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Frequency Dependence of the Two-Magnon Ferrimagnetic Resonance Linewidth

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The Sparks-Loudon-Kittel (SLK) two-magnon linewidth theory is modified in a way that takes into account a more reasonable model of the scattering center due to magnetic inhomogeneities. The inverse-square frequency dependence of the linewidth at high frequencies given by SLK is eliminated, and a weak frequency dependence similar, but not identical, to that found by the theories of Geschwind and Clogston and of Schlömann is obtained. Experimental data ranging over a factor of about ten in applied field divided by magnetization ($\propto$frequency) gives excellent agreement with the modified SLK theory.

INTRODUCTION

THERE exists a considerable body of work[1–9] on the ferrimagnetic resonance linewidth due to magnetic inhomogeneities (e.g., porosity, variations of density or magnetization, anisotropy in polycrystals) in the sample under investigation. The effects of these inhomogeneities have been analyzed in detail[1–3,7] and the dependence of the linewidth on porosity and anisotropy in the case of polycrystalline samples[6,9] and on surface imperfections[4] in the case of single crystals has been well documented. The frequency dependence for low frequencies[5] has also been investigated and is in reasonable accord with experiment. The only serious flaw in the theory is the frequency dependence at high frequencies. The weak frequency dependence found by the classical theories of Geschwind and Clogston[2] and of Schlömann[3] (GC–S) is borne out experimentally. However, the quantum-mechanical calculation of Sparks, Loudon, and Kittel[7] (SLK), which roughly agrees with the classical calculation for low frequencies (strictly speaking for large ratios $4\pi M/H$, where $4\pi M$ is the saturation magnetization, and H is the applied field), yields an inverse square frequency dependence at large frequencies (small $4\pi M/H$) which is definitely not observed.[9] We have therefore undertaken the examination of this point and resolved the difficulty by considering a more realistic model of the two-magnon scattering process.

The SLK two-magnon linewidth theory[7] predicts that $\Delta H \sim \omega^{-2}$ for *spherical* samples in the high-field limit of large applied fields with respect to $4\pi M$. The ω^{-2} frequency dependence is not expected for samples of geometries other than spherical. The measurements we have made indicate that the linewidth is very nearly independent of frequency for this case of spherical samples at high fields. In the low-field limit, where the uniform precession comes near the top of the magnon band, a strong frequency dependence has been observed.[5] These results are explained below, where we see that the ω^{-2} frequency dependence arises from the dipole angle factor $3\cos^2\theta_k - 1$ in the scattering Hamiltonian. This factor arises from the spherical symmetry of the model used for the scattering center. When this spherical symmetry is destroyed by any of several possible means, the dipole factor must be replaced by a more general factor. This results in a nearly frequency-independent linewidth in the high-field limit.

EXTENSION OF THE SPARKS-LOUDON-KITTEL MODEL

First we shall review the SLK theory[7] of linewidth induced by surface roughness, voids between polycrystalline grains or nonmagnetic inclusions. The model of a spherical cavity in an infinite medium is taken for the scattering center. The standard transition probability result[10] is used for the probability per unit time TP of a transition in which a uniform precession magnon u is annihilated and a $k \neq 0$ magnon is created.

$$TP = (2\pi/\hbar)\,|\langle n_u - 1, n_k + 1 | \mathcal{H} | n_u, n_k \rangle|^2 \delta(\hbar\omega_k - \hbar\omega_u)\,, \quad (1)$$

where n_i denotes the number of magnons in state i and the scattering Hamiltonian is

