

# Ferromagnetic Relaxation. I. Theory of the Relaxation of the Uniform Precession and the Degenerate Spectrum in Insulators at Low Temperatures\*

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A scheme is proposed for the relaxation processes at low temperatures accompanying excitation of uniform precession spin waves in a ferromagnetic resonance experiment, with particular reference to highly pure yttrium iron garnet. The processes are: (1) scattering of uniform precession spin waves into the degenerate spin-wave modes by polishing imperfections on the sample surface; (2) equalization of the populations of the degenerate spin-wave modes, also by surface imperfections; and (3) relaxation of the degenerate modes by Raman scattering of thermal spin waves through the magnetic dipole interaction. Relaxation times for the three processes are calculated and compared with experimental values with reasonable agreement.

WE now know that ferromagnetic relaxation is a complicated phenomenon because of the multiplicity of processes which may play a part between the initial excitation in a microwave resonance experiment of the uniform precession of the magnetization (or of spin waves of low wave vector) and the ultimate establishment of thermal equilibrium with the lattice. Many different relaxation processes have been considered in the past fifteen years, and various processes may be important under appropriate conditions. Several recent developments make it profitable for the first time to undertake a systematic quantitative discussion of the entire relaxation process under well-defined conditions. Many of the theoretical methods for treating relaxation problems were discussed earlier by various workers in the U.S.S.R., starting with Akhiezer,<sup>1</sup> for application to somewhat different conditions. We are also greatly indebted to the work of Kasuya,<sup>2</sup> who delineated the major processes affecting the relaxation of magnons of wave vectors not equal to zero.

The recent developments include:

1. The discovery by Anderson and Suhl<sup>3</sup> of the portion of the magnon spectrum degenerate with the uniform precession, and the recognition<sup>4-6</sup> of the central role the degenerate modes play in the initial stages of relaxation process in insulators.

2. The discovery<sup>7</sup> in France of yttrium iron garnet (YIG), which has the outstanding attributes of (a) cubic structure, (b) excellent insulator, (c) all magnetic ions identical and in an *S* state, and (d) no detectable

magnetic disorders. One may say that YIG is at present to ferromagnetic resonance research what the fruit fly is to genetics research.

3. The recognition<sup>8,9</sup> of the sensitivity of the relaxation observations to the presence of trace amounts of impurity atoms which are themselves rapidly relaxed by spin-lattice mechanisms. In the present paper we are concerned with intrinsic and geometrical relaxation processes, rather than with the processes associated with rapidly relaxing impurities.

4. The prediction by Kaganov and Tsukernik,<sup>10</sup> Morgenthaler,<sup>11</sup> and Schlömann, Green, and Milano<sup>11</sup> of direct nonresonant microwave excitation of magnons having well-defined wave vectors.

5. The experimental program of LeCraw and Spencer<sup>12</sup> and associates, in which a number of individual relaxation processes in YIG have been studied under a wide range of conditions.

In the present paper we discuss aspects of the theory of the initial stages of relaxation in pure ferromagnetic insulators at low temperatures ( $T \ll T_c$ ) with particular reference to yttrium iron garnet. By initial stages we mean, first, the mixing or scattering of the uniform precession with the degenerate spectrum and, second, the interaction of the degenerate spectrum with the thermal magnons by processes in which not more than three magnons are involved.<sup>13</sup>

We do not consider here the intrinsic processes (such as the 4-magnon dipolar process) which relax the

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<sup>1</sup> A. I. Akhiezer, *J. Phys. U.S.S.R.* **10**, 217 (1946). For more recent references see A. I. Akhiezer, V. G. Bar Yakhtar, and S. V. Peletninskii, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **36**, 216 (1959) [translation: *Soviet Phys.—JETP* **36**, 146 (1959)].

<sup>2</sup> T. Kasuya, *Progr. Theoret. Phys. (Kyoto)* **12**, 802 (1954).

<sup>3</sup> P. W. Anderson and H. Suhl, *Phys. Rev.* **100**, 1788 (1955).

<sup>4</sup> A. M. Clogston, H. Suhl, L. R. Walker, and P. W. Anderson, *J. Phys. Chem. Solids* **1**, 129 (1956).

<sup>5</sup> H. B. Callen, *J. Phys. Chem. Solids* **4**, 256 (1958).

<sup>6</sup> H. B. Callen and E. Pittelli (to be published).

<sup>7</sup> F. Bertaut and F. Forrat, *Compt. rend.* **242**, 382 (1956); S. Geller and M. A. Gillo, *Acta Cryst.* **10**, 239 (1957).

<sup>8</sup> C. Kittel, *Phys. Rev.* **115**, 1587 (1959); P.-G. de Gennes, C. Kittel, and A. M. Portis, *Phys. Rev.* **116**, 323 (1959).

<sup>9</sup> R. L. White, *Phys. Rev. Letters* **2**, 465 (1959).

<sup>10</sup> M. I. Kaganov and V. M. Tsukernik, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **37**, 823 (1959) [translation: *Soviet Phys.—JETP* **37**, 582 (1960)].

<sup>11</sup> F. R. Morgenthaler, *J. Appl. Phys.* **31**, 95S (1960), and doctoral thesis proposal, Massachusetts Institute of Technology, 1959 (unpublished); E. Schlömann, J. J. Green, and U. Milano, *J. Appl. Phys.* **31**, 386S (1960).

<sup>12</sup> See, for example, E. G. Spencer and R. C. LeCraw, *Phys. Rev. Letters* **4**, 130 (1960); R. C. Fletcher, R. C. LeCraw, and E. G. Spencer, *Phys. Rev.* **117**, 955 (1960); also several papers presently in preparation.

<sup>13</sup> A short summary of the calculation for the three-magnon relaxation process was made by M. Sparks and C. Kittel, *Phys. Rev. Letters* **4**, 232, 320 (1960).

uniform mode directly. A subsequent paper will discuss several of these processes in detail.

#### MIXING OF THE UNIFORM PRECESSION WITH THE DEGENERATE SPECTRUM BY SURFACE IMPERFECTIONS

In a single-crystal spherical specimen of pure YIG at sufficiently low temperatures the principal contribution to the width of the uniform mode ferromagnetic resonance line is believed to come from surface imperfections as shown by LeCraw, Spencer, and Porter.<sup>14</sup> This relaxation process is a two-magnon process: It involves the destruction ( $\Delta n_0 = -1$ ) of a uniform mode magnon and the creation of a magnon ( $\Delta n_s = +1$ ) in the degenerate spectrum, containing what we shall refer to as  $S$  magnons or Suhl-Anderson magnons. The number of magnons remains constant in the scattering process, and therefore the total  $z$  component  $S_z$  of spin is constant, but the destruction of a  $\mathbf{k}=0$  magnon decreases the total spin  $S$  by one unit. The scattering process is therefore a relaxation process only for the transverse component of the total magnetic moment and may be specified as a relaxation time  $T_{20}$ . The surface pits are important in causing two spin-wave collisions, but can be shown not to make any significant contribution to the three-spin-wave collision interaction. The linewidth associated with the pits may be in the range 0.1 to 50 oersteds, according to the polish.

Many other static interactions besides surface pit scattering contribute to the elastic scattering which converts uniform magnons into degenerate  $S$  magnons, but all of these contributions in YIG appear to be small. We have calculated relaxation times for a number of point processes by standard methods, similar to those employed by Callen and Pittelli.<sup>6</sup>

(1) Vacancies and diamagnetic ions in ferric ion sites. Controlled experiments by Spencer and LeCraw<sup>15</sup> show that the substitution of vast amounts of diamagnetic  $\text{Ga}_2\text{O}_3$  for  $\text{Fe}_2\text{O}_3$  in YIG produces a remarkably small contribution to the linewidth, of the order of 0.1 oersted per atomic percent Fe sites occupied by Ga. The value calculated for magnetic dipole interactions is of the order of  $5 \times 10^{-4}$  oe per atomic percent Ga, and this width is negligible. The pseudodipolar interaction constant is presumably of the order of  $(\Delta g/g)^2 J$  and should not be greater than  $10^{-20}$  erg for  $(\Delta g/g)^2 \approx 10^{-6}$  and may be neglected. Pincus, in unpublished work, has shown that the effect on the linewidth of the Dzialoshinski-Moriya interaction is also negligible in YIG.

(2) Hyperfine interaction from  $\text{Fe}^{57}$  nuclei. The relaxation time is estimated to be  $\sim 10^3$  sec, or  $10^{-4}$  oe.

(3) Adsorbed molecular oxygen. The linewidth asso-

ciated with the dipole moment of static electronic paramagnetic impurities may be roughly estimated to be of the order of  $10^{-4}f$  oe, where  $f$  is the fraction of the total lattice sites thus occupied. We may note, however, that if adsorbed paramagnetic moments on the surface are rapidly relaxing in the sense of reference 7, then their effects may be on the limits of detectability.

(4) Pinned spins. It may be possible for the spin directions of certain random ferric ions to be frozen in direction either by strong exchange coupling with a paramagnetic ion having a large crystal field splitting with a nondegenerate ground state, or by (with the aid of the Meiklejohn-Bean effect)<sup>16</sup> exchange interaction with an antiferromagnetic region. It is hard to say anything very specific about such effects without using an *ad hoc* model, but a pinning field of  $10^5$  oe would give a linewidth in YIG of the order of 1 oe per atomic percent pinning sites, which would make such defects the most important static *point* defects for the present problem.

The specimens of YIG used in the ferromagnetic resonance experiments are spherical, having been polished into this shape by the Bond process. Examination reveals that the surface is covered with pits having diameters about two-thirds the diameter of the grit of the polish used. The experimental ferromagnetic resonance linewidths  $\Delta H$  for several sizes of polishing grit show that  $\Delta H$  increases approximately linearly with the radius  $R$  of the surface pits on the sample;  $\Delta H$  was observed over a range of about 0.5 to 10 oe. We now calculate the linewidth for the pit scattering process. The result has approximately the correct order of magnitude and depends linearly on the pit radius.

We treat in the Born approximation the problem of scattering by a small spherical cavity in an infinite medium, and later we interpret the result in terms of surface pits. The scattering potential  $V$  which arises from the interaction of the magnetostatic field  $\mathbf{H}_c$  of the cavity and magnetization  $\mathbf{M}$  is

$$V_c = -\frac{1}{2} \int \mathbf{H}_c \cdot \mathbf{M} d\mathbf{r}, \quad (1)$$

where the factor  $\frac{1}{2}$  is appropriate to a self-energy.

The Holstein-Primakoff transformation of the magnetization to spin-wave operators correct to second order is

$$M^+(\mathbf{r}) = (4\mu M_s/V)^{\frac{1}{2}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}} a_{\mathbf{k}}; \quad (2)$$

$$M^z(\mathbf{r}) = M_s - (2\mu/V) \sum_{\mathbf{k}_1, \mathbf{k}_2} e^{i(-\mathbf{k}_1 + \mathbf{k}_2) \cdot \mathbf{r}} a_{\mathbf{k}_1}^\dagger a_{\mathbf{k}_2}; \quad (3)$$

here  $\mu$  is  $g|\mu_B|/2$ , and  $V$  is the volume of the specimen. This representation does not diagonalize the magnetic dipolar energy, but the approximation should be adequate provided  $H > \sim 2\pi M_s$ . The macroscopic sig-

<sup>14</sup> R. C. LeCraw, E. G. Spencer, and C. S. Porter, Phys. Rev. **110**, 1311 (1958); see also reference 5. Further relevant experiments are reported by A. G. Gurevich and I. E. Gubler, Soviet Physics—Solid State **1**, 1693 (1960).

