Magnetostatic mode excitations in EuS

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The first observation of many well-resolved magnetostatic (MS) modes in a Heisenberg ferromagnet is reported. The experiments were carried out on a thin, nearly square, platelet of EuS ($T_c = 16.7$ K) at X-band frequencies and at 4.2 K. The experimental values of the resonance fields H_r of the various MS modes (for static field $\vec{H}$ applied in the plane of the disk) are used to test the Damon-Eshbach theory modified to include the demagnetizing fields in a finite disk. Except for the (1,1) and (1,3) modes which appear as surface MS modes, all the other modes $(1,n_z)$ with $n_z = 5, 7, 9, \ldots, 41$ appear as volume MS modes. The experimental values of $H_r(1,n_z)$ agree (within $\pm 3\%$) with the values predicted by the modified Damon-Eshbach theory. However, for $n_z > 31$, there is increasing deviation between theory and experiment leading to an absorption edge whose field position is about 8% lower than the predicted value. This is not fully understood. The linewidth of the modes varies approximately as $(1/n_z)$. For $\vec{H}$, at an angle θ out of the plane of the disk, the observed resonance fields of the first several modes varies as $H_r(\theta) = H_r(0)/\cos\theta$ for $\theta < 75°$. An explanation for this result is given.

I. INTRODUCTION

Since the first observation of the magnetostatic (MS) modes in ferromagnetic resonance experiments in 1956,[1] considerable progress has been made in understanding these multiple resonances. The first detailed theory of the MS modes for spheroidal samples was given by Walker.[2] Damon and Eshbach[3] (DE) adapted this theory for a magnetic disc of infinite in-plane dimensions. By far, the most popular material for the investigation of the MS modes has been yttrium iron garnet (YIG), a ferrimagnet. The recent studies by Tittman[4] and Storey *et al.*[5] were also carried out on thin films of YIG. (A good source of references to the earlier studies is the review by Walker.[6]) Storey *et al.*[5] have attempted a quantitative comparison of the field position of the first several modes in YIG with the calculated values using the DE theory. Although they were able to identify several of the modes adjacent to the Kittel (uniform-precession) mode, there remained a discrepancy of about 5% between the calculated and observed resonance fields. In this connection it is noted that the DE theory was developed for an infinite disc. However, Story *et al.*[5] argued that if the dimensions b and c of the disc were much greater its thickness s, the DE calculations could still be valid, with the continuous values of the wave vectors replaced by quantization conditions, viz., $k_y = n_y \pi / b$ and $k_z = n_z \pi / c$, where n_y are n_z are integers defining the mode numbers. For their samples, b and c were both about two orders of magnitude larger than s, and the demagnetizing fields were negligible.

A second set of systems where the MS modes have been observed are the layer ferromagnets $CrBr_3$ (Ref. 7) and $(C_nH_{2n+1}NH_3)_2CuCl_4$ (Ref. 8) [i.e., C(n)Cu]. In these compounds the anisotropy plays a significant role which has been included in an extension[9] of the DE theory. The measurements of Reimann and Waldner in C(n)Cu (Ref. 8) are found to be in good agreement with the calculations[7] for both the volume MS modes (about 15 in number) and the surface MS modes (about 10 in number). In this comparison, the demagnetizing fields were also included in addition to the anisotropy. Recently, the MS modes have also been observed and analyzed in MnF_2, an antiferromagnet.[10]

To the best of our knowledge, the observations of the MS modes in a three-dimensional ferromagnet has not been reported so far. In this paper[11] we report the observation of up to 20 well-resolved MS modes in EuS, a good example of Heisenberg ferromagnet with $T_c = 16.7$ K.[12] Like the ferrimagnet YIG, EuS has the useful features of: (i) cubic structure; (ii) all magnetic ions being identical and in an S state; and (iii) sufficiently high resistivity so that microwave skin effects are negligible. However, there are several new features in this work on EuS, as compared to the reported studies on YIG.[4,5] First, in the sample used in the present work, the disc dimensions b and c are about one order of magnitude larger than the thickness s, compared to about two orders of magnitude difference in YIG. Second, the saturation magnetization M_s for EuS is about an order of magnitude larger than that for YIG. These two considerations make the demagnetization fields quite important in EuS, unlike the studies in YIG. We have included the demagnetization fields in our calculations, thereby providing an extension of the DE theory. In the present work, a quantitative test of the DE theory has been made by accurately measuring the resonance field for about 20 MS modes and comparing these with the calculated values. The change in the resonance field of these modes as the magnetic field is rotated out of the plane of the disc-shaped sample, has also been investigated. Although the DE theory does not consider the relaxation effects, we have also investigated the variation of the linewidth of the MS modes with the mode number.