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

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where $\mathbf{H}_D$ is the demagnetizing field of the cavity. This demagnetizing field is determined by

$$\nabla\cdot\mathbf{B}=\nabla\cdot[\mathbf{H}_D+4\pi\mathbf{M}\theta(r-R)]=0;\quad \nabla\times\mathbf{H}_D=0,$$

where R is the radius of our model cavity and the unit step function $\theta(x)=0$ for $x<0$ and 1 for $x>0$. With $\mathbf{H}_D=-\nabla\phi$, we have

$$-\nabla^2\phi=-4\pi\nabla\cdot\mathbf{M}\theta(r-R). \tag{3}$$

The solution is

$$\begin{aligned}\phi(\mathbf{r})&=-\int d\mathbf{r}'\frac{\nabla\cdot[\mathbf{M}(\mathbf{r}')\theta(r'-R)]}{|\mathbf{r}'-\mathbf{r}|}\\&=\phi_S(\mathbf{r})+\phi_V(\mathbf{r}),\end{aligned} \tag{4}$$

where the surface, $\phi_S(\mathbf{r})$, and volume, $\phi_V(\mathbf{r})$, terms are

$$\phi_S(\mathbf{r})=-\int d\mathbf{r}\frac{\delta(r'-R)\mathbf{M}(\mathbf{r}')\cdot\hat{r}'}{|\mathbf{r}'-\mathbf{r}|}, \tag{5a}$$

$$\phi_V(\mathbf{r})=-\int d\mathbf{r}'\frac{\theta(r'-R)\nabla\cdot\mathbf{M}(\mathbf{r}')}{|\mathbf{r}'-\mathbf{r}|}, \tag{5b}$$

where the caret denotes a unit vector. In the volume term the small cavity approximation is made, with $\mathbf{M}(\mathbf{r})\sim\exp(i\mathbf{k}\cdot\mathbf{r})$,

$$\phi_V(\mathbf{r})=-\int_{\text{all space}}d\mathbf{r}'\frac{\nabla\cdot\mathbf{M}(\mathbf{r}')}{|\mathbf{r}'-\mathbf{r}|}+\int_{\text{cavity}}d\mathbf{r}'\frac{\nabla\cdot\mathbf{M}(\mathbf{r}')}{|\mathbf{r}'-\mathbf{r}|}, \tag{6}$$

in which the integral over the cavity is neglected. The surface and volume terms make equal contributions to $\mathbf{H}_D$. The results are,[7,11] expanding $\mathbf{M}$ in magnon variables,

$$\mathcal{H}=16\pi^2R^3(\mu M/V)\sum_k(3\cos^2\theta_k-1)\times\frac{j_1(kR)}{kR}a_u{}^\dagger a_k+\text{c.c.}, \tag{7}$$

where V is the sample volume, 2μ=spectroscopic splitting factor times the Bohr magneton, M is the saturation magnetization, θ_k=the angle between the applied field and the wave vector $\mathbf{k}$, $j_1(kR)$ is the first spherical Bessel function, $a^\dagger$ and a are magnon creation and annihilation operators and u denotes the uniform precession mode. From Eq. (1) it is found[7,11] that the linewidth is

$$\Delta H_{\text{SLK}}=2\pi^2M(v/V)G_{\text{SLK}}(\cos\theta_u,\omega), \tag{8}$$

with

$$G_{\text{SLK}}(\cos\theta_u,\omega)=\frac{(3\cos^2\theta_u-1)^2}{\cos\theta_u}\frac{\omega_u}{\omega_i}, \tag{9}$$

where v is the total volume of all pits,

$$\omega_i=\gamma[H-(4\pi/3)M],\quad \omega_u=\gamma H$$

(for spherical samples), θ_u is the value of θ_k when $k=0$, and $\omega_k=\omega_u$, and is obtained below from Eq. (10). The correction factor ω_u/ω_i, which is important at low frequencies, arises[11] from using the true dispersion relation, Eq. (10), in the δ function in Eq. (1) rather than the approximate one previously used.[7] It has been assumed that the pits scatter independently; thus the total linewidth is given by the number of pits times $1/\gamma T_{\text{pit}}$ for a single pit. The frequency dependence arises only from the factor G. A graph of $(3\cos^2{}_u\theta e-1)^2/\cos\theta_u$ as a function of $\cos\theta_u$ is shown in Fig. 1. The zero of the function at $\cos^2\theta_u=\frac{1}{3}$ is at the value of $\cos\theta_u$ for the uniform precession for a spherical sample in the high-field limit. Changes in the applied field give rise to *small* changes in $\cos\theta_u$, but the corresponding changes in $(3\cos^2\theta_u-1)^2$ are *large* since we are near a zero of this function. This is the reason for the strong ω^{-2} frequency dependence. If there were no zero at $\cos^2\theta_u=\frac{1}{3}$, the linewidth would be very nearly independent of frequency. The dispersion relation[12] is given by