<sup>15</sup> E. G. Spencer and R. C. LeCraw, Bull. Am. Phys. Soc. **5**, 58 (1960).

<sup>16</sup> W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956).

nificance of this approximation is that the precessional motion within a spin wave is taken to be circular, whereas it is actually elliptical. The terms in  $V_c$  which relax the uniform mode are, as derived in Appendix A,

$$V_c(0, \mathbf{k}) = 16\pi^2 R^3 (\mu M_s / V) (3 \cos^2 \theta_k - 1) \times [j_1(kR)/kR] (a_0 a_{\mathbf{k}}^\dagger + a_0^\dagger a_{\mathbf{k}}), \quad (4)$$

where  $R$  is the radius of the cavity and  $j_1$  is a spherical Bessel function. It is convenient to write

$$V_c(0, \mathbf{k}) = F(\mathbf{k}, R) (a_0 a_{\mathbf{k}}^\dagger + a_0^\dagger a_{\mathbf{k}}), \quad (5)$$

so that, if the occupation numbers  $n_{\mathbf{k}}$  (for  $k \neq 0$ ) of the  $S$  magnons are all equal to some value  $\bar{n}_0$ , the kinetic equation for  $n_0$  is

$$dn_0/dt = -(n_0 - \bar{n}_0)/T_{20}, \quad (6)$$

where the transverse relaxation time  $T_{20}$  is given by

$$\frac{1}{T_{20}} = \frac{2\pi}{\hbar} \int_0^{k_m} |F(\mathbf{k}, R)|^2 \rho_k dk, \quad (7)$$

where  $\rho_k dk$  is the number of states per unit energy range with  $|\mathbf{k}|$  in  $dk$  at  $k$ . The dispersion relation for the  $S$  magnons in a sphere is

$$\omega^2 = \gamma^2 [H_0 - (4\pi M_s/3) + (D/\hbar\gamma)k^2] \times [H_0 - (4\pi M_s/3) + (D/\hbar\gamma)k^2 + 4\pi M_s \sin^2 \theta_k]. \quad (8)$$

Here  $D$  is the exchange constant and  $\theta_k$  is the angle between  $\mathbf{k}$  and the  $z$  axis;  $H_0$  is the external applied magnetic field; in (7) the upper limit  $k_m$  is given by  $(D/\hbar\gamma)k_m^2 = 4\pi M_s/3$  and has the value  $k_m \sim 4 \times 10^5 \text{ cm}^{-1}$  in YIG. The evaluation of the density of states for (8) is fairly complicated (reference 5). For  $H_0 \gg 4\pi M_s/3$  and  $k_m R \gg 10$ ,

$$\frac{1}{T_{20}} \cong \frac{16\pi^2}{\hbar} R^3 (\mu M_s / V) \frac{(3 \cos^2 \theta_0 - 1)^2}{\cos \theta_0} \int_0^\infty [j_1(kR)]^2 d(kR). \quad (9)$$

Here  $\theta_0$  is the value of  $\theta$  satisfying (8) for  $k \rightarrow 0$ ; the definite integral has the value  $\pi/6$ . We now convert the result from a spherical cavity to a hemispherical pit having half the volume and situated on the surface of the specimen. We suppose plausibly that the scattering matrix elements are reduced by a factor  $\frac{1}{2}$ , of which  $\frac{1}{2}$  arises from the reduction in the effective magnetic moment of the scatterer and a further  $\frac{1}{2}$  arises from the reduced range of angular integration; thus  $1/T_{20}$  is reduced by  $(\frac{1}{2})^2$ . If we suppose that the surface of the specimen is covered entirely with pits, their number will be  $\sim 4r_0^2/R^2$ , where  $r_0$  is the radius of the specimen.

Finally, if the pits scatter independently,

$$\Delta H \approx 1/\gamma T_{20} \approx 4M_s(R/r_0)G(\theta_0), \quad (10)$$

where

$$G(\theta_0) = (3 \cos^2 \theta_0 - 1)^2 / \cos \theta_0. \quad (11)$$

The predicted dependence of  $\Delta H$  on pit radius  $R$  and specimen radius  $r_0$  is in qualitative agreement with the results<sup>14</sup> of Spencer and LeCraw and of Gurevich and Gubler. For  $H = 3300$  oe,  $M_s = 195$  gauss, and a representative (reference 14) specimen radius  $r_0 = 0.018$  cm, we have  $\theta_0 \approx 62$  deg; this estimate is based on  $D \approx 0.55 \times 10^{-28}$  erg cm<sup>2</sup>. Then, from (10),

$$\Delta H \approx 1.2 \times 10^4 R \text{ oe}. \quad (12)$$

For  $R = 5 \times 10^{-4}$  cm, we calculate  $\Delta H \approx 6$  oe from (12), which may be compared with the observed 6 oe. The agreement with experiment is better than one could expect, in view of the irregularities in the actual shape of the surface, with the possibility of superposition of fracture indentations of various shapes and sizes.

For the constants given,  $G(\theta_0)$  has a value near  $\frac{1}{8}$ . We have used this value of the angular factor in (12) above, but one should note, first, that the factor would be appreciably smaller at room temperature because the numerator of (11) will vanish at some temperature and, second, the angular factor is a consequence of our particular model of a spherical cavity and it may be modified somewhat for the distribution of pits and fractured cleavages on the actual surfaces. It is likely that quadrupolar and higher terms in (A.6) contribute significantly to the broadening, with the net effect of smoothing out the dipolar variation calculated for  $G(\theta_0)$ . Nevertheless, it would appear interesting to carry out measurements of  $\Delta H$  in pure but rough YIG under conditions such that one sweeps through the zero of  $G(\theta_0)$ .

The linewidth resulting from a single spherical cavity of volume  $v$  is

$$\Delta H \approx 2\pi^2 M_s (v/V) G(\theta_0). \quad (13)$$

It is interesting to compare this result with a linewidth calculated by Schlömann.<sup>17</sup> He assumed that the linewidth is just the width of the distribution of the fluctuations in magnetic field caused by the demagnetizing effect of the cavity, and obtained the result:

$$\Delta H \approx 6\pi M_s (v/V) / [1 + (v/V)]. \quad (14)$$

For YIG at 0°K, Eq. (13) gives  $\Delta H \approx 4M_s v/V$ , so that the linewidth given by spin-wave theory is smaller by a factor of about 5 than that obtained when correlation between the spins is neglected. We have here an example of the dipolar narrowing which occurs when the linewidth is due to a perturbation causing field fluctuations with a period large compared to the lattice constant but small compared to sample dimensions.<sup>18,19</sup>

#### RELAXATION OF DEGENERATE S MAGNONS BY THREE-MAGNON DIPOLAR PROCESSES

A preliminary account of the first calculation of this section has appeared.<sup>13</sup> The calculation is concerned

<sup>17</sup> E. Schlömann, Proceedings of the Boston Magnetism Conference, 1956 (unpublished), p. 600.

<sup>18</sup> S. Geschwind and A. M. Clogston, Phys. Rev. **108**, 49 (1957).

<sup>19</sup> A. M. Clogston, J. Appl. Phys. **29**, 334 (1958).

with the results reported by Spencer and LeCraw<sup>12</sup> for the relaxation of the magnetic-moment component parallel to the static magnetic field, in ferromagnetic resonance experiments on highly purified YIG at low temperatures. It is appropriate, with reference to the Bloch-Bloembergen equations, to denote this relaxation time by  $1/T_1$ . The best experimental estimate<sup>12</sup> of  $1/T_1$  at 9.34 kMc/sec between 2 and 30°K is fitted roughly, after correction for what may be impurity relaxation, by

$$1/T_1 \cong 1 \times 10^5 T \text{ sec}^{-1}, \quad (15)$$

with  $T$  in deg K; below 5°K the curve appears to flatten out above the values given by this relation. The result is roughly independent of pit radius.

It is possible to relax the uniform precession directly, as by intrinsic four-magnon dipolar (*not* exchange) processes, but such processes have a rate proportional to  $T^2$  and will be slower than the observed rate at sufficiently low temperatures, particularly in view of the low magnetoelastic coupling believed to characterize materials such as YIG which contain only  $S$ -state ions ( $\text{Fe}^{3+}$ ). The three-magnon dipolar process discussed below does not relax the uniform precession, but does relax the  $S$  magnons in the degenerate spectrum at a rate directly proportional to  $T$  and with an absolute value in quite fair agreement with (15).

Some general assumptions should be noted. We give results only for a spherical specimen, because this is the only geometry used so far in the more relevant experiments. We assume that all magnons involved have the form of uniform plane waves; this is not always true<sup>20</sup> at wave vectors below  $\sim 10^4 \text{ cm}^{-1}$ . We have not made the second Holstein-Primakoff transformation on the spin operators which diagonalizes all the quadratic terms in the Hamiltonian; this transformation takes account of the elliptical motion of the local magnetization in a spin wave—we treat this motion as circular, although we do use the exact dispersion relation. The relevant correction has been studied by Schlömann<sup>21</sup> and at most amounts to 4% in YIG at  $K$  band and 5% at  $X$  band. We also neglect spin pinning at the surface. We assume that the exchange constant  $D$  is independent of the state of excitation of the spin system. Keffer and Loudon<sup>22</sup> have shown that Dyson's criterion for the validity of this assumption has a simple, firm, and cogent physical basis. We assume further that the mean free path  $\Lambda$  of all magnons involved is long in comparison with the wavelength  $\lambda$  of the incident magnon.<sup>23</sup> In the 3-magnon process the thermal magnons which have the most important effect in relaxing microwave magnons  $k_1$  turn out to be those having  $k_2 \approx \hbar\omega_1/2Dk_1 \sim 10^6$  to  $10^7 \text{ cm}^{-1}$ . These have a group velocity of  $2 \times 10^5$  to  $2 \times 10^6 \text{ cm/sec}$  and will have a mean free path  $> 2 \times 10^{-4} \text{ cm}$  provided that  $\tau > 10^{-9} \text{ sec}$ .

The inequality  $\Lambda > \lambda$  appears to be satisfied in most contemporary experiments on pure YIG at room temperatures or below, except for the uniform mode. The 4-magnon exchange process is, at high temperatures, a promising vehicle for the relaxation of the relevant thermal magnons.

We neglect the ferrimagnetic nature of YIG and treat it as a ferromagnet having the same macroscopic properties; we neglect the optical branches of the magnon spectrum. Several workers have suggested that ferrimagnetic effects are important above the low-temperature region.

