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P. Dorsey

J. B. Sokoloff
Northeastern University

C. Vittoria
Northeastern University

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P. Dorsey, J. B. Sokoloff, and C. Vittoria
Northeastern University, Boston, Massachusetts 02115

We present calculations of the ferrimagnetic resonance lineshapes resulting from second order Suhl instabilities for thin films and spheres. We find that whereas a spherical sample has a lineshape which is symmetrical around the resonant frequency, a thin film has an asymmetrical lineshape. The calculations are in agreement with measurements that we have performed of the lineshape as a function of input power for thin film samples of both barium ferrite and yttrium iron garnet. When the magnetic field direction is changed from perpendicular to parallel to the film plane, the asymmetry of the lineshape at magnetic resonance changes in opposite sense relative to the resonant field. Theoretical estimates of the critical microwave field necessary for second order Suhl instabilities to occur are in agreement with measured critical fields.

I. INTRODUCTION

It is well known that nonlinear microwave effects manifest themselves quite readily in ordered magnetic materials which exhibit narrow ferromagnetic resonance (FMR) linewidths, such as yttrium iron garnet (YIG) and barium ferrite.[1] The FMR linewidth of YIG at 9.6 GHz is as small as 0.25 Oe and that of barium ferrite is 23 Oe at 70 GHz. In FMR experiments with the microwave magnetic field transverse to the applied static magnetic field, nonlinear effects have been observed as a broadening of the resonance linewidth and the appearance of a broad subsidiary maximum at half the resonant field. At higher power, hysteretic behavior known as foldover effects have also been observed in FMR experiments.[2] Besides changes in the lineshape chaotic effects have been observed in single crystal samples of YIG.[1] In a chaos experiment the response as a function of time and power are measured. In order to describe either type of experiment, one must invoke a nonlinear theory.

In this article, we apply Suhl's theory of a second order instability[3,4] due to nonlinearity to calculate the FMR lineshape for films excited at the bottom and top of the zero wavevector spinwave manifold. This corresponds to putting the external magnetic field perpendicular and parallel to the film plane. We predict a characteristic asymmetry of the lineshape which depends on the direction of the external field relative to the film plane. The calculated lineshape is in reasonable agreement with our FMR measurements at high power on thin film samples of both barium ferrite and YIG. We take advantage of the fact that in the thin film configuration the uniform mode can be made to fall at the top or bottom of the zero wavevector magnon manifold. We propose that by observing the lineshape as a function of microwave power in an FMR experiment, it should be possible to identify the onset of nonlinearity. Specifically, the occurrence of an asymmetric lineshape in an FMR experiment signifies the onset of nonlinear spin interactions in ordered magnetic materials. In Sec. II, we describe the nonlinear theory based on Suhl's work on the theory of spinwave excitation at high power and in Sec. III, the FMR experiments at high power. The concluding remarks are given in Sec. IV.

II. THEORY

Suhl instabilities near the FMR frequency (i.e., second order Suhl instabilities) lead to a broadening of the line and a reduction in its intensity.[1,3,4] For a thin film sample with the dc magnetic field applied perpendicular to the film, the resonance frequency ω_{res} occurs at the bottom edge of the magnon manifold. Thus, for frequencies slightly above ω_{res}, the uniform precession ($k=0$) magnon is energetically degenerate with other (primarily $k \neq 0$) magnons which could become unstable, but for frequencies below ω_{res}, there are no magnon modes. In this section we calculate the effect of the interaction between the ($k=0$) uniform magnon and the other magnons degenerate with it. We shall see that since this interaction can occur only for $\omega > \omega_{res}$, it leads to an asymmetric lineshape.