$$\hbar\omega_k=[(Dk^2+\hbar\omega_i)(Dk^2+\hbar\omega_i+\hbar\omega_m\sin^2\theta_k)]^{1/2}, \tag{10}$$

where $\omega_m=4\pi\gamma M$. Setting $k=0$ and $\omega_k=\omega_u=\gamma H$, we easily find

$$\cos\theta_u=\left[\frac{3-2\alpha}{3(3-\alpha)}\right]^{1/2}, \tag{11}$$

where $\alpha=4\pi M/H$. For small α (high frequencies) the factor G_{SLK} then becomes

$$G_{\text{SLK}}=\alpha^2/(3)^{3/2}=1/(3)^{3/2}[\omega_m/\omega_u]^2 \tag{12}$$

and gives rise to the inverse square frequency dependence.

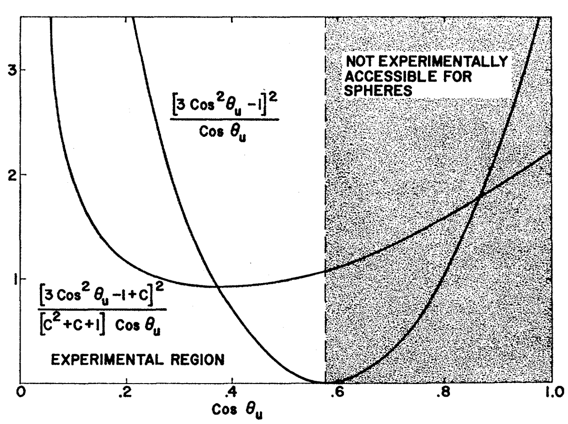

FIG. 1. Plot of the angular factors for both the original and modified SLK model.

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Now the factor $(3\cos^2\theta_u-1)^2$ in Eq. (9) which gives rise to the zero of G_{SLK} at $\cos^2\theta_u=\frac{1}{3}$ comes directly from the $(3\cos^2\theta_k-1)$ factor in the Hamiltonian Eq. (7) and is very strongly model-dependent. It arises only for a model with complete spherical symmetry. The order of magnitude of the other factors in Eq. (7), however, are expected to be less sensitive to the particular model used. This spherical symmetry of Eq. (7) can be destroyed in any of several ways: (1) The actual pits have quite irregular shapes, not spherical in general. (2) For surface pits, the shape would be more closely approximated by a hemisphere, rather than a sphere, and they lie on a surface rather than in an infinite medium. (3) When two or more pits appear close together the spherical symmetry is destroyed. (4) In the polycrystalline case, when a void lies near the surface of the sample, the spherical symmetry is destroyed. The arithmetic becomes quite complicated when any nonspherical symmetry model of which we have thought is introduced. However, some general considerations of nonspherical models can be made.

We consider a simple model which gives the observed weak frequency dependence. The model takes into account the fact that even for a nonspherical cavity the dipole contribution to the demagnetization is expected to be rather large. Therefore, we keep the dipole term and replace all higher order multipoles by an average value which we take to be constant (c). Specifically, in the square of the Hamiltonian in Eq. (7) we replace

$$(3\cos^2\theta_k-1)^2\rightarrow\frac{(3\cos^2\theta_k-1+c)^2}{\int_0^1 d\cos^2\theta_k[3\cos^2\theta_k-1+c]^2}=\frac{(3\cos^2\theta_k-1+c)^2}{c^2+c+1},\quad(13)$$

the integral in the denominator being a normalizing factor which averages linearly over magnetic field. Thus, Eq. (8) still gives the linewidth if we replace G_{SLK} by the modified function

$$G_{\mathrm{SS}}=\frac{\omega_u}{\omega_i}\frac{(3\cos^2\theta_u-1+c)^2}{(c^2+c+1)\cos\theta_u}.\quad(14)$$