In the three-magnon process the magnetostatic field of an  $S$  magnon couples it to the magnetization of a thermal magnon (and vice versa), producing a third magnon at slightly higher energy. Two magnons are destroyed in the process and one magnon is created. All three magnons must have  $k \neq 0$ , so that  $S$  is unchanged while  $\Delta S_z = +1$  in the process. The rate of this process appears to control the intrinsic relaxation of  $S_z$  in the low-temperature range, provided that the coupling between the uniform and the  $S$  modes is not a bottleneck.

Recently nonresonant double-frequency pumping experiments have been carried out by LeCraw and Spencer<sup>24</sup> in which the relaxation rate of spin waves of given  $\mathbf{k}$  are studied directly, using a method of nonlinear rf excitation proposed by Schlömann.<sup>11</sup> These studies give highly detailed evidence for the three-magnon relaxation process and have the advantage that they do not involve special assumptions about the excitation of the  $S$  magnons by surface scattering.

We first calculate the relaxation frequency  $1/T_{1k}$  of an individual  $S$  mode of wave vector  $\mathbf{k}_1$  by magnetic dipolar Raman processes, which change  $M_z V$  by  $\pm 2\mu_B$ . The role of magnetic dipole coupling in the related problem of relaxation, averaged over a thermal distribution, has been treated by Akhiezer.<sup>25</sup> A qualitative calculation similar to ours was made by Kasuya<sup>2</sup> in 1954, before the line-broadening process was understood; so our present calculation amounts to a numerical refinement, on a firmer physical basis, of his result.

The magnetic dipole part of the Hamiltonian for the process is determined by

$$\mathcal{H} = -\frac{1}{2} \int d\mathbf{r} \mathbf{M} \cdot \mathbf{H}_s, \quad (16)$$

where  $\mathbf{M}$  is the magnetization and  $\mathbf{H}_s$ , the volume demagnetization field of the system of spin waves, is determined by

$$\nabla \cdot (\mathbf{H}_s + 4\pi \mathbf{M}) = 0; \quad \nabla \times \mathbf{H}_s = 0. \quad (17)$$

<sup>20</sup> P. Fletcher and C. Kittel, Phys. Rev. **120**, 2004 (1960).

<sup>21</sup> E. Schlömann, Phys. Rev. **121**, 1312 (1961).

<sup>22</sup> F. Keffer and R. Loudon (to be published).

<sup>23</sup> C. Kittel (to be published).

<sup>24</sup> R. C. LeCraw and E. C. Spencer (to be published).

<sup>25</sup> A. I. Akhiezer, J. Phys. U.S.S.R. **10**, 217 (1946).

Setting  $\mathbf{H}_s = -\nabla\psi$ , the solution is

$$\psi(\mathbf{r}) = - \int d\mathbf{r}' \frac{\nabla \cdot \mathbf{M}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}. \quad (18)$$

In terms of spin-wave creation and destruction operators  $a_\nu^\dagger$  and  $a_\nu$  for spin wave  $\mathbf{k}_\nu$ , the magnetization components are given by, taking  $\mu = g|\mu_B|/2$ ,

$$M^+(\mathbf{r}) = (4\mu M_s/V)^{\frac{1}{2}} \left[ \sum_\nu e^{i\mathbf{k}_\nu \cdot \mathbf{r}} a_\nu - (\mu/2VM_s) \right. \\ \left. \times \sum_{\lambda\mu\nu} e^{i(-\mathbf{k}_\nu + \mathbf{k}_\mu + \mathbf{k}_\lambda) \cdot \mathbf{r}} a_\nu^\dagger a_\mu a_\lambda \right] + \dots; \quad (19)$$

$$M^z(\mathbf{r}) = M_s - (2\mu/V) \sum_{\mu\nu} e^{i(-\mathbf{k}_\nu + \mathbf{k}_\mu) \cdot \mathbf{r}} a_\nu^\dagger a_\mu + \dots$$

We now write

$$\psi = \psi^{(1)} + \psi^{(2)} + \psi^{(3)} + \dots,$$

with

$$\psi^{(i)} = - \int d\mathbf{r}' \frac{\nabla \cdot \mathbf{M}^{(i)}}{|\mathbf{r} - \mathbf{r}'|};$$

here  $\mathbf{M}^{(i)}$  is the part of  $\mathbf{M}$  in (19) containing  $i$  spin-wave operators. In a short straightforward calculation using

$$\int d\mathbf{r}' \frac{e^{i\mathbf{k} \cdot \mathbf{r}'}}{|\mathbf{r} - \mathbf{r}'|} = \frac{4\pi}{k^2} e^{i\mathbf{k} \cdot \mathbf{r}},$$

we find

$$\mathbf{H}_s^{(1)} = -4\pi(\mu M_s/V)^{\frac{1}{2}} \sum_\lambda \mathbf{k}_\lambda k_\lambda^{-1} (k_\lambda)^{-2} e^{i\mathbf{k}_\lambda \cdot \mathbf{r}} a_\lambda + \text{c.c.};$$

$$\mathbf{H}_s^{(2)} = (8\pi\mu/V) \sum_{\lambda\mu} (-\mathbf{k}_\lambda + \mathbf{k}_\mu) (-k_\lambda^2 + k_\mu^2) \\ \times |-\mathbf{k}_\lambda + \mathbf{k}_\mu|^{-2} e^{i(-\mathbf{k}_\lambda + \mathbf{k}_\mu) \cdot \mathbf{r}} a_\lambda^\dagger a_\mu;$$

$$\mathbf{H}_s^{(3)} = 2\pi M_s (\mu/V M_s)^{\frac{1}{2}} \sum_{\lambda\mu\nu} (-\mathbf{k}_\lambda - \mathbf{k}_\mu + \mathbf{k}_\nu) \\ \times (-k_\lambda^2 - k_\mu^2 + k_\nu^2) |-\mathbf{k}_\lambda - \mathbf{k}_\mu + \mathbf{k}_\nu|^{-2} \\ \times e^{i(-\mathbf{k}_\lambda - \mathbf{k}_\mu + \mathbf{k}_\nu) \cdot \mathbf{r}} a_\lambda^\dagger a_\mu^\dagger a_\nu + \text{c.c.}; \quad (20)$$

where c.c. denotes Hermitean conjugate.

The three-magnon terms of the dipolar Hamiltonian are

$$\mathcal{H}^{(3)} = -\frac{1}{2} \sum_{i=0}^2 \int d\mathbf{r} \mathbf{M}^{(i)} \cdot \mathbf{H}_s^{(3-i)}, \quad (21)$$

whence

$$\mathcal{H}^{(3)} = \sum_{lmn} C_{lmn} a_l a_m^\dagger a_n + \text{c.c.}; \quad (22)$$

$$C_{lmn} = -8\pi\mu(\mu M_s/V)^{\frac{1}{2}} k_l^2 k_l^{-1} (k_l)^{-2} \Delta(\mathbf{k}_l - \mathbf{k}_m + \mathbf{k}_n),$$

and where  $\Delta(\mathbf{k})=1$  for  $\mathbf{k}=0$  and zero otherwise. In this calculation, direct evaluation of the integral  $\int d\mathbf{r} M_z^{(0)} H_{sz}^{(3)}$  gives the indeterminate value 0/0. This integral may be shown to vanish by writing  $H_{sz}^{(3)} = -\partial\psi^{(3)}/\partial z$  and integrating by parts. The result (22) is also obtained from the magnetic dipole term of

the Holstein-Primakoff Hamiltonian, as shown by Akhiezer.<sup>25</sup>

The nonvanishing matrix elements of  $\mathcal{H}^{(3)}$  in the exchange magnon representation are

$$\langle n_\lambda + 1, n_\mu - 1, n_\nu + 1 | \mathcal{H}^{(3)} | n_\lambda, n_\mu, n_\nu \rangle \\ = [(n_\lambda + 1)n_\mu(n_\nu + 1)]^{\frac{1}{2}} (C_{\lambda\mu\nu} + C_{\nu\mu\lambda}), \quad (23)$$

and

$$\langle n_\lambda - 1, n_\mu + 1, n_\nu - 1 | \mathcal{H}^{(3)} | n_\lambda, n_\mu, n_\nu \rangle \\ = [n_\lambda(n_\mu + 1)n_\nu]^{\frac{1}{2}} (C_{\lambda\mu\nu} + C_{\nu\mu\lambda}). \quad (24)$$

This representation does not diagonalize the lower order dipole terms, but the approximation should be adequate when  $H > \sim 2\pi M_s$ . By standard perturbation theory, the probability per unit time

$$W(n_\lambda, n_\mu, n_\nu; n_\lambda - 1, n_\mu + 1, n_\nu - 1)$$

of a transition from state  $|n_\lambda n_\mu n_\nu\rangle$  to state  $|n_\lambda - 1, n_\mu + 1, n_\nu - 1\rangle$  is

$$W(n_\lambda, n_\mu, n_\nu; n_\lambda - 1, n_\mu + 1, n_\nu - 1) \\ = (2\pi/\hbar) |\langle n_\lambda - 1, n_\mu + 1, n_\nu - 1 | \mathcal{H}^{(3)} | n_\lambda, n_\mu, n_\nu \rangle|^2 \\ \times \delta(\hbar\omega_\lambda - \hbar\omega_\mu + \hbar\omega_\nu), \quad (25)$$

where  $\hbar\omega_\lambda$  is the energy of magnon  $\mathbf{k}_\lambda$ . By taking  $n_\lambda = n_1$ , where the  $\mathbf{k}_1$  magnon is the  $S$  magnon whose relaxation frequency  $1/T_{1k}$  we are calculating, and summing Eq. (25) over  $\mu$  and  $\nu$  magnons, we get<sup>26</sup> the probability per unit time  $W^-$  that  $n_1$  decreases by one:

$$W^- = \sum_{\mu\nu} W(n_1, n_\mu, n_\nu; n_1 - 1, n_\mu + 1, n_\nu - 1). \quad (26)$$

The probability per unit time  $W^+$  that  $n_1$  increases by one is obtained from (26) by interchanging all plus and minus signs. The net rate of change of  $n_1$  is

$$dn_1/dt = W^+ - W^- = (2\pi/\hbar) \sum_{\mu\nu} |C_{1\mu\nu} + C_{\nu\mu 1}|^2 \\ \times [(n_1 + 1)n_\mu(n_\nu + 1) - n_1(n_\mu + 1)n_\nu] \\ \times \delta(\hbar\omega_1 - \hbar\omega_\mu + \hbar\omega_\nu). \quad (27)$$

With an earlier expression for  $C_{1\mu\nu}$  and  $C_{\nu\mu 1}$ , we have

$$dn_1/dt = (128\pi^3\mu^3 M_s/\hbar V) \\ \times \sum_{\mu\nu} |k_1^2 k_1^{-1} (k_1)^{-2} + k_\nu^2 k_\nu^{-1} (k_\nu)^{-2}|^2 \\ \times \Delta(\mathbf{k}_1 - \mathbf{k}_\mu + \mathbf{k}_\nu) [(n_1 + 1)n_\mu(n_\nu + 1) \\ - n_1(n_\mu + 1)n_\nu] \delta(\hbar\omega_1 - \hbar\omega_\mu + \hbar\omega_\nu). \quad (28)$$

The evaluation of this expression is considered in detail in Appendix B. The detail is essential to an understanding of the region of validity of the results. The relative sizes of  $k_B T$ ,  $Dk_1^2$ , and  $\hbar\omega_0$  dictate the assumptions which must be made in evaluating (28).