Our calculation of the asymmetric lineshape follows the discussion of Ref. 4, in which ω was set equal to ω_{res}. We extend Suhl's calculations to $\omega < \omega_{res}$ and $\omega > \omega_{res}$. It is necessary to extend the calculations to all frequencies near ω_{res} in order to obtain a lineshape. Suhl's discussion begins with the following equation of motion for the magnetization $\mathbf{M}(\mathbf{r},t)$ in an effective field $\mathbf{H}_{eff}$, which includes both the applied and exchange fields,

$$\frac{d\mathbf{M}(\mathbf{r},t)}{dt} = -\gamma[\mathbf{M}(\mathbf{r},t) \times \mathbf{H}_{eff}] + \text{damping term}. \quad (1)$$

Since the precessional motion is circular, it is convenient to introduce the variables $M^{\pm} = M_x \pm iM_y$. Then, Eq. (1) becomes

$$\dot{M}^+ = iM^+[\omega_H + \omega_{dz} + \omega_{ex}l^2\nabla^2 M_z] - M_z[\gamma h_s e^{i\omega t}$$
$$+ \omega_{ex}l^2\nabla^2 M^+ + \omega_{dx} + i\omega_{dy}], \quad (2)$$

where $\omega_H = \gamma H$, $\omega_{d\alpha} = \gamma N_\alpha M_\alpha$, and $\alpha = x, y,$ and z. H is the external applied field, N_α is the demagnetizing factor in the α direction and $\gamma = ge/2mc$. The following transformation:

$$a_k = \lambda_k b_k - \mu_k b^*_{-k}, \quad (3a)$$

$$a^*_{-k} = -\mu^*_k b_k + \lambda_k b_{-k}, \quad (3b)$$

where $a_k = (2\pi)^{-1}\int d^3r M^+(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}}$, the Fourier transform of M^+, diagonalizes the linear approximation to the

equations of motion in terms of the spinwave variable b_k. λ_k and μ_k are chosen so that the linear approximation to Eq. (2) will reduce to

$$\dot{b}_k = i\omega_k b_k, \tag{4a}$$

$$\dot{b}^*_{-k} = i\omega_k b^*_{-k}, \tag{4b}$$

for $k \neq 0$, where ω_k is the spinwave frequency. The solution of the linear approximation to Eq. (2) for $k = 0$ (i.e., the uniform precession mode) is[3]

$$a_0 = \gamma h_s e^{i\omega t}[(\omega_{res} - \omega) + i\eta_0]^{-1} = a_0^0 e^{i\omega t}, \tag{5}$$

where h_s is the microwave field, η_0 is the intrinsic linewidth, and a_0 is the amplitude of the uniform magnon. To second order in a_0^0, if we write b_k as $b_k^0 e^{i\omega_k t}$, Eq. (2) becomes[3]

$$\dot{b}_k^0 = \xi_k a_0^2 (b_{-k}^0)^* e^{2i(\omega - \omega_k)t}, \tag{6}$$

where ξ_k is a combination of various parameters in the problem, given in Eq. (5) of Ref. 3. The other nonlinear terms in Eq. (6) are neglected here because they oscillate much more rapidly than the term on the right hand side of Eq. (6) for ω close to ω_k. Setting $b_k = v_k e^{i(\omega - \omega_k)t}$ and substituting it in Eq. (6) gives

$$\left(\frac{d}{dt} + i(\omega - \omega_k)\right)v_k = i\xi_k (a_0^0)^2 v_{-k}^*, \tag{7}$$

which when combined with the corresponding equation for v_{-k}^* gives

$$\left(\frac{d^2}{dt^2} + (\omega - \omega_k)^2 - \xi_k^2 |a_0^0|^4\right)v_k = 0, \tag{8}$$

whose solution increases exponentially for $|a_0^0|^4 \rangle (\omega - \omega_{res})^2$, i.e., for sufficiently large h_s or microwave power to make $|a_0^0|^4$ greater than this critical value [as seen from Eq. (5)]. Adding a dissipative term and a linear coupling to the uniform mode (which is responsible for the scattering of the uniform mode into the $k \neq 0$ modes due to scattering by surface pits) to Eq. (7), we obtain

$$i\dot{v}_k = \omega_k v_k + \nu_{k0} a_0 + \xi_k a_0^2 v_{-k}^* - i\eta_k v_k, \tag{9a}$$

$$-i\dot{v}_{-k}^* = \omega_k v_{-k}^* + \nu_{k0} a_0^* + \xi_k^* (a_0^*)^2 v_k + i\eta_k v_{-k}^*, \tag{9b}$$

where η_k is the linewidth of the magnon of wavevector k described by Eq. (9), ν_{k0} is the strength of its coupling to the uniform magnon mode, and $\xi_k = 0.5\lambda_k^2(\omega_{ex}l^2 k^2 - N_T \omega_d + \omega_d(k_z^2/k^2))$, where N_T is the demagnetizing factor in the plane of the film. Equations (9a) and (9b) are identical to equations above Eq. (iii) in Ref. 4. The corresponding linearized equation for the uniform mode is

$$i\dot{a}_0 = \omega_{res} a_0 + \sum_k \nu_{0k} v_k - i\eta_0 a_0 + h_s e^{i\omega t}. \tag{9c}$$