Notice that the factor $\omega_u/\cos\theta_k\omega_i$ comes from energy conservation and not from the geometry of the model. It is a density of degenerate states factor and is expected to remain for any model. A graph of

$$(3\cos^2\theta_u-1+c)^2/(c^2+c+1)\cos\theta_u$$

is shown in Fig. 1.[13] The zero at $\cos^2\theta_u=\frac{1}{3}$ has disappeared and consequently so has the inverse square frequency dependence. This approximation cannot, of course, be expected to give the exact dependence of linewidth on $\cos^2\theta_u$, but the order of magnitude, the absence of a zero at $\cos^2\theta_u=\frac{1}{3}$, and the general weak frequency dependence for $\alpha\ll1$ are expected to be correct.

The following consideration shows that this approximation is not unreasonable: Consider as a model for a surface pit, or a cavity between polycrystalline grains, or a nonmagnetic inclusion, a cavity of arbitrary irregular shape in an infinite medium. The demagnetization field is still described by Eq. (5) if the spherical surface $r'=R$, reflected in $\delta(r'-R)$ and $\theta(r'-R)$, is replaced by the actual irregular surface of the cavity. The small cavity approximation in which the integral over the volume of the cavity in Eq. (6) is neglected in the volume term ϕ_V, can be applied to our irregularly shaped cavity as well as to the spherical cavity. Thus, the demagnetization field arising from ϕ_V is the same as for a spherical cavity and gives rise to a factor $(3\cos^2\theta_k-1)$. In the spherical case this volume term accounts for one-half the total demagnetization field, i.e., for one-half the Hamiltonian. The surface term ϕ_S for the irregular cavity consists of many higher order multipole terms in addition to a dipole term. The approximation made in the model is then to replace the spherical result $(3\cos^2\theta_k-1)$ in the surface contribution by a constant.[14]

Since Eq. (14) does not have a zero for $\cos^2\theta_u$ near $\frac{1}{3}$, the model predicts a linewidth nearly independent of frequency for spherical samples at large applied fields. The detailed comparison with experiment will be given in the next section. Before proceeding to the experiment, however, we should examine how the above considerations compare with the classical theories. This may be done quite easily for the results of Geschwind and Clogston by looking at Eq. (7) of Ref. 2, which reduces to the following for a spherical sample[15]

$$G_{\mathrm{GC}}=\tfrac{1}{6}(\omega_u/\omega_i)(1/\cos\theta_u)\left[1+\frac{5}{6}\frac{9+(3-\alpha)^2}{3-\alpha}\right].\quad(15)$$

The factors $\omega_u/\omega_i\cos\theta$ arise from the density of states and therefore should be present in any theory. The third factor is just that arising from the Hamiltonian implicit in the problem. Since Schlömann's[3] analysis yields the same result as GS, a similar comparison is possible and in fact is most obvious from his Eq. (47b) which is of a similar form to our Eq. (1), i.e., an effective Hamiltonian matrix element times a δ function in frequency. The δ function yields the density of states factors $\omega_u/\omega_i\cos\theta$ so that the essential difference in the calculation is again in the form of the implicit Hamiltonian. Unfortunately, since the Hamiltonian is not explicit it is difficult to compare it to ours on a physical basis.

[13] The value $c=2.44$, the experimental value, is used.

[14] For the case of surface pits, which are approximately hemispherical, ϕ_V will also contain an appreciable nondipolar part which we also consider to be contained in the constant term.

[15] The symbol J is used in Ref. 2 for what we here call G.