<sup>26</sup> Thus we neglect the process in which the  $S$  spin wave splits into two spin waves; this process cannot conserve energy in the high-field limit.

For the regime in which the exchange part of the energy of the magnon of wave vector  $\mathbf{k}_1$  is much less than the total energy  $\hbar\omega_1$  of the magnon (Zeeman + magnetostatic + exchange), we have

$$\frac{1}{T_{1k}} = \frac{16\pi\mu^3 M_s k_1 k_B T}{3D\hbar^2\omega_0} [1 + (17/2) \sin^2\theta_1 - (35/4) \sin^4\theta_1], \quad (29)$$

where  $\theta_1$  is the angle between  $\mathbf{k}_1$  and the  $z$  axis; the dispersion relation (8) connects  $\omega_1$ ,  $k_1$ , and  $\theta_1$ . This result is valid provided: (a)  $8Dk_1^2 \ll \hbar\omega_1$  and (b)  $k_B T \gg (\hbar\omega_1)^2/4Dk_1^2$ .

We note several features of (29). The minimum value of the angular function falls at  $\theta_1 = \pi/2$ ; near this angle the factor in the square brackets is  $[\frac{3}{4} + 9\alpha^2]$ , where  $\alpha = \theta_1 - \frac{1}{2}\pi$ . The fact that the minimum is at  $\pi/2$  enhances the likelihood that only  $\pi/2$  magnons will be produced at the threshold rf field in a parallel-pumping experiment, provided that the 3-magnon process dominates the magnon relaxation.

At constant  $\omega_1$  and  $\theta_1$  the relaxation frequency of the 3-magnon process is directly proportional to  $k_1$ . In parallel-pumping experiments we are chiefly concerned with the  $\pi/2$  magnons, and it is not difficult to arrange an experiment to vary  $k_1$  over a wide range while holding  $\omega_1$  constant, or nearly constant. We note that the process of relaxation by impurities which relax rapidly is roughly independent of  $\mathbf{k}$  at fixed  $\omega_1$ , as is also the 4-magnon dipolar coupling process. The 3-magnon dipolar process is rigorously zero in a sphere when one of the magnons has  $\mathbf{k} = 0$ ; this is not true of the 4-magnon dipolar processes.

A convenient form of (29) for numerical work is, with  $M_s = 195$  and  $\mu = 0.93 \times 10^{-20}$  erg/oe:

$$\frac{1}{T_{1k}} = 3.27 \times 10^4 \left\{ \frac{T(k_1 \times 10^{-5})}{(\omega_1 \times 10^{-10})(D \times 10^{28})} \right\} \times [1 + (17/2) \sin^2\theta_1 - (35/4) \sin^4\theta_1], \quad (30)$$

where  $T_{1k}$  is in sec;  $T$  in deg K;  $k_1$  in  $\text{cm}^{-1}$ ;  $\omega_1$  in rad/sec; and  $D$  in erg  $\text{cm}^2$ . At the time of preparation of this paper it appears likely that for YIG at 0°K the value of  $D$  is close to  $0.9 \times 10^{-20}$  erg  $\text{cm}^2$ , but a value near  $0.5 \times 10^{-28}$  has not yet been excluded.

The Schlömann correction<sup>21</sup> amounts to multiplication of (30) by a factor which varies between unity for  $\theta_1 = 0$  and

$$H_0 / \left[ \left( H_0 - \frac{4\pi}{3} M_s \right) \left( H_0 + \frac{8\pi}{3} M_s \right) \right]^{\frac{1}{2}}$$

for  $\theta_1 = \pi/2$ ; at the latter angle the factor in YIG at 0°K is 0.95 for  $H \sim 3000$  oe and 0.94 for  $H_0 \sim 8000$  oe.

It has been established experimentally<sup>27</sup> that  $D$  is essentially independent of temperature in YIG from

<sup>27</sup> R. C. LeCraw (private communication); see also reference 22.

4°K up to room temperature. It appears reasonable that one should use for  $M_s$  the value at the actual temperature rather than at 0°K.

The results we have just given are intended to be applicable within the approximations stated to that part of the relaxation of individual magnons which is linear in  $T$  and in  $k_1$ . The calculations may therefore be compared directly with measurements of  $1/T_{1k}$  in parallel-pumping experiments.

In uniform-mode resonance experiments the  $z$  component of the magnetization is relaxed at low temperatures only through the  $S$  modes. The uniform mode is scattered into the  $S$  modes with conservation of  $M_z$ , and then the 3-magnon process allows  $M_z$  to relax. A broad spectrum of  $S$  modes necessarily results from the scattering process, and to calculate the relaxation time for  $M_z$  we must take the appropriate average over (29).

We have some reason for believing that under ordinary conditions with  $\gamma(\Delta H)_0 \gg T_{1k}^{-1}$  it is not a bad approximation to assume that all the degenerate modes are populated equally; this is in contrast to assuming that the output spectrum of  $V_c(0, \mathbf{k})$  in Eq. (4) for a particular pit size  $R$  should be used as the input spectrum of the 3-magnon process. First, the Fourier spectrum of the surface roughness will cover a substantial range in  $k$ , because sharp cracks and boundaries between cleavage planes will contribute higher Fourier components than those contained in the analysis of the basic spherical pits related to the size of the grinding particle. Further, there is scattering among the various  $S$  modes. We have calculated the scattering potential between two modes  $\mathbf{k}_1, \mathbf{k}_2$  interacting by a spherical cavity. The result contains too many terms to be worth quoting here, but the dominant term in the scattering potential connecting two degenerate states  $\mathbf{k}_1$  and  $\mathbf{k}_2$ , neither being zero, is

$$V_c(\mathbf{k}_1, \mathbf{k}_2) \cong (8/3)\pi^2 R^3 (\mu M_s / V) \times [12 \cos^2\theta_K - 8 + 3 \cos^2\theta_{\mathbf{k}_1} + 3 \cos^2\theta_{\mathbf{k}_2}] \times [j_1(|K|R)/|K|R][a_{\mathbf{k}_1} a_{\mathbf{k}_2}^\dagger + a_{\mathbf{k}_1}^\dagger a_{\mathbf{k}_2}], \quad (31)$$

where  $\mathbf{K} = \mathbf{k}_1 - \mathbf{k}_2$ . This is the dominant term when  $k_1 R, k_2 R \gg 1$ . Now the relaxation rate from  $\mathbf{k}_1$  to  $\mathbf{k}_2$ , will be faster than from 0 to  $\mathbf{k}$ , for small  $K$ , because of density-of-states factors. On integrating the transition probability over the degenerate spectrum we find, for pit scattering,

$$1/T_{2\mathbf{k}_1} \approx 2k_1 R (1/T_{20}), \quad (32)$$

roughly, where  $T_{20}$  is given by (10). As  $k_1 R \gg 1$ , the diffusion of the  $S$  waves among the degenerate states proceeds quite rapidly.

Making then the assumption of uniform mixing, we need to calculate

$$1/T_1 = \int_0^{k_m} (1/T_{1k}) \rho(k) dk / \int_0^{k_m} \rho(k) dk, \quad (33)$$

where  $\rho(k)dk$  is the number of states per unit energy having  $|k|$  in  $dk$  at  $|k|$ , where the energy is taken at  $\hbar\omega_0$ . Here  $T_{1k}$  is given by (B.28).

In Appendix B we obtain (B.28) by evaluating the summations in (27) without making assumption (a), which is not well satisfied at the upper limit  $k_1 = k_m$  of the integral in (33). On substituting (B.28) into (33) and making the transformation  $\sigma = (k_1/k_m)^2$ , we find

$$\frac{1}{T_1} = \frac{2(3\pi M_s)^{1/2} \mu^3 k_B T}{\gamma^4 (\hbar D)^{3/2}} \times \frac{\int_0^1 d\sigma \frac{s_0 + s_2 \sin^2 \theta_1 + s_4 \sin^4 \theta_1}{[(3\omega_i/\omega_s) + \sigma] \cos \theta_1}}{\int_0^1 d\sigma \frac{\sigma^{1/2}}{[(3\omega_i/\omega_s) + \sigma]}}, \quad (34)$$

where  $\omega_i = \gamma[H_0 - (4\pi M_s/3)]$ ;  $\omega_s = 4\pi\gamma M_s$ ; and  $s_0, s_2$ , and  $s_4$  are functions of  $\sigma$  which are defined under (B.28). The integrands in (34) are functions of applied field  $H_0 = \omega_0/\gamma$ , but are independent of  $D$ . The calculation was carried out numerically for the frequency 9.34 kMc/sec used by Spencer and LeCraw.<sup>12</sup> We find, taking  $D = 0.9 \times 10^{-28}$  erg cm<sup>2</sup>,

$$1/T_1 = 0.24 \times 10^5 T \text{ sec}^{-1}, \quad (35)$$

where  $T$  is in degrees Kelvin. This should be lowered by not more than 5% by the Schlömann correction, and it would be increased by a factor of 2.1 by using the earlier value  $D = 0.55 \times 10^{-28}$  erg cm<sup>2</sup> for the exchange constant. In any event (35) is in satisfactory agreement with (15) within the accuracy of the experiment.

Also, it now appears that the original experimental result (15) may be too large<sup>28</sup> because the correction for what may be impurity relaxation may have been underestimated.

#### RELAXATION OF THE $\pi/2$ MAGNONS BY THREE-MAGNON DIPOLAR PROCESSES

We now consider the relaxation of the  $\pi/2$  magnons as a function of their wave-vector amplitude  $k_1$ . The result (29) is valid for  $\pi/2$  magnons with small exchange energy [assumption (a) under (29)] in the high-temperature limit [assumption (b) under (29)] provided the high-field approximation  $H_0 > \sim 2\pi M_s$  is satisfied. In the parallel-pumping experiments it is easy to measure the relaxation frequency of the  $\pi/2$  magnons when these have such a large exchange energy that assumption (a) is not valid. Now, (B.28) does not apply to magnons with large exchange energy because the assumption  $\omega_i \cong \omega_0$  is not valid for such exchange magnons. For high-energy magnons, the "splitting" process, in which the input magnons split into two magnons, must also be considered.

<sup>28</sup> R. C. LeCraw (private communication).

We consider only the high-field limit  $H_0 > \sim 2\pi M_s$ , i.e.,  $\omega_0 > \sim 2\omega_s/3$ , where we define  $\omega_s$  by the relation

$$\omega_s = 4\pi\gamma M_s. \quad (36)$$

The reader is reminded that  $\omega_0 > \omega_s/3$  is the condition required for saturation of the magnetization in spheres; hence, the present calculations are inapplicable only over the small range of applied magnetic field  $\omega_s/3 < \omega_0 < \sim 2\omega_s/3$  (i.e., 0.8 koe  $< H_0 < 1.6$  koe in YIG).