Solving Eq. (9b) for v_k^*, assuming that $v_k \propto e^{-i\omega t}$, we obtain

$$v_{-k}^* = \frac{\nu_{k0} a_0^* + \xi_k^* (a_0^*)^2 v_k}{\omega - \omega_k - i\nu_k}$$

and substituting this expression for v_k^* in Eq. (9a), we obtain

$$v_k = \nu_{k0} a_0 \frac{\xi_k |a_0|^2 + (\omega - \omega_k - i\nu_k)}{(\omega - \omega_k)^2 + \nu_k^2 - |\xi_k|^2 |a_0|^4}.$$

This expression is then is substituted into Eq. (9c) for v_k to give

$$a_0^0 = h_s[(\omega - \omega_{res} - \eta_{sp} S_1) + i(\eta_0 + \eta_{sp} S_2)]^{-1}, \tag{10a}$$

where a_0^0 is defined by $a_0 = a_0^0 e^{-i\omega t}$ and

$$S = S_1 + iS_2 = \sum_k N(\omega_k)^{-1} \frac{\omega - \omega_k + \xi_k |a_0^0|^2 - i\eta_k}{\eta_k^2 - \xi_k^2 |a_0^0|^4 + (\omega_k - \omega)^2}, \tag{10b}$$

where $N(\omega_k)$ is the magnon density of states and η_{sp} is the contribution to the spinwave linewidth due to two magnon scattering (primarily by surface pits) in the low power limit. To obtain Eq. (10), we have used the Fermi golden rule expression for η_{sp}, namely $\eta_{sp} = \pi \nu_{k0}^2 N(\omega_k)$ to eliminate ν_{k0}. The quantity S represents the "self-energy" whose real part gives the frequency shift and whose imaginary part gives the damping of the uniform magnon mode caused by its interaction with other magnons. The appearance of $|a_0^0|$ in Eq. (10b) indicates that S includes the effects of nonlinearity. In the limit as a_0^0 becomes zero, the real and imaginary parts of S reduce to usual expression for the frequency shift and damping, respectively, in the low power limit. Following Ref. 4, we replace the summation over k by an integral over ω_k, neglecting the variation of $N(\omega_k)$, η_k and ξ_k with k since most of the contribution to the integral comes from the vicinity of $\omega_k = \omega$. Furthermore, we will assume that the contribution to S_1 from the integral resulting from the term with the $\omega - \omega_k$ factor in the numerator of the expression that is being summed over in Eq. (10b) has been absorbed into ω_{res}. This is justified if η_k and ξ_k are sufficiently small. In that case this contribution will have very little dependence on $|a_0^0|^4$. Then, we find for the imaginary and real parts of S for applied field perpendicular to the film

$$S_2 = (1 - |a_0^0/a_c|^4)^{-0.5}\{\pi/2 - \arctan[\eta_0^{-1}(\omega_{res} - \omega)$$

$$\times (1 - |a_0^0/a_c|^4)^{-0.5}]\}, \tag{11a}$$

$$S_1 = \pi^{-1} |a_0^0/a_c|^2 S_2, \tag{11b}$$

where $a_c = (\eta_k/\xi_k)^{0.5}$ for $|a_0^0| < a_c$, and for $|a_0^0|$ greater than this value the quantity $(1 - |a_0^0/a_c|^4)^{-0.5}$ is replaced in Eq. (11a) and (11b) by an infinitely large quantity. The quantity a_c is the value of a_0^0 at which nonlinearity becomes important. Let us take the square of the absolute value of Eq. (10a),

$$|a_0^0|^2 = h_s^2[(\omega - \omega_{res} - \eta_{sp} S_1)^2 + (\eta_0 + \eta_{sp} S_2)^2]^{-1}. \tag{12}$$