EXPERIMENTAL RESULTS AND COMPARISON WITH THEORY

Since the frequency factors in the two-magnon relaxation theory under consideration here always enter in the ratio of magnetization to frequency

$$(\alpha=4\pi M/H=\omega_m/\omega_u),$$

we may conveniently study the linewidth as a function of α by two different experiments, firstly by linewidth measurements directly as a function of frequency, and secondly, measurements as a function of magnetization (changing the magnetization by varying the temperature). Both these measurements have been made on single crystal spheres of yttrium iron garnet whose surfaces have been sufficiently roughened to insure dominance of the two-magnon process ($\Delta H=15$ Oe at 10 kMc/sec and room temperature). In addition, the frequency dependence has been measured on a crystal of lithium ferrite ($\Delta H=31$ Oe at 10 kMc/sec).[16] The frequency measurements extend from 8 to 35 kMc/sec and the temperature measurements from 77 to 530°K which corresponds to changes in $4\pi M$ from the order of 2500 to 700 G.

The data are plotted in Fig. 2 in a manner which allows easy comparison between the present theory and the classical theory (GC–S). Since in both these theories the linewidth reaches a constant value as $\alpha\rightarrow 0$ we plot $G/G_{\alpha=0}$ to obtain the theoretical curves. The experimental points are then plotted by using the relation

$$\Delta H=K4\pi M(G/G_{\alpha=0}). \tag{16}$$

The value of K is found by extrapolation from the datum of smallest α. Since the value of $G/G_{\alpha=0}$ is very close to the same for both theories in this range we have a valid one-point normalization of the data. The frequency dependence is then clearly observed in Fig. 2 from the plot of $G/G_{\alpha=0}$ as a function of α^{-1}. The curve for the G_{SS} is obtained by using $c=2.44$ and the fit to the data is seen to be very good,[17] much better than the fit to G_{GC-S} which has previously been used. The reason that the discrepancy of the fit to G_{GC-S} was not previously observed is that for the work on yttrium iron garnet α^{-1} varied from only 1.2 to 2.6 which results in a maximum deviation from G_{GC-S} of only about 15%, which is not very large compared to experimental error and is of the order of observed deviations found in Ref. 9. For the work on manganese ferrite,[9] the correction due to finite crystal linewidth was large enough to swamp out appreciably the difference between G_{SS} and G_{GC-S}. In the experiments reported here, α^{-1} runs from 0.75 to 7.2 and the deviation from G_{GC-S} is obvious.

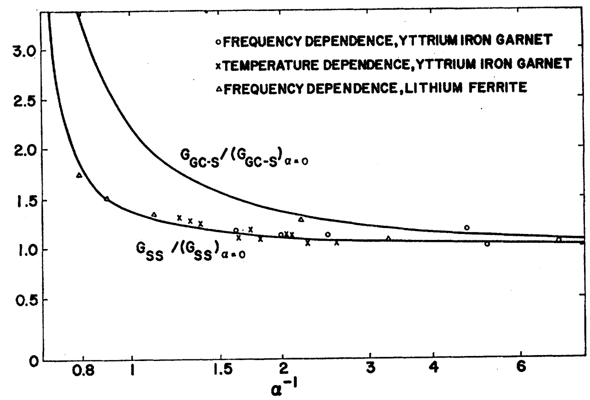

FIG. 2. G_{GC-S} and G_{SS} as a function of α^{-1} together with the experimental data.

CONCLUSION

We have shown that the calculation of two-magnon scattering by the theory of Sparks, Loudon, and Kittel, modified by considering a more realistic model of the scattering center, eliminates the inverse square frequency dependence previously found and gives a result for the frequency dependence in accord with experiment. Although it has not been possible to calculate a rigorous Hamiltonian in this case, the approximation used is certainly justified on the basis of the known structure of polycrystalline samples.

[16] Some polycrystalline measurements have previously been done over a smaller range of α and yield similar results if one takes into account the effects of both anisotropy and porosity. See Refs. 5 and 9.

[17] This value for c is roughly twice the root-mean-square value of the dipole term and suggests that the other higher order multipoles might be important.