Schlömann<sup>21</sup> has calculated the relaxation frequency of the  $\pi/2$  magnons for the small-magnetic-field case in the limit of vanishing wave vector of the  $\pi/2$  magnon. The splitting process relaxes the uniform mode when  $\omega_0 < 2\omega_s/3$  (even at absolute zero temperature).

#### Relaxation of the $\pi/2$ Magnons by the Confluence Process

For the "confluence" process, in which two magnons are destroyed and one is created, it would be easy to reevaluate the integral in (B.13) without making the assumptions  $8Dk_1^2 \ll \hbar\omega_1$  and  $\omega_1 \cong \omega_0$ ; however, for both the confluence and splitting processes, we simplify the calculations of  $1/T_{1k}$  by neglecting the angular dependence of the interaction. The result of the calculation in Appendix C is, for the confluence process,

$$\left(\frac{1}{T_{1k}}\right)_{\text{confl}} = \frac{\pi \mu^2 M_s k_B T}{\hbar D^2 k_1} \times \ln \left[ 1 + \left(\frac{\omega_1}{\omega_0}\right) \frac{1}{1 + (\hbar\omega_0/4Dk_1^2)} \right]. \quad (37)$$

This result, which is valid in the high-field

$$(\omega_0 > \sim 2\omega_s/3),$$

high-temperature

$$\{k_B T \gg \hbar\omega_0 [1 + (\hbar\omega_0/4Dk_1^2)]\}$$

limit, is represented by the solid curve in Fig. 1. We

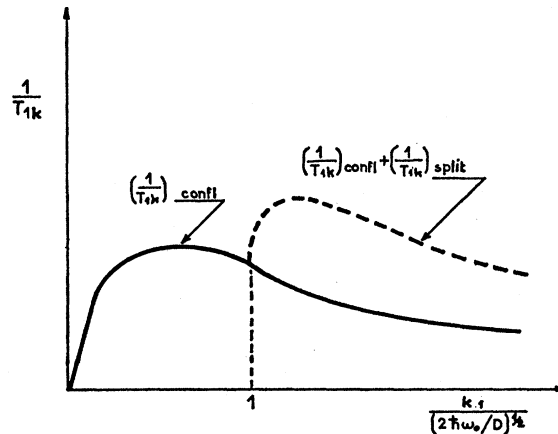


FIG. 1. Variation of relaxation frequency with wave-vector amplitude for the confluence and splitting processes. The magnitude of  $(1/T_{1k})_{\text{confl}}$  at the maximum is  $(2 \times 10^6) (T/300) \times [M_s(T)/M_s(300)] (9.34 \text{ kMc/sec}/\nu_0) \text{ sec}^{-1}$ .

now show that  $(1/T_{1k})_{\text{confl}}$  is linear in  $k_1$  in the Zeeman limit and varies as  $k_1^{-1} \ln k_1$  in the exchange limit.

### Confluence Process in the Zeeman Limit

In the Zeeman limit  $4Dk_1^2 \ll \hbar\omega_0$ , expansion of the logarithm in (37) gives

$$\left(\frac{1}{T_{1k}}\right)_{\text{confl}} = \frac{4\pi\mu^3 M_s k_1 k_B T}{D\hbar\omega_0}. \quad (38)$$

### Confluence Process in the Exchange Limit

In exchange limit  $4Dk_1^2 \gg \hbar\omega_0$ , (37) reduces to

$$\left(\frac{1}{T_{1k}}\right)_{\text{confl}} = \frac{\pi\mu^3 M_s k_B T}{\hbar D^2 k_1} \ln\left(\frac{\omega_1}{\omega_0}\right). \quad (39)$$

### Relaxation of the $\pi/2$ Magnons by the Splitting Process

The essential features of the rather complicated result (C.7) for the splitting relaxation frequency follow: In the high-field approximation  $\omega_0 > \sim 2\omega_s/3$ , the splitting process relaxes only those  $\pi/2$  magnons with  $k_1 \geq (2\hbar\omega_0/D)^{1/2}$ ; the splitting process cannot conserve energy and momentum for magnons with wave vectors smaller than this threshold value.

### Splitting Process for $k_1$ near the Threshold Value

For  $k_1$  near the threshold value  $k_{td} = (2\hbar\omega_0/D)^{1/2}$  the relaxation frequency of the  $\pi/2$  magnons is

$$\left(\frac{1}{T_{1k}}\right)_{\text{split}} = \frac{4\pi\mu^3 M_s k_B T}{3\hbar D^3 (\hbar\omega_0)^{1/2}} \left(\frac{k_1 - k_{td}}{k_{td}}\right)^{1/2}. \quad (40)$$

### Splitting Process in the Extreme Exchange Limit

In the extreme exchange limit  $Dk_1^2 \gg 2\hbar\omega_0$ , the relaxation frequency of the  $\pi/2$  magnon is

$$\left(\frac{1}{T_{1k}}\right)_{\text{split}} = \frac{\pi\mu^3 M_s k_B T}{\hbar D^2 k_1} \ln\left(\frac{\omega_1}{\omega_0}\right). \quad (41)$$

In the extreme exchange limit, the splitting and confluence processes contribute equally to the relaxation frequency. The results (40) and (41) are valid in the high-field ( $\omega_0 > \sim 2\omega_s/3$ ), high-temperature  $k_B T \gg \hbar\omega_1$  limit. In Fig. 1 we present these results as the dotted curve  $(1/T_{1k})_{\text{confl}} + (1/T_{1k})_{\text{split}}$ .

### Relaxation of Exchange Magnons by Other Processes

In addition to the three-magnon processes, other processes are important at high temperature in relaxing magnons with large exchange energy. The four-magnon processes will be considered in a subsequent publication.

In order to observe the sharp increase in  $1/T_{1k}$  from the splitting process (Fig. 1), the temperature must be lowered below room temperature to reduce the contribution to  $1/T_{1k}$  from the higher order processes. Of these higher order processes, the most effective in relaxing the exchange magnons at high temperature will probably be the four-magnon exchange process. Using Dyson's result<sup>29</sup> for the relaxation frequency of the four-magnon exchange process, we estimate that for  $\omega_0 = 2\omega_s/3$ , i.e.,  $H_0 = 1.6$  koe in YIG, the effect of the splitting process should be observable at 120°K. Lower applied magnetic fields should be avoided in this experiment, for corrections associated with the noncircular precession of the spins must then be applied. In the above estimation of the temperature at which the splitting process is observable, the rather low value of field  $\omega_0 = 2\omega_s/3$  was chosen to put the microwave frequency within the present experimentally available range. The sharp rise in  $1/T_{1k}$  as a function of  $k_1$  at the onset of the splitting process (Fig. 1) occurs at  $\sim 30$  kMc/sec in YIG for this value of field. (The magnon frequency is one-half of this microwave frequency.) *Note added in proof.* LeCraw has recently observed<sup>30</sup> the initial departure of the relaxation frequency of the  $\pi/2$  magnons from linearity in  $k_1$ , as shown in Fig. 1. The onset of this concave downward region was observed at  $k_1 \cong 1.6 \times 10^5 \text{ cm}^{-1}$  for magnons with a frequency of 5.7 kMc/sec. From (37), with a slight correction for the four-magnon process, we estimate that the onset of the bendover should occur at  $k_1 \cong 1.5 \times 10^5 \text{ cm}^{-1}$ , in agreement with the observed value within the accuracy of the estimation. The inflection point between the concave downward region (three-magnon confluence) and the concave upward region (four-magnon exchange) should occur at  $k_1$  near  $3 \times 10^5 \text{ cm}^{-1}$  for  $T = 300^\circ\text{K}$  and  $= 5.7$  kMc/sec.

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### APPENDIX A. SCATTERING OF UNIFORM MODE SPIN WAVES; DERIVATION OF EQUATION (4)

The components of the magnetization  $\mathbf{M}$  are given in (2) and (3) of the text. In calculating the interaction which relaxes the uniform mode, the important terms

<sup>29</sup> Freeman J. Dyson, Phys. Rev. **102**, 1217 (1956).

<sup>30</sup> T. Kasuya and R. C. LeCraw, Phys. Rev. Letters **6**, 223 (1961).



are those in  $a_0^\dagger a_k$  and  $a_0 a_k^\dagger$ , and it is convenient to split up the magnetization into parts  $M_0$  and  $M_k$  defined by:

$$M_0^+ = (4\mu M_s/V)^{1/2} a_0, \quad M_0^- = (4\mu M_s/V)^{1/2} a_0^\dagger; \\ M_0^z = M_s; \quad (\text{A.1})$$

$$M_k^+ = (4\mu M_s/V)^{1/2} \exp(i\mathbf{k} \cdot \mathbf{r}) a_k; \\ M_k^- = (4\mu M_s/V)^{1/2} \exp(-i\mathbf{k} \cdot \mathbf{r}) a_k^\dagger, \quad (\text{A.2}) \\ M_k^z = -(2\mu/V) [\exp(-i\mathbf{k} \cdot \mathbf{r}) a_k^\dagger a_0 + \exp(i\mathbf{k} \cdot \mathbf{r}) a_0^\dagger a_k].$$

The required interaction is then contained in

$$V_c = -\frac{1}{2} \sum_{\mathbf{k} \neq 0} \int [\mathbf{M}_0 \cdot \mathbf{H}_k + \mathbf{M}_k \cdot \mathbf{H}_0] d\mathbf{r}, \quad (\text{A.3})$$

the terms in  $-\mathbf{M}_0 \cdot \mathbf{H}_0$  and  $\mathbf{M}_k \cdot \mathbf{H}_k$  making no contribution to the scattering.

In the above expression  $\mathbf{H}_0$  and  $\mathbf{H}_k$  represent the magnetostatic fields due to the magnetizations  $\mathbf{M}_0$  and  $\mathbf{M}_k$ , respectively. There are two types of magnetostatic field to consider: the magnetostatic field due to the dipole moment induced at the cavity and the magnetostatic field in the bulk material of the sample. This latter makes a contribution to the cavity spin-wave scattering, since the integrated magnetostatic energy of the sample is changed by the introduction of the cavity.