II. EXPERIMENTAL DETAILS

The magnetic resonance measurements reported here were carried out on a standard X-band reflection-type ESR spectrometer. The klystron was stabilized to the sample cavity (employing a TE_{101} mode) so that only the field derivative of the absorption was observed. A rectangular disk of EuS was shaped from a larger single-crystal piece by cutting, followed by grinding on a fine Emery paper and cotton cloth in that order. The measured dimensions of the disk were: $b=2.26$ mm, $c=2.55-2.65$ mm, and the thickness $s=189-196$ μm was estimated from the weight (6.5 mg) of the sample. (Since the sample is quite brittle, we found it difficult to completely smooth the c direction—hence the variations in the c and s measurements. However, this is properly taken into account in the calculations.) The measurements were taken for the static field $\vec{H} \parallel b$ as well as $\vec{H} \parallel c$. The sample was placed on the yz plane of the cavity, near the position of the maximum rf field so that the magnetic field $\vec{H}$ could be rotated out of the plane of the disk, eventually to a position normal to the disk, while keeping $\vec{H}$ normal to the rf field. In the data presented here, the crystal orientations of EuS are left unspecified since its intrinsic anisotropy is only a few oersteds.[12] Magnetization versus field data for the magnetic field $\vec{H}$ applied along the b axis were taken with a Faraday balance at several temperatures below T_c. The experimental procedures for these measurements have been described elsewhere.[13] These measurements allowed us to provide a check on the demagnetization fields.

III. THEORETICAL CONSIDERATIONS

Before presenting the experimental data, an overview of the DE theory as it pertains to the calculation of the resonance field for the various MS modes is presented. The resonance fields for the various MS modes are solutions of the DE equation,[3] given by

$$(1+\eta^2)+2|(1+\eta^2)^{1/2}|\left(-\frac{1+\eta^2+\kappa}{1+\kappa}\right)^{1/2} \times(1+\kappa)\cot\left[|k_y|s\left(-\frac{1+\eta^2+\kappa}{1+\kappa}\right)^{1/2}\right] +(1+\kappa)^2\left(\frac{1+\eta^2+\kappa}{1+\kappa}\right)-\nu^2=0 \quad , \tag{1}$$