Although it will be seen shortly that for nonzero values of η_{sp} there is really not a sharp threshold value of the microwave field at which the second order Suhl instability begins to occur, we may still define a characteristic field near which the Suhl instability becomes important (h_c) by

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$$a_c = h_c \chi_0 = h_c [\omega - \omega_{\text{res}} + i(\eta_0 + \eta_{\text{sp}} S_2^0)]^{-1}, \qquad (13)$$

where χ_0 is the susceptibility one would have if there were no nonlinearity (i.e., if the parameter ξ_k were zero) and S_2^0 is the value of S_2 at a_0^0 equal to zero. S_2^0 is nearly equal to π for ω well into the magnon continuum and 0 well below the continuum. Then the field h_c represents the microwave field that would produce a uniform precession amplitude equal to a_c if there were no nonlinearity present. Combining Eqs. (12) and (13), we have

$$|a_0^0/a_c|^2 P_c/P = \frac{(\omega - \omega_{\text{res}})^2 + (\eta_0 + \eta_{\text{sp}} S_2^0)^2}{(\omega - \omega_{\text{res}} + S_1)^2 + (\eta_0 + S_2)^2}, \qquad (14)$$

where the ratio of the microwave power to the power at microwave field h_c is given by $P = h_s^2/h_c^2$. Equation (14) can be solved graphically by plotting its right and left hand sides as a function of $|a_0^0|^2$, as was done in Ref. 3 for the first order Suhl instability. This is illustrated in Fig. 1. It should be noted that the plot of the right hand side of Eq. (14) drops to zero when $\omega - \omega_{\text{res}}$ greater than 0 but not when it is less than 0. This occurs because in the second case, S_2 in Eq. (11a) becomes equal to $(\omega_{\text{res}} - \omega)^{-1}$ when the factor $(1 - |a_0^0/a_c|^2)^{-0.5}$ becomes infinitely large, whereas for the case of $(\omega - \omega_{\text{res}})$ greater than zero, S_2 becomes infinitely large. In Ref. 3 the right hand side was a step function, which implied that when the intersection occurred on the vertical part of the step function, $|a_0^0|$ would remain fixed at the value at which the vertical part of the step function occurs, which means that $|a_0^0|$, the uniform mode amplitude will saturate at a fixed value once the microwave field is above the threshold for the first order Suhl instability. In our case the right hand side is not a step function, and hence, the uniform mode amplitude will not saturate until the microwave field becomes infinitely large. (Actually, in the limit as η_{sp} becomes zero, the right hand side of our equation will become a step function, and we will again get saturation of the uniform mode amplitude above threshold.) From Eq. (14) and the definition of the susceptibility $\chi = a_0^0/h_s$, we find

$$|\chi/\chi_0|^2 = \frac{(\omega - \omega_{\text{res}})^2 + (\eta_0 + \eta_{\text{sp}} S_2^0)^2}{(\omega - \omega_{\text{res}} + S_1)^2 + (\eta_0 + S_2)^2}, \qquad (15)$$

and

$$\text{Im} \, \chi = \text{Im} \, \chi_0 |\chi/\chi_0|^2 (\eta_0 + \eta_{\text{sp}} S_2)/(\eta_0 + \eta_{\text{sp}} S_2^0). \qquad (16)$$

Equation (14) was solved graphically for $|a_0^0/a_c|^2$ and $\text{Im} \, \chi$ was calculated from Eqs. (15) and (16) for a few values of the microwave power $P \propto h_s^2$. The results are illustrated in Fig. 2, where $\text{Im} \, \chi$ is plotted as a function of $\omega_{\text{res}} - \omega$ for several values of the microwave power. Since our samples have narrow FMR linewidths, η_{sp} is expected to be comparable to η_0 (the intrinsic limit), and hence, we took $\eta_{\text{sp}} = \eta_0$ in these calculations. Since ω_{res} increases approximately linearly with the dc field H, this is effectively a plot of $\text{Im} \, \chi$ as a function of H. What we see is that since the resonant peak occurs right at the lower edge of the magnon continuum, the half of the resonant peak in $\text{Im} \, \chi$ which is inside the magnon continuum (i.e., for $\omega > \omega_{\text{res}}$) is suppressed in intensity more than the half which is below