Consider for simplicity the case where the spherical cavity of radius  $R$  is at the center of a spherical sample of radius  $r_0$ . We treat first the magnetostatic field of the cavity alone. The magnetic potential  $\phi_c$  in the sample, caused by the introduction of the cavity, can be evaluated by ordinary potential theory. We suppose that  $\phi_i$  is the magnetic potential in the interior of the cavity and we let  $\phi_M$  be the potential of the magnetization, such that  $\mathbf{M} = -\nabla\phi_M$ . Then the boundary conditions at the surface of the cavity are

$$\phi_i = \phi_c; \quad \frac{\partial\phi_i}{\partial r} = \frac{\partial\phi_c}{\partial r} + 4\pi \frac{\partial\phi_M}{\partial r}. \quad (\text{A.4})$$

We have

$$\partial\phi_M/\partial r = (1/2) \sin\theta [M^- \exp(i\phi) - M^+ \exp(-i\phi)] - M^z \cos\theta. \quad (\text{A.5})$$

The spatial dependence of the components of  $\mathbf{M}$  is given by terms in

$$\exp(i\mathbf{k} \cdot \mathbf{r}) = \sum_{n=0}^{\infty} (2n+1) i^n j_n(kr) P_n^0(\cos\theta). \quad (\text{A.6})$$

It follows that  $\partial\phi_M/\partial r$  can be expressed in the following way:

$$\partial\phi_M/\partial r = \sum_{n=0}^{\infty} \sum_{m=-1,0,1} A_{nm}(r) P_n^m(\cos\theta) e^{im\phi}. \quad (\text{A.7})$$

Now  $\phi_i$  and  $\phi_c$  can be expanded in a series of spherical

harmonics as follows:

$$\phi_i = \sum_{n=0}^{\infty} \sum_{m=-1,0,1} B_{nm} r^n P_n^m(\cos\theta) e^{im\phi}; \quad (\text{A.8})$$

$$\phi_c = \sum_{n=0}^{\infty} \sum_{m=-1,0,1} C_{nm} r^{-n-1} P_n^m(\cos\theta) e^{im\phi}; \quad (\text{A.9})$$

because  $\phi_i$  and  $\phi_c$  can contain only those spherical harmonics contained in  $\partial\phi_M/\partial r$ . The components of  $H$  are expressions of the form

$$H^i = \sum_{n=1}^{\infty} \sum_{m=0,\pm 1,\pm 2} D_{nm} r^{-n-2} P_n^m(\cos\theta) e^{im\phi}. \quad (\text{A.10})$$

It is important to notice that  $H^i$  contains no term in  $P_0(\cos\theta)$ , i.e., no spherically symmetric term. Because of this, only those terms in  $M$  which vary spatially can make a contribution to  $V_c$ , i.e., we need consider only the second term in the integrand of (A.3), as far as the cavity is concerned. The calculation of  $\mathbf{H}_0$  is simple, since  $\mathbf{M}_0$  is constant; and  $\phi_c$  therefore contains only a dipole part:

$$\phi_c = (4\pi R^3/3) (4\mu M_s/V)^{1/2} (1/2) [a_0^\dagger \exp(i\phi) - a_0 \exp(-i\phi)] r^{-2} \sin\theta - (4\pi R^3/3) M_s r^{-2} \cos\theta. \quad (\text{A.11})$$

Hence, the contribution to the scattering interaction is

$$-\pi R^3 (2\mu M_s/V) \sum_{\mathbf{k}} \int (3z^2 - r^2)/r^5 [a_0 a_k^\dagger \exp(-i\mathbf{k} \cdot \mathbf{r}) + a_0^\dagger a_k \exp(i\mathbf{k} \cdot \mathbf{r})] d\mathbf{r}. \quad (\text{A.12})$$

The radial integral falls off as  $j_1(kr)/kr$  and the integral may be extended to a good approximation over all space, excluding the cavity volume, to give

$$8\pi^2 R^3 (\mu M_s/V) \sum_{\mathbf{k}} (3 \cos^2\theta_k - 1) [j_1(kR)/kR] \times (a_0 a_k^\dagger + a_0^\dagger a_k). \quad (\text{A.13})$$

Now consider the magnetostatic field which exists in the absence of the cavity. A uniform magnetization has no such demagnetizing field, so now it is the first term in the integrand of (A.3) which contributes to scattering interaction. Now  $\mathbf{H}_k$  is determined by

$$\nabla \cdot (\mathbf{H}_k + 4\pi \mathbf{M}_k) = 0, \quad \nabla \times \mathbf{H}_k = 0. \quad (\text{A.14})$$

From (20),

$$\mathbf{H}_k = \{ -2\pi (4\mu M_s/V)^{1/2} (1/k^2) [k^+ a_k^\dagger \exp(-i\mathbf{k} \cdot \mathbf{r}) + k^- a_k \exp(i\mathbf{k} \cdot \mathbf{r})] + 4\pi (2\mu/V) (k^z/k^2) \times [a_k^\dagger a_0 \exp(-i\mathbf{k} \cdot \mathbf{r}) + a_0^\dagger a_k \exp(i\mathbf{k} \cdot \mathbf{r})] \} \mathbf{k}. \quad (\text{A.15})$$

The contribution to the scattering interaction is,

therefore,

$$\begin{aligned}
 & -\pi(2\mu M_s/V)\sum_{\mathbf{k}}(3\cos^2\theta_{\mathbf{k}}-1)(a_0^\dagger a_{\mathbf{k}}+a_0 a_{\mathbf{k}}^\dagger) \\
 & \times \int \exp(i\mathbf{k}\cdot\mathbf{r})d\mathbf{r} \\
 & = -4\pi^2(2\mu M_s/V)\sum_{\mathbf{k}}(3\cos^2\theta_{\mathbf{k}}-1) \\
 & \times (a_0^\dagger a_{\mathbf{k}}+a_0 a_{\mathbf{k}}^\dagger)[r^3 j_1(kr)/kr]^{r_0}. \quad (\text{A.16})
 \end{aligned}$$

For sufficiently large  $r_0$ , the value at this limit is vanishingly small. The value at the lower limit is equal to (A.13). Both terms in (A.3), therefore, make equal contributions to the scattering interaction, which is:

$$\begin{aligned}
 V_c &= 16\pi^2 R^3 (\mu M_s/V) \sum_{\mathbf{k}} (3\cos^2\theta_{\mathbf{k}}-1) \\
 & \times [j_1(kR)/kR] (a_0 a_{\mathbf{k}}^\dagger + a_0^\dagger a_{\mathbf{k}}). \quad (\text{A.17})
 \end{aligned}$$

#### APPENDIX B. EVALUATION OF THE TRANSITION RATE FOR S MAGNONS BY THE THREE-MAGNON PROCESS

We first consider the factor

$$[(n_1+1)\bar{n}_\mu(\bar{n}_\nu+1)-n_1(\bar{n}_\mu+1)\bar{n}_\nu]$$

in Eq. (28), which we denote by  $[n]$ , where the bars indicate thermal equilibrium values. We subtract from  $[n]$  the same expression but with  $n_1$  replaced by  $\bar{n}_1$ ; the quantity which we subtract is identically zero. The result is

$$[n] = (n_1 - \bar{n}_1)(\bar{n}_\mu - \bar{n}_\nu). \quad (\text{B.1})$$

We substitute the Bose factors for  $\bar{n}_\mu$  and  $\bar{n}_\nu$  and use  $\omega_\mu = \omega_1 + \omega_\nu$  to obtain

$$\begin{aligned}
 [n] &= -(n_1 - \bar{n}_1)[\exp(\hbar\omega_1/\tau) - 1] \\
 & \times \exp(\hbar\omega_\nu/\tau)/[\exp(\hbar\omega_\nu/\tau) - 1] \\
 & \times \{\exp[\hbar(\omega_\nu + \omega_1)/\tau] - 1\}. \quad (\text{B.2})
 \end{aligned}$$

We may eliminate  $\mu$  as an independent variable in (28) by using momentum conservation, so that  $\omega(\mathbf{k}_\mu) = \omega(\mathbf{k}_1 + \mathbf{k}_\nu) \equiv \omega_{1+\nu}$ . We then replace  $\sum_\nu$  by  $[V/(2\pi)^3] \times \int d\mathbf{k}_\nu$ ; then, with the help of (B.2), (28) may be written

$$dn_1/dt = -(1/T_{1k})(n_1 - \bar{n}_1), \quad (\text{B.3})$$

where

$$\begin{aligned}
 1/T_{1k} &= (16\mu^3 M_s/\hbar)[\exp(\hbar\omega_1/\tau) - 1] \\
 & \times \int_0^\infty dk_\nu k_\nu^2 \int_0^{2\pi} d\phi_\nu \int_{-1}^1 d(\cos\theta_\nu) \\
 & \times |k_1^z k_1^-(k_1)^{-2} + k_\nu^z k_\nu^-(k_\nu)^{-2}|^2 \\
 & \times \delta(\hbar\omega_1 + \hbar\omega_\nu - \hbar\omega_{1+\nu}) \\
 & \times \exp(\hbar\omega_\nu/\tau)/[\exp(\hbar\omega_\nu/\tau) - 1] \\
 & \times \{\exp[\hbar(\omega_\nu + \omega_1)/\tau] - 1\}. \quad (\text{B.4})
 \end{aligned}$$

The energy delta function is simplified by applying

the high-frequency approximation  $\hbar\omega \cong Dk^2 + \hbar\omega_0$  to  $\hbar\omega_\nu$  and  $\hbar\omega_\mu$ , so that

$$\hbar\omega_\nu \cong Dk_\nu^2 + \hbar\omega_0; \quad \hbar\omega_\mu \cong D|\mathbf{k}_\nu + \mathbf{k}_1|^2 + \hbar\omega_0. \quad (\text{B.5})$$

With the help of (B.5) the delta function becomes, writing  $\mathbf{k}_\nu \cdot \mathbf{k}_1/k_\nu k_1 = \xi_{\nu 1}$ :

$$\delta(\hbar\omega_1 + \hbar\omega_\nu - \hbar\omega_\mu) \cong (1/2Dk_1 k_\nu) \delta(\xi_{\nu 1} - k_0 k_\nu^{-1}), \quad (\text{B.6})$$

where

$$k_0 = (\hbar\omega_1 - Dk_1^2)/2Dk_1. \quad (\text{B.7})$$

In evaluating the angle integrals in (B.4) we choose the polar axis along  $\mathbf{k}_1$ ; the integral over  $\theta_\nu$  may be written

$$\int_{-1}^1 d\xi_{\nu 1} \delta(\xi_{\nu 1} - k_0 k_\nu^{-1}) = \begin{cases} 0 & \text{for } k_\nu < k_0 \\ 1 & \text{for } k_\nu > k_0. \end{cases} \quad (\text{B.8})$$

With  $k_\nu dk_\nu = (\tau/2D)d(\hbar\omega_\nu/\tau)$ , (B.4) becomes

$$\begin{aligned}
 1/T_{1k} &= (4\mu^3 M_s/\hbar D^2 k_1) [\exp(\hbar\omega_1/\tau) - 1] \\
 & \times \int_{Dk_0^2 + \hbar\omega_0}^\infty d(\hbar\omega_\nu) \\
 & \times \int_0^{2\pi} d\phi_\nu |k_1^z k_1^-(k_1)^{-2} + k_\nu^z k_\nu^-(k_\nu)^{-2}|_{\xi_{\nu 1} = k_0 k_\nu^{-1}} \\
 & \times \exp(\hbar\omega_\nu/\tau)/[\exp(\hbar\omega_\nu/\tau) - 1] \\
 & \times \{\exp[\hbar(\omega_\nu + \omega_1)/\tau] - 1\}. \quad (\text{B.9})
 \end{aligned}$$

The angular term in (B.9) may be written, referred now to the  $z$  axis as the polar axis,

$$\begin{aligned}
 & |k_1^z k_1^-(k_1)^{-2} + k_\nu^z k_\nu^-(k_\nu)^{-2}|_{\xi_{\nu 1} = k_0 k_\nu^{-1}} \\
 & = [\cos^2\theta_1 \sin^2\theta_\nu + \cos^2\theta_\nu \sin^2\theta_1 + 2\cos\theta_1 \cos\theta_\nu \\
 & \quad \times (\cos\theta_{\nu 1} - \cos\theta_1 \cos\theta_\nu)]_{\cos\theta_{\nu 1} = k_0 k_\nu^{-1}}. \quad (\text{B.10})
 \end{aligned}$$

