a_\mu^\dagger a_\nu a_\sigma\Delta(\mathbf{k}_\lambda+\mathbf{k}_\mu-\mathbf{k}_\nu-\mathbf{k}_\sigma).$$

A transition-probability calculation gives for this process

$$1/T_{2-2}\cong(KS^2V/2\pi N)^2[(k_BT)^2/4\pi\hbar D^3]\times[45(\alpha_\eta^4+\alpha_\zeta^4+\alpha_\xi^4-27]^2, \tag{4}$$

where $V$ is the volume of the sample. For YIG, $N/V\approx0.6\times10^{22}/\text{cm}^3$, $K\approx1.8\times10^{-3}$ cm$^{-1}$, $S=5/2$, $D\approx9\times10^{-29}$ erg cm$^2$. Then at 300°K, along the easy [111] axis, $T_{2-2}^{-1}$ is about $10^5$ sec$^{-1}$, still an order of magnitude smaller than the total experimental value.

When the sample is not magnetized along a symmetry axis, the cubic anisotropy also gives rise to three-one processes, which relax $M_z$ directly. In a similar way this relaxation rate $T_{3-1}^{-1}$ may be calculated to give

$$1/T_{3-1}\cong(17KS^2V/2\pi N)^2\times[(k_BT)^2/8\pi\hbar D^3][1+(\alpha_\eta^4+\alpha_\zeta^4+\alpha_\xi^4)^2-2(\alpha_\eta^4+\alpha_\zeta^4+\alpha_\xi^4)-12\alpha_\eta^2\alpha_\zeta^2\alpha_\xi^2]. \tag{5}$$

Note that $T_{3-1}^{-1}$ vanishes along the easy [111] axis. Along other directions $T_{3-1}$ is of the same order of magnitude as $T_{2-2}$. Thus, we conclude that four-magnon

[11] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).
[12] E. H. Turner, Phys. Rev. Letters **5**, 100 (1960).
[13] G. P. Rodrigue, H. Meyer, and R. V. Jones, Suppl. J. Appl. Phys. **31**, 376S (1960).

anisotropy relaxation is not sufficiently rapid to make a major contribution to the total relaxation rate.

### RELAXATION OF MAGNONS WITH LARGE WAVE VECTOR BY FOUR-MAGNON EXCHANGE PROCESS

In I the relaxation frequency of magnons as a function of wave-vector amplitude $k$ was calculated for three-magnon dipolar processes. For the confluence process, in which two magnons are destroyed and one is created, the relaxation frequency was found to be proportional to $k$ for small $k$ and inversely proportional to $k$ for large $k$, with a maximum between these two regions. The splitting process, in which the input magnon splits into two magnons, relaxes only magnons with wave amplitude greater than the threshold value $k_{td}=(2\hbar\omega_0/D)^{\frac{1}{2}}$. For values of wave vector slightly greater than this threshold, the relaxation frequency increases rapidly from zero as $\lambda$ increases; for further increase in $k$ the relaxation frequency goes through a maximum, and for large $k$ is proportional to $k^{-1}\log k$. The relaxation frequency is linear in temperature for both the splitting and confluence processes. Figure 1 of I represents these results, which are valid in the high-field, high-temperature limit.

We now consider the effect of the four-magnon exchange process on the magnon relaxation frequency, comparing the result with the three-magnon results. Dyson's[14] four-magnon exchange result may be written in terms of a relaxation frequency

$$(1/T_{1k})_{4e}=[2\zeta(\tfrac{3}{2})D/\hbar](\mu/M_s)^2(k_BT/4\pi D)^{\frac{5}{2}}k^3, \quad (6)$$

where $\zeta(\frac{3}{2})$ is the Reimann zeta function of argument $\frac{3}{2}$. With $D=9\times10^{-29}$ erg cm$^2$ and $M_s=140$ gauss at 300°K, this reduces to the convenient form

$$(1/T_{1k})_{4e}=1.6\times10^7[M_s(300)/M_s(T)]^2 \times(k/10^6)^3(T/300)^{\frac{5}{2}}\ \mathrm{sec}^{-1}. \quad (7)$$

Comparing this result with the three-magnon results described above, it is seen that the four-magnon exchange relaxation frequency increases more rapidly with increasing $k$ than that of the three-magnon process and also increases more rapidly with temperature than the three-magnon process. Thus the four-magnon process becomes important for some large value of $k$, this value of $k$ being a function of temperature and of the applied magnetic field. Let us now examine this relative importance of the three- and four-magnon processes by comparing (7) with the three-magnon results (3.7) and (C.7) of I. First, for $\omega_0=2\omega_s/3$, where $\omega_0$ is the applied magnetic field in frequency units, and $\omega_s=4\pi\gamma M_s$, and $T=300$°K, the two contributions to the relaxation frequency are equal when $k=7\times10^5$ cm$^{-1}$, and the threshold wave vector for the splitting process is also at $k_{td}=7\times10^5$ cm$^{-1}$. Thus the $k$ dependence of the relaxation frequency should be the following: The three-magnon confluence process is dominant at low values of $k$, giving a linear $k$ dependence as $k$ increases from zero, then for larger values of $k$ there is a bending down of the relaxation frequency toward the $k$ axis. This behavior has been verified[15] in parallel pumping experiments by one of us (R. C. L.). Before the relaxation frequency goes through the maximum, the four-magnon contribution becomes large enough to cause a bending up of the curve away from the $k$ axis.