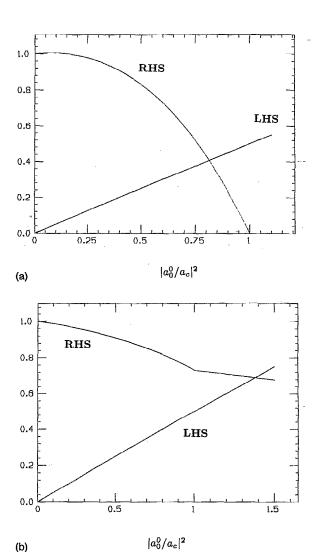(a) $|a_0^0/a_c|^2$

(b) $|a_0^0/a_c|^2$

FIG. 1. The solution of Eq. (14) is illustrated. The left and right hand sides of Eq. (14) (denoted by LHS and RHS, respectively) are plotted as a function of $|a_0^0/a_c|^2$ for $P/P_c = 2$ and for (a) $(\omega - \omega_{\text{res}})/\eta_{\text{sp}} = 1$ and (b) for $(\omega - \omega_{\text{res}})/\eta_{\text{sp}} = -1$.

(above) the continuum in frequency (field). [Incidentally, there is still some effect of nonlinearity, even below the continuum because of the mixing of the magnon states caused by scattering from surface pits (i.e., because of the ν_{k0} parameter in Eq. (9)]. The net result is that for the power P well above P_c there is a peak for ω just below the magnon continuum, which has a steeper slope on the high frequency side of the peak than on the low frequency side. Just above this peak in frequency there exists a shoulder, which peaks just above the bottom of the magnon continuum. For P/P_c equal to 2, the shoulder and the peak have nearly the same intensity, and for $P = P_c$ they have merged into one peak, although the peak is still asymmetrical. (Of course, for the limit as a_0^0 becomes zero, the peak is a Lorentzian symmetrically centered around ω_{res}.)

Now let us consider the FMR lineshapes expected for two sample geometries, spherical and planar. The self-energy term due to nonlinearity S in Eq. (12) will become infinitely large only when $|a_0^0|$ becomes equal to or greater
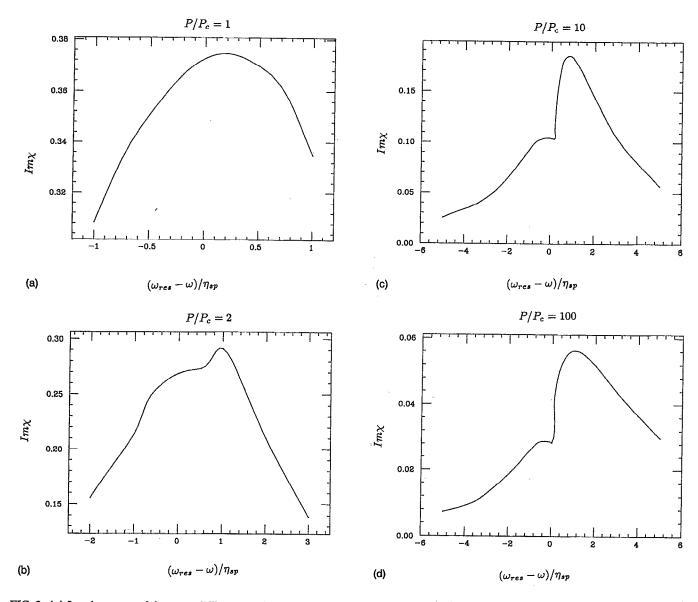
FIG. 2. (a) Imaginary part of the susceptibility versus $(\omega_{res}-\omega)/\eta_{sp}$ for $\eta_0=\eta_{sp}$ and for (a) $P/P_c=1$, (b) $P/P_c=2$, (c) $P/P_c=10$, (d) $P/P_c=100$ for the dc magnetic field perpendicular to the sample.