After some tedious trigonometry we find

$$\begin{aligned}
 & \int_0^{2\pi} d\phi_{\nu 1} |k_1^z k_1^-(k_1)^{-2} + k_\nu^z k_\nu^-(k_\nu)^{-2}|_{\xi_{\nu 1} = k_0 k_\nu^{-1}} \\
 & = \pi[L + M(k_0/k_\nu)^2 + N(k_0/k_\nu)^4], \quad (\text{B.11})
 \end{aligned}$$

where

$$\begin{aligned}
 L &= \sin^2\theta_1 - (3/4)\sin^4\theta_1, \\
 M &= 2 - 3\sin^2\theta_1 + (3/2)\sin^4\theta_1, \\
 N &= -2 + 10\sin^2\theta_1 - (35/4)\sin^4\theta_1. \quad (\text{B.12})
 \end{aligned}$$

On substituting (B.11) into (B.9) we find

$$\begin{aligned}
 \frac{1}{T_{1k}} &= \frac{4\pi\mu^3 M_s}{\hbar D^2 k_1} [\exp(\hbar\omega_1/\tau) - 1] \int_{Dk_0^2 + \hbar\omega_0}^\infty d(\hbar\omega_\nu) \\
 & \times \frac{\exp(\hbar\omega_\nu/\tau)}{[\exp(\hbar\omega_\nu/\tau) + 1]\{\exp[\hbar(\omega_\nu + \omega_1)/\tau] - 1\}} \\
 & \times \left[ L + M\left(\frac{k_0}{k_\nu}\right)^2 + N\left(\frac{k_0}{k_\nu}\right)^4 \right]. \quad (\text{B.13})
 \end{aligned}$$

The  $L$  term in (B.13) can be integrated exactly and  $M$  and  $N$  terms can be integrated approximately. Experiments involving  $1/T_{1k}$  have been performed at temperatures ranging from a few degrees Kelvin up to the Curie temperature, and in the parallel-pumping experiments it is possible to measure the relaxation frequency of magnons with any energy from the Zeeman region through the exchange region. Thus, the conditions of any particular experiment dictate the assumptions which must be made in evaluating the integral in (B.13); the results vary markedly for different experiments.

**Case I. Relaxation Rate of  $S$  Magnons for (a):**  
 $8Dk_1^2 \ll \hbar\omega_1$ , and (b):  $k_B T \gg (\hbar\omega_1)^2/4Dk_1^2$

We consider first case I, where the exchange energy of the input magnon is much smaller than its total energy and where the high-temperature approximation  $[\exp(\hbar\omega/\tau) - 1] \cong \hbar\omega/\tau$  is valid for each of the three magnons. We show later that under these conditions

$$Dk_v^2 \gg 2\hbar\omega_1, \quad (\text{B.14})$$

and

$$\hbar\omega_1 \cong \hbar\omega_0. \quad (\text{B.15})$$

Thus, in (B.13) we set

$$\exp(\hbar\omega_v/\tau)/[\exp(\hbar\omega_v/\tau) - 1] \\ \times \{\exp[\hbar(\omega_v + \omega_1)/\tau] - 1\} \cong \tau^2/(\hbar\omega_v)^2,$$

and  $Dk_v^2 \cong \hbar\omega_v$ ; and in the limit of the integral in (B.13) we set  $Dk_0^2 + \hbar\omega_0 \cong Dk_0^2$ . We examine the assumptions below. With these assumptions we find from (B.13)

$$\frac{1}{T_{1k}} = \frac{4\pi\mu^3 M_s \tau^2}{3\hbar D^2 k_1 k_0^2} [\exp(\hbar\omega_1/\tau) - 1] F(\theta_1), \quad (\text{B.16})$$

where

$$F(\theta_1) = 1 + (17/2) \sin^2 \theta_1 - (35/4) \sin^4 \theta_1, \quad (\text{B.17})$$

and

$$k_0^2 \cong (\hbar\omega_1/2Dk_1^2)^2.$$

This approximation for  $k_0^2$  is valid for

$$Dk_1^2 \ll \hbar\omega_1. \quad (\text{B.18})$$

Finally, for

$$k_B T \gg \hbar\omega_1 \quad (\text{B.19})$$

the exponential factor in (B.16) may be approximated by  $\hbar\omega_1/\tau$ . Using these approximations

$$1/T_{1k} = (16\pi\mu^3 M_s k_1 k_B T / 3D\hbar^2 \omega_0) F(\theta_1), \quad (\text{B.20})$$

with  $F(\theta_1)$  defined by (B.17). The angle  $\theta_1$  is determined by the dispersion relation (8).

The result (B.20) is valid provided (a):  $8Dk_1^2 \ll \hbar\omega_1$  and (b):  $k_B T \gg (\hbar\omega_1)^2/4Dk_1^2$ . We now show that assumptions (a) and (b) are equivalent to the earlier assumptions (B.14), (B.15), (B.18), and (B.19). For the

convenience of the reader we list the following equations:

$$2\hbar\omega_1 \ll Dk_0^2, \quad (\text{B.21})$$

$$\hbar\omega_v \cong Dk_v^2, \quad (\text{B.22})$$

$$Dk_0^2 \ll k_B T, \quad (\text{B.23})$$

$$k_0^2 \cong (\hbar\omega_1/2Dk_1^2)^2, \quad (\text{B.24})$$

and

$$k_B T \gg \hbar\omega_1. \quad (\text{B.25})$$

Equation (B.15) is satisfied exactly in the modulation experiments and is well satisfied for  $\pi/2$  magnons which have negligible exchange energy in the high-field limit  $\omega_0 > \sim 2\omega_s/3$ , where  $\omega_s$  is defined by (36). For example, from the dispersion relation (8) with  $Dk^2 = 0$  and  $\sin^2 \theta = 1$ , it is easy to show that  $\omega_0$  and  $\omega_1$  differ by no more than 6% for  $\omega_0 \geq 1.8(\omega_s/3)$ .

In (B.13) the minimum value of  $Dk_v^2$  is  $Dk_0^2$ ; with (B.21) this gives (B.14). With  $k_0$  given by (B.24), (B.21) is assumption (a) and (B.23) is assumption (b). The validity of (B.24) requires that  $Dk_1^2 \ll \hbar\omega_1$ , a less severe restriction than (a). In (B.13),  $Dk_v^2 \geq Dk_0^2$ ; hence, using (B.24)

$$Dk_v^2 \geq (\hbar\omega_1/4Dk_1^2)\hbar\omega_1.$$

With assumption (a) this gives  $Dk_v^2 \gg \hbar\omega_1$  and (B.22) is satisfied. Finally, (a) and (b) are consistent only if (B.23) is satisfied.

In the low-temperature resonance experiments<sup>12</sup> of Spencer and LeCraw, the maximum wave-vector magnitude  $k_m$  for  $S$  modes, as determined by the dispersion relation (8), is

$$k_m = (\hbar\omega_s/3D)^{1/2}, \quad (\text{B.26})$$

with  $\omega_s$  defined by (36). In YIG, where  $M_s = 195$  gauss and  $D = 9 \times 10^{-29}$  erg cm<sup>2</sup>,  $k_m = 4.1 \times 10^5$  cm<sup>-1</sup>. The resonance experiments reported are at 9.34 kMc/sec; hence,  $\hbar\omega_0 = 0.45 k_B$  erg. Assumption (b) is satisfied for all  $k \gg 1.2 \times 10^5$  cm<sup>-1</sup> if  $T \geq 5^\circ$  Kelvin. The maximum value of  $Dk_1^2$  for magnons degenerate with the uniform mode is  $Dk_m^2 = \hbar\omega_s/3 = 0.11 k_B$  erg, and (a) is not well satisfied for the largest  $k_1$ 's. We therefore evaluate the integrals in (B.14) for case II, in which we do not make assumption (a).

**Case II. Relaxation of  $S$  Magnons for (b):**

$$k_B T \gg (\hbar\omega_1)^2/4Dk_1^2 \text{ and (c): } k_B T \gg 2\hbar\omega_1$$

In (B.13) we set

$$\exp(\hbar\omega_v/\tau)/[\exp(\hbar\omega_v/\tau) - 1] \{ \exp[\hbar(\omega_v + \omega_1)/\tau] - 1 \} \\ \cong \tau^2/(Dk_v^2 + \hbar\omega_0)(Dk_v^2 + \hbar\omega_0 + \hbar\omega_1)$$

and define  $z = Dk_v^2/\hbar\omega_0$ . In the high-field limit,  $\omega_1 \cong \omega_0$ ; thus

$$\frac{1}{T_{1k}} = \frac{4\pi\mu^3 M_s \tau}{\hbar D^2 k_1} \int_{Dk_0^2/\hbar\omega_0}^{\infty} dz \frac{1}{(z+1)(z+2)} \\ \times \left[ L + M \left( \frac{Dk_0^2}{\hbar\omega_0 z} \right) + N \left( \frac{Dk_0^2}{\hbar\omega_0 z} \right)^2 \right]. \quad (\text{B.27})$$

On evaluating the integrals by partial fractions, we find

$$1/T_{1k} = (4\pi\mu^3 M_s \tau / \hbar D^2 k_1) (s_0 + s_2 \sin^2 \theta_1 + s_4 \sin^4 \theta_1), \quad (\text{B.28})$$

where

$$\begin{aligned} s_0 &= m - (1/2)n, \\ s_2 &= l - (3/2)m + (5/2)n, \\ s_4 &= -(3/4)l + (3/4)m - (35/16)n; \end{aligned}$$

and where

$$\begin{aligned} l &= \ln[(z_0 + 2)/(z_0 + 1)], \\ m &= z_0 \ln[(z_0 + 1)^2 / z_0(z_0 + 2)], \end{aligned}$$

$$n = z_0^2 \left[ \ln \left( \frac{z_0^3(z_0 + 2)}{(z_0 + 1)^4} \right) + \frac{2}{z_0} \right],$$

and

$$z_0 = \frac{3\omega_0}{4\omega_s} \left( \frac{k_1}{k_m} \right)^2 \left[ \left( \frac{k_m}{k_1} \right)^2 - \frac{\omega_s}{3\omega_0} \right]^2.$$

### Case III. Relaxation of $S$ Magnons for (a): $8Dk_1^2 \ll \hbar\omega_1$ and (c): $k_B T \ll (\hbar\omega_1)^2 / 4Dk_1^2$

We now evaluate the integral in (B.13) for the other limiting value of  $Dk_0^2/\tau$ , i.e.,  $Dk_0^2 \gg \tau$ . In this limit

$$\begin{aligned} \exp(\hbar\omega_\nu/\tau) / [\exp(\hbar\omega_\nu/\tau) - 1] \{ \exp[\hbar(\omega_\nu + \omega_1)/\tau] - 1 \} \\ \cong \exp[-\hbar(\omega_\nu + \omega_1)/\tau], \end{aligned}$$

and the integrand is a rapidly decreasing function of  $\hbar\omega_\nu$  at the lower limit of the integral. Thus defining  $x = Dk_\nu^2/\tau$ ,

$$\begin{aligned} \int_{Dk_0^2/\tau}^{\infty} dx \exp\{-[x + (2\hbar\omega_0/\tau)]\} (Dk_0^2/\tau x)^u \\ \cong \exp\{-[(Dk_0^2/\tau) + (2\hbar\omega_0/\tau)]\}, \end{aligned}$$

for  $u=0, 1$ , or  $2$ . This gives, for  $Dk_0^2 \gg 2\hbar\omega_0$ ,

$$\begin{aligned} 1/T_{1k} &= (32\pi\mu^3 M_s \tau / \hbar D^2 k_1) [\exp(\hbar\omega_0/\tau) - 1] \\ &\times \exp\{-[(\hbar\omega_0)^2 / 4Dk_1^2 \tau]\} \sin^2 \theta_1 \cos^2 \theta_1. \quad (\text{B.29}) \end{aligned}$$

This is a good approximation to  $1/T_{1k}$  provided assumption (a) is satisfied, and further (c):  $k_B T \ll (\hbar\omega_1)^2 / 4Dk_1^2$ .