where

$$\eta=k_z/k_y \quad , \tag{2}$$

$$\kappa=\Omega_H/(\Omega_H^2-\Omega^2) \quad , \tag{3}$$

$$\nu=\Omega/(\Omega_H^2-\Omega^2) \quad , \tag{4}$$

$$\Omega_H=H_i/4\pi M_0 \quad , \tag{5}$$

and

$$\Omega=\omega/4\pi\gamma M_0 \quad . \tag{6}$$

In the above equations M_0 is the magnetization, γ is the gyromagnetic ratio, ω is the angular frequency of the microwave field, H_i is the internal static field along the z direction, and k_y and k_z are, respectively, the wave vectors along y and z directions. For a disk of infinite lateral dimensions and magnetized in its own plane, $H_i=H$(applied). But for the practical case of finite lateral dimensions, the demagnetization fields need to be considered in order to determine H_i. Towards this end, we reexamined the DE calculations, now by including the demagnetizing fields, namely, using $h_{ix}=h_x-4\pi N_x M_x$ and $h_{iy}=h_y-4\pi N_y M_y$ for the rf fields and $H_i=H-4\pi N_z M_0$ for the static field. Here N_x, N_y, and N_z are the demagnetizing factors ($N_x+N_y+N_z=1$) along x, y, and z directions, respectively. This analysis shows that Eq. (1) is still valid except that $H_i=H-4\pi N_z M_0$ is now the internal field. Following Storey *et al.*[5] and as noted in the Introduction, we take $k_y=n_y\pi/b$ and $k_z=n_z\pi/c$, with n_y and n_z being positive integers specifying various modes. Modes with even n_y and n_z are forbidden except the (0,0) mode with $n_y=0$ and $n_z=0$. However, strictly speaking this mode is possible only in an infinite plate, the DE geometry. In the DE theory, the (0,0) mode occurs at $\Omega=(\Omega_H^2+\Omega_H)^{1/2}$ which agrees with the expression for Kittel's uniform-precession mode for an infinite plate.[14] The allowed modes with nonzero values of n_y and n_z are classified as volume MS or surface MS modes according to whether $\eta >$ or $< \{\Omega^2/[\frac{1}{2}-(\frac{1}{4}+\Omega^2)^{1/2}]^2-1\}^{1/2}$. Thus whether a mode appears as a volume or a surface mode depends on the sample dimensions, the operating frequency ω and the direction of $\vec{H}$. Usually, the modes $(n_y,1)$ with $n_y=3, 5, 7, \ldots,$ appear as surface modes and the modes with $(1,n_z)$ with $n_z=3, 5, 7, \ldots,$ appear as volume modes. The absorption edge for volume modes in the DE theory is at $\Omega = \Omega_H$, which translated into the magnetic fields units with demagnetizing fields included, is given by

$$H_e = (\omega/\gamma) + 4\pi N_z M_0 \ . \tag{7}$$

The other limit is the theoretical position of the (0,0) mode noted earlier, which in magnetic field units and corrected for demagnetizing factors, is given by

$$H_0 = -2\pi M_0 + [(4\pi M_0)^2 + \omega^2/\gamma^2]^{1/2} + 4\pi N_z M_0 \ . \tag{8}$$

The volume MS modes appear between H_0 and H_e and the surface MS modes appear for $H < H_0$.

To calculate the resonance field H_r of a particular mode, one needs to solve Eq. (1) numerically. For this, we have essentially followed the procedure of Storey *et al.*[5] A computer program was written which allowed us to determine the zeros of the left-hand side of Eq. (1) for a particular n_y and n_z as the field is lowered from H_e. The field position where the first zero occurs is the H_r of that (n_y,n_z) mode. In this way positions of the various modes were calculated. These calculated values are compared with the observations in the following sections.

IV. EXPERIMENTAL RESULTS AND COMPARISON WITH THEORY

A. Experimental parameters

First we consider the values of the experimental parameters entering Eq. (1). The demagnetizing factors have been estimated from the work of Osborn[15] using the measured dimensions of the sample, yielding $N_y = 0.064-0.065$ and $N_z = 0.052-0.056$. As a check on these values, we measured the initial susceptibility (χ_y) for the field applied along the b direction of our sample at several temperatures below T_c. The results, magnetization versus field plot, are shown in Fig. 1. It is well known that for a ferromagnet, below T_c, $\chi_y = 1/4\pi N_y$. Results of Fig. 1 show that χ_y is indeed constant below T_c and its value yields $N_y = 0.062 \pm 0.002$, in close agreement with the calculated value noted above. (In Fig. 1, the absolute magnitude of the magnetization has an uncertainty of $\pm 3\%$, primarily from the uncertainty in determining the mass of this small sample.) At 4.2 K, the magnetization is nearly saturated above 1.5 kOe and its value is 1242 Oe. However, in our calculations we have used $M_0 = 1209$ Oe at 4.2 K as determined by Schwob *et al.*[12] because of the larger uncertainty in our measurements of M_0 as noted above. The two values, however, agree within experimental errors. We have also calculated the resonance fields