than a_c. When ω lies in the spinwave continuum, this critical value can be fairly small, and there will be a solution to Eq. (2) for small critical fields. Using Ref. 3 or 5 we estimate that for barium ferrite, for which the intrinsic linewidth is about 30 Oe at a frequency of 80 GHz, the critical electromagnetic field is only 1 Oe and about 1 mOe for a YIG sample with a linewidth of 1 Oe at 9.5 GHz. When we are outside the continuum, the required critical field becomes much larger. Thus, for the case of thin films, for which the resonant frequency lies at the bottom of the magnon continuum,[5] the resonant lineshape will be asymmetrical as illustrated in Fig. 2 because the Suhl effect will only occur for frequencies in the magnon continuum (i.e., above the resonant frequency).

If the dc magnetic field is applied parallel to the film, the uniform magnon will occur at the top of the spinwave manifold (see Fig. 3). This corresponds to magnons propagating within the plane of the film for which the parameter ξ_k is zero if $k=0$ because in the expression for ξ_k given

under Eq. (9), N_T is zero and at the top of the $k=0$ magnon manifold $k_z=0$. This implies that a_c is infinite. For nonzero k, however, the threshold field is not infinite. Using the fact that the spinwave stiffness for YIG is $\sim 10^{-9}$ Oe cm^2 and that ω_d is about 140 Oe, we estimate that for k of about 10^5 cm^{-1} the value of a_c and thus the critical field will be comparable to the value it would have if ω_{res} were at the bottom of the magnon manifold. When ω is increased sufficiently far above the top edge of the $k=0$ continuum (see Fig. 3), however, the Suhl instability will no longer occur. Even though nonzero wavevector magnons degenerate with ω to become unstable still exist, they have values of k much larger than the inverse size of a surface pit (about 10^5 cm^{-1}). Since the parameter ν_{k0} drops to zero when k becomes much larger than the inverse pit size,[5] it can be taken to be negligibly small. Thus, all (nonzero k) modes above a certain frequency near the top of the magnon manifold are decoupled from the uniform mode. Therefore, we expect an asymmetric lineshape to
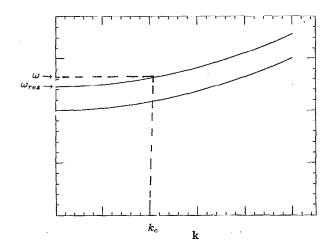
FIG. 3. A sketch is given of the magnon manifold for the case of H parallel to the film. Here ω is the driving frequency and k_c is the wavevector of the unstable mode corresponding to frequency ω.

occur at the top of the spinwave manifold like that which occurs when the resonance occurs at the bottom of the continuum, except that the lineshape will be the mirror image of the lineshape which occurred for the latter case. By comparison, when the sample is spherical, the resonant frequency will lie within the spinwave manifold,[5] and hence, the lineshape will be symmetrical, although the Suhl instability will lead to a broadening of the linewidth.

III. EXPERIMENTAL RESULTS

We have performed FMR experiments on films of both barium ferrite (measured linewidth 30 Oe at 80 GHz) and YIG (measured linewidth 1 Oe at 9.5 GHz) produced by the laser ablation technique.[6] High power experiments were performed on YIG films with the dc magnetic field both perpendicular and parallel to the film plane using standard FMR cavity perturbation technique. The resonant frequency of the cavity was 9.5 GHz. Barium ferrite films, however, were placed in a waveguide arrangement and the frequency of operation was 80 GHz. The experiments consisted of sweeping the magnetic field through the magnetic resonance at fixed frequency. The input power was measured for each sweep. The critical power for the onset of nonlinearity was determined by noting the power at which the lineshape was visibly distorted. Representative scans are shown in Figs. 4 and 5. As can be seen, these results compare quite favorably with our theoretical calculations based on Suhl's theory shown in Fig. 2 in the sense that the slope of the absorption spectrum is steeper for $\omega > \omega_{res}$ or H less than the resonant field (i.e., inside the spinwave continuum) than for $\omega < \omega_{res}$ (as can be seen in Fig. 4 by the fact that the peak in the derivative spectrum shown in this figure on the low field side of the resonance is larger than the one on the high field side). That is, the experiments qualitatively agree with the shape of the major peak in the calculated absorption spectrum shown in Fig.
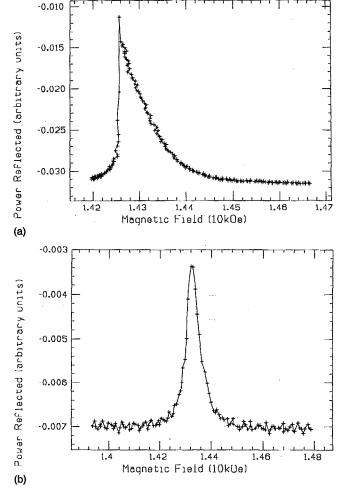