In the Spencer and LeCraw experiments (c) is satisfied at  $T=5^\circ$  Kelvin for  $k_1 \ll 0.29k_m$ . The factor  $\exp\{-[(\hbar\omega_0)^2 / 4Dk_1^2 \tau]\}$ , when  $(\hbar\omega_1 / 4Dk_1^2 \tau) \gg 1$ , makes  $1/T_{1k}$  very small when assumption (c) is satisfied. We notice also that  $1/T_{1k} = 0$  for  $\theta_1 = 0$  or  $\pi/2$  in (B.29).

### APPENDIX C. EVALUATION OF THE TRANSITION RATE FOR $\pi/2$ MAGNONS FOR THE THREE-MAGNON PROCESSES

#### Relaxation Rate of $\pi/2$ Magnons for the Confluence Process

We shall first derive (37), the relaxation rate for the  $\pi/2$  magnons by the three-magnon confluence process. We neglect the angular dependence of the interaction;

thus, in (B.9) we set

$$|k_1^z k_1^- (k_1)^{-2} + k_\nu^z k_\nu^- (k_\nu)^{-2}|^2 = \frac{1}{8}.$$

The constant  $\frac{1}{8}$  is chosen to give agreement between the exact result (29) and the result of the present calculation in the limit of low  $k_1$ . In the high-temperature approximation  $k_B T \gg \hbar\omega_\nu + \hbar\omega_1$ , (B.9) becomes

$$\frac{1}{T_{1k}} = \frac{\pi\mu^3 M_s \tau}{\hbar D^2 k_1} \ln \left[ 1 + \frac{\hbar\omega_1}{Dk_0^2 + \hbar\omega_0} \right], \quad (\text{C.1})$$

where  $k_0$  is given by (B.7). We make the approximation  $\hbar\omega_1 - Dk_1^2 \cong \hbar\omega_0$ , which is well satisfied for all  $\pi/2$  magnons provided  $\omega_0 > \sim 2\omega_s/3$ . For example,  $\hbar\omega_1 - Dk_1^2$  and  $\hbar\omega_0$  differ by 6% for  $3\omega_0/\omega_s = 4$  and  $Dk_1^2 \ll \hbar\omega_0$ . Thus

$$Dk_0^2 \cong (\hbar\omega_0)^2 / 4Dk_1^2.$$

On substituting this value of  $Dk_0^2$  into (C.1), we obtain (37).

#### Relaxation Rate of $\pi/2$ Magnons for the Splitting Process

We now calculate  $1/T_{1k}$  for the splitting process. The nonvanishing matrix elements of  $\mathcal{H}^{(3)}$  [Eq. (22)] for the splitting process in the exchange magnon representation are

$$\begin{aligned} |\langle n_1 + 1, n_\lambda - 1, n_\nu - 1 | \mathcal{H}^{(3)} | n_1, n_\lambda, n_\nu \rangle|^2 \\ = (64\pi\mu^3 M_s / V) \Delta(\mathbf{k}_1 - \mathbf{k}_\lambda - \mathbf{k}_\nu) n_\lambda n_\nu (n_1 + 1) \\ \times |k_\lambda^z k_\lambda^- (k_\lambda)^{-2} + k_\nu^z k_\nu^- (k_\nu)^{-2}|^2. \end{aligned}$$

Setting the angle factor equal to  $\frac{1}{8}$  as in the confluence calculation, we find by the method of Appendix B that in the high-temperature limit  $k_B T \gg \hbar\omega_1$ ,

$$\begin{aligned} (1/T_{1k})_{\text{split}} &= (\pi\mu^3 M_s \tau / 2\hbar D^2 k_1) \\ &\times \int_{\hbar\omega_{LL}}^{\hbar\omega_{uL}} d(\hbar\omega_\nu) / \hbar\omega_\nu (\hbar\omega_1 - \hbar\omega_\nu), \quad (\text{C.2}) \end{aligned}$$

where  $\hbar\omega_{LL} = Dk_{LL}^2 + \hbar\omega_0$ ,  $\hbar\omega_{uL} = Dk_{uL}^2 + \hbar\omega_0$ , and  $k_{LL}$  and  $k_{uL}$  are the two roots of the equation

$$\frac{\hbar\omega_0}{2Dk_1^2} - \left( \frac{k}{k_1} \right) \left( 1 - \frac{k}{k_1} \right) = 0; \quad (\text{C.3})$$

thus

$$k_{LL} = \frac{k_1}{2} \left[ 1 - \left( 1 - \frac{2\hbar\omega_0}{Dk_1^2} \right)^{\frac{1}{2}} \right],$$

and

$$k_{uL} = \frac{k_1}{2} \left[ 1 + \left( 1 - \frac{2\hbar\omega_0}{Dk_1^2} \right)^{\frac{1}{2}} \right]. \quad (\text{C.4})$$

In deriving (C.2) a factor of  $\frac{1}{2}$  is included in  $1/T_{1k}$  to account for the equivalent output magnons, and in obtaining the expression (C.3) we made use of the

equation

$$\begin{aligned} \int_{-1}^1 d(\cos\theta_{v1})\delta(\hbar\omega_1 - \hbar\omega_v - \hbar\omega_\mu) \\ = (1/2Dk_1k_v) \int_{-1}^1 d(\cos\theta_{v1})\delta(\cos\theta_{v1} + \xi_0) \\ = \begin{cases} (1/2Dk_1k_v) & \text{for } \xi_0 \leq 1 \\ 0 & \text{otherwise,} \end{cases} \end{aligned}$$

where  $\xi_0 = [Dk_v^2 + (\hbar\omega_0/2)]/Dk_1k_v$ . With this expression for  $\xi_0$ , the inequality  $\xi_0 \leq 1$  may be written

$$\frac{\hbar\omega_0}{2Dk_1^2} - \frac{k_v}{k_1} \left(1 - \frac{k_v}{k_1}\right) \leq 0. \quad (\text{C.5})$$

This equation has real solutions for all  $k_v$ 's between the two values of  $k_v$  defined by the equality, which is just (C.3).

The splitting process cannot conserve energy (in the high-field approximation  $\omega_0 > \sim 2\omega_s/3$ ) unless  $k_1 \geq k_{td}$ , where the threshold wave vector  $k_{td}$  is defined by

$$k_{td} = (2\hbar\omega_0/D)^{1/2}. \quad (\text{C.6})$$

We obtain this threshold by noticing that momentum conservation requires that  $0 \leq k_v \leq k_1$ , and since the maximum value of  $(k_v/k_1)[1 - (k_v/k_1)]$  for  $k_v$  on this range is  $\frac{1}{4}$ , (C.5) has no real solutions unless

$$(\hbar\omega_0/2Dk_1^2) < (\frac{1}{4}).$$

On evaluating the integral in (C.2), we find

$$\left(\frac{1}{T_{1k}}\right)_{\text{split}} = \frac{\pi\mu^3 M_s \tau}{2\hbar D^2 k_1} \ln \left[ \left( \frac{\hbar\omega_1}{\hbar\omega_{LL}} - 1 \right) / \left( \frac{\hbar\omega_1}{\hbar\omega_{uL}} - 1 \right) \right]. \quad (\text{C.7})$$

We simplify this expression in the two limiting cases  $Dk_1^2 \gg 2\hbar\omega_0$  and  $Dk_1^2 \cong 2\hbar\omega_0$ .

### Relaxation Rate of $\pi/2$ Magnons for the Splitting Process in the Limit $Dk_1^2 \gg 2\hbar\omega_0$

For  $Dk_1^2 \gg 2\hbar\omega_0$ , (C.4) may be approximated by

$$k_{LL} \cong (\hbar\omega_0/2Dk_1), \quad k_{uL} \cong k_1 - k_{LL}.$$

Thus

$$\hbar\omega_{LL} \cong \hbar\omega_0 + [(\hbar\omega_0)^2/2Dk_1^2], \quad \hbar\omega_{uL} \cong \hbar\omega_1 - \hbar\omega_0.$$

On substituting these expressions into (C.7) we find

$$\left(\frac{1}{T_{1k}}\right)_{\text{split}} = \frac{\pi\mu^3 M_s \tau}{\hbar D^2 k_1} \ln \left( \frac{\omega_1}{\omega_0} \right),$$

which is valid for  $2\hbar\omega_0 \ll Dk_1^2 \ll k_B T$ .

### Relaxation Rate of $\pi/2$ Magnons for the Splitting Process in the Limit $Dk_1^2 \cong 2\hbar\omega_0$

For the other limiting case of  $Dk_1^2$  only slightly greater than  $2\hbar\omega_0$  we define  $k'$  as the small difference between  $k_1$  and  $k_{td}$ , where  $k_1$  is the magnitude of  $\mathbf{k}_1$ , and  $k_{td}$  is defined in (C.6); thus  $k' = k_1 - k_{td}$ . Then for  $k' \ll k_{td}$ , Eqs. (C.4) approximate closely to

$$k_{LL} \cong (k_{td}/2)[1 - (2k'/k_{td})^{1/2}],$$

and

$$k_{uL} \cong (k_{td}/2)[1 + (2k'/k_{td})^{1/2}].$$

We substitute these values of  $k_{LL}$  and  $k_{uL}$  into the equation  $\hbar\omega_{LL} = Dk_{LL}^2 + \hbar\omega_0$  and the same equation with  $LL$  replaced by  $uL$  and find

$$\hbar\omega_{LL} = (Dk_{td}^2/4)[1 - (2)^{1/2}(k'/k_{td})^{1/2}] + \hbar\omega_0,$$

and

$$\hbar\omega_{uL} = (Dk_{td}^2/4)[1 + (2)^{1/2}(k'/k_{td})^{1/2}] + \hbar\omega_0.$$

Finally, on substituting these expressions into (C.7) and expanding the logarithm, we obtain the result (40).