It is unlikely that the sharp rise in relaxation frequency at $k=k_{td}$ due to the onset of the splitting process will be observable in the presence of the four-magnon process at room temperature. In order to observe this sharp increase, the temperature must be lowered below room temperature to reduce the contribution from the four-magnon exchange process. We estimate the required temperature by calculating the temperature at which $(1/T_{1k})_{4e}$ equals one-fourth the confluence value for $k=k_{td}$. In order to put the microwave pump frequency within the experimentally available range, we again take $\omega_0=2\omega_s/3$. Lower applied fields should be avoided in this experiment, for correction associated with the noncircular precession of the spins must then be applied. With $k=k_{td}$ the microwave pump frequency $\omega_{\mathrm{pump}}=2(Dk^2+\hbar\omega_0)$ is 30 kMc/sec, a reasonable value. From (7) above and (37) of I with $k=k_{td}$, we find that $(1/T_{1k})_{4e}$ equals one-fourth the confluence value for $T=120$°K; the effect of the splitting process should be easily observable at this temperature.

### ESTABLISHING A SPIN TEMPERATURE

In I a complete scheme of relaxing the magnetization in low-temperature resonance experiments was proposed. The uniform precession mixes with the degenerate $S$ through the dipole depolarization field of small pits left on the surface of the sample by the polishing process. The relaxation frequency for this process determines the resonance linewidth. The $S$ magnons are relaxed by the three-magnon dipole process in a time which governs the rate of relaxation of $M_z$. The output magnons of this Raman process, which we call subthermal magnons, then relax by additional three-magnon processes on four-magnon processes, the relative importance of the two processes depending on the temperature. The output magnons of these processes relax in turn by three- and four-magnon processes, etc., until thermal equilibrium is established in the spin system. The spin temperature will be somewhat higher than the lattice temperature, the temperature difference depending on the speed of the magnon-photon processes.

In I the relaxation frequencies of the first two processes in this sequence were calculated and found to be in reasonable agreement with experimental linewidth and $M_z$ time. One of us (M. S.) has considered[16] the

[14] Freeman J. Dyson, Phys. Rev. **102**, 1217 (1956).

[15] See Fig. 1 of reference 2.

[16] M. Sparks, Doctoral thesis, University of California, 1961 (unpublished).

subsequent processes which are dominant in establishing a spin temperature. It is shown that in the YIG resonance-modulation experiments,[9] the subthermal magnons are relaxed more effectively by the four-magnon exchange process than by the three-magnon processes for temperatures above $\sim 50°$K. In general, both the three-magnon dipole and the four-magnon exchange processes are important in the subsequent processes leading to a spin temperature. Finally, it is shown that these three- and four-magnon processes are more effective than the two-magnon–one-phonon processes, induced by magnetoelastic and exchange interactions, in relaxing all magnons which are important in the resonance experiments. These important magnons include the $S$ magnons, the subthermal magnons, and the thermal magnons. This justifies the assumption made[5,16] in magnon-phonon calculations that a spin temperature exists. The calculations[5] of a magnon-magnon relaxation frequency averaged over all magnons to justify the assumption of a spin temperature is not sufficient for the relaxation scheme proposed in I.

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# Use of Thin Films for the Study of Stress Effects on the Superconducting Transition of Indium

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Measurements on evaporated thin films of superconductors provide a powerful technique for studying stress effects on the superconductivity transition. Utilization of the differential thermal expansion of film and substrate by suitable choice of substrate and substrate temperature during evaporation provides a convenient way for controlling the stress in the films. In this manner, uniform tensile and compressive stresses of substantial magnitude can be obtained and are tolerable because of the great strength of thin films. To illustrate how this technique can be used, studies are shown of similarity effects in indium films under the influence of biaxial stress and a generalization of the usual similarity relations is suggested.

It is the purpose of this paper to suggest that studies of evaporated thin films of superconductors provide a powerful technique for studying stress effects on the superconductivity transition. Utilization of the differential thermal expansion of film and substrate by suitable choice of substrate and substrate temperature during evaporation provides a convenient way for controlling the stress in the films. In this manner, uniform tensile and compressive stresses of substantial magnitude can be obtained and are tolerable because of the great strength of thin films.[1]

Table I. Specimen characteristics.

| | In 57 | In 58 |
|---|---|---|
| Substrate | fused quartz | high-expansion glass |
| Thickness | 1253 A | 1225 A |
| Residual resistivity | 0.492 $\mu$ohm-cm | 0.473 $\mu$ohm-cm |
| Critical temperature | 3.473°K | 3.437°K |
| Critical field at 0°K | 836 oersted | 840 oe |
| Tensile stress at 4°K | $2.3\times10^3$ atm | $1.0\times10^3$ atm |
| $H_c(\sigma,0)/T_c$ | 240.7 oe/°K | 244.3 oe/°K |
| Bulk critical field[a] | 285.7 oe | 285.7 oe |
| Zero-stress critical temperature[a] | 3.408°K | 3.408°K |
| Ratio of weak-field penetration depth at 0°K to film thickness | 0.600 | 0.605 |

[a] R. W. Shaw, D. E. Mapother, and D. C. Hopkins, Phys. Rev. **120**, 88 (1960).

To illustrate how this technique can be used, studies are shown of similarity effects in indium films under the influence of biaxial stress and a generalization of the usual similarity relations is suggested.

Upon the application of hydrostatic pressures to bulk superconducting samples, in general, similarity is not obeyed. The term similarity usually implies the validity of two independent conditions[2]:

$$H_c(X,t)/H_c(X,0)=f(t), \tag{1}$$

and

$$H_c(X,0)/T_c(X)=\text{const}, \tag{2}$$

where $H_c$ is the critical magnetic field, $T_c$ is the critical temperature, $t$ is the reduced temperature, $T/T_c$, and $X$ is an independent variable such as pressure, or

[1] A. M. Toxen, Phys. Rev. **123**, 442 (1961).
[2] M. Garfinkel and D. E. Mapother, Phys. Rev. **122**, 459 (1961).