(a)

(b)

FIG. 4. Measurements of the FMR power absorption as a function of the dc field, applied perpendicular to the film, for frequency fixed at 80 GHz for films of barium ferrite, (a) for $P > P_{crit}$, (b) for $P < P_{crit}$.

2. The shoulder found in these calculations was not observed, probably because of the uncertainty in the location of the baseline in the scan.

In order to support the idea that the asymmetry comes about because of the location of the resonant frequency, we have also presented FMR measurements with the dc field parallel to the film. The results are presented in Fig. 6. As can be seen, the resulting spectrum is approximately the mirror image of the FMR spectrum with the field perpendicular to the film, which is what is expected since changing the dc field direction shifts the resonant frequency (field) from the bottom (top) to the top (bottom) of the $k = 0$ magnon continuum. As mentioned earlier, although for ω_{res} well above the zero wavevector magnon continuum unstable modes still exist, since these modes must be of nonzero wavevector and since v_{0k} becomes negligibly small for k greater than the reciprocal of a typical pit size,[5] these modes will make a negligible contribution and hence there will effectively exist a top edge of the $k = 0$ magnon continuum, namely the value of ω_k for which v_{k0} becomes nearly zero. The critical microwave fields for the onset of nonlinearity determined in these experiments were 5.8 mOe for YIG and 300 mOe for barium ferrite, which are
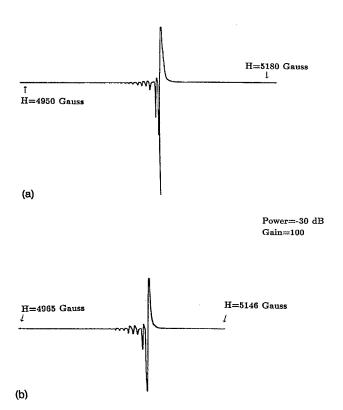
H=5180 Gauss

H=4950 Gauss

(a)

H=2514 Gauss

H=2695 Gauss

(a)

H=4965 Gauss

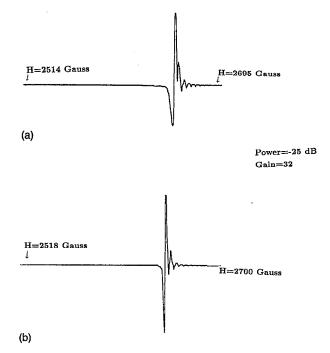H=5146 Gauss

(b)

H=2518 Gauss

H=2700 Gauss

(b)

FIG. 6. Measurements of the derivative of the FMR power absorption as a function of dc field, applied parallel to the film, for fixed frequency for films of YIG (a) for $P > P_{crit}$, (b) for $P < P_{crit}$.

FIG. 5. Measurements of the derivative of the FMR power absorption as a function of dc field, applied perpendicular to the film, for frequency fixed at 9.5 GHz for films of YIG (a) for $P > P_{crit}$, (b) for $P < P_{crit}$.

comparable to the estimates given earlier of the critical fields for the occurrence of Suhl instabilities, lending further credence to our suggestion that the asymmetric lineshapes observed by us are due to nonlinear effects. We find no evidence of "foldover" effects[2] since we obtain the identical spectrum no matter which way we sweep the dc field. Furthermore, our microwave fields are well below the threshold for "foldover" effects.[2] In order to rule out the possibility that our observed line assembly was due entirely to heating effects, we have repeated our measurements on barium ferrite films with the sample mounted on the metallic short. The asymmetry was still observed. The very

fine cusp [in Fig. 4(a)], however, was not observed in these data.

ACKNOWLEDGMENT

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