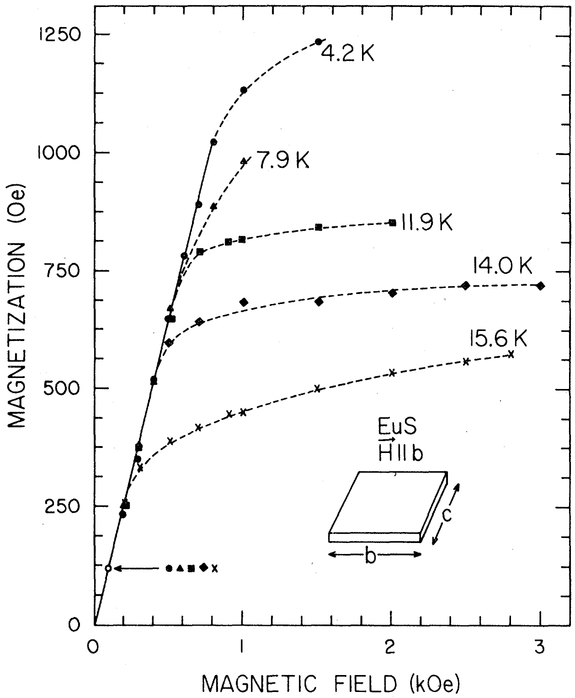

FIG. 1. Magnetization vs applied field at several temperatures for $\vec{H} \parallel b$. The dimensions of the nearly square platelet are: $b = 2.26$ mm, $c = 2.55-2.65$ mm, and thickness $= 189-196$ μm. A density of 5.748 for EuS is used to determine the thickness and to obtain the magnetization from the measured (per gram) values. The solid line gives the initial susceptibility and the dotted lines are drawn through the points for visual aid.

for $M_0 = 1150$ Oe, in order to demonstrate the effect of M_0 on H_r. To determine γ, we used $g = 2$, as determined by ESR experiments above T_c.

B. Identification of the modes

The observed spectrum of the MS modes for EuS at 4.2 K for $\vec{H} \parallel b$ is shown in Fig. 2, for the range of 400 Oe to 5 kOe. For aid in the identification of the modes, we followed the uniform-precession ESR line from above T_c to 4.2 K. This mode is labeled as the (1,1) mode in Fig. 2 [rather than the (0,0) mode] for reasons discussed in Sec. III. The other major lines are then the allowed modes such as (1,3), (1,5), etc. In this fashion modes up to (1,41) have been observed and identified. Several other modes of lower intensity and superposed on the main modes are also observed. However, these are difficult to label unambiguously, because their calculated H_r are not clearly resolved within the uncertainties. In any case, the identification of the major modes is sufficient to provide a clear check on the theory.

In the literature, the uniform-precession mode has

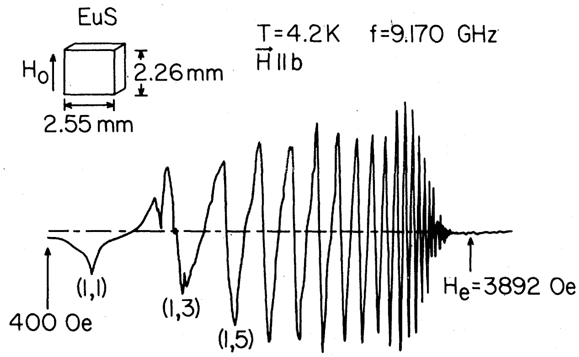

FIG. 2. A copy of the chart recording of the modes, with first few modes labeled. The edge position H_e and the resonance fields for various modes were determined from separate expanded sweeps.

been usually identified as the (1,1) mode of the DE theory, rather than the (0,0) mode. In our view, as noted earlier, this is not strictly correct, although in cases where the demagnetizing fields are unimportant, the calculated resonance fields for the two cases may nearly be the same. In our case, Eq. (8) for $\vec{H} \parallel b$ yields $H_0 = 1648$ Oe, whereas the calculated value of H_r for the (1,1) mode from the DE theory is 754 Oe (experimental $H_r = 771$ Oe). This difference between H_0 and $H_r(1,1)$ is primarily the demagnetizing field $4\pi N_z M_0$. On the other hand, if we use Kittel's equation for ferromagnetic resonance, for a finite plate[14] using the demagnetizing factors, we get $H_r = 926$ Oe. These comparisons suggest that the (1,1) mode is correctly labeled, but it is not strictly the uniform-precession mode.

C. Resonance fields for the modes

The resonance fields for various modes were carefully measured by slowly sweeping the field through each mode on an expanded field scale. The results for $\vec{H} \parallel b$, along with the calculated values, are given in Table I. The calculated values are for $b = 2.26$ mm, $c = 2.55$ mm, $s = 196$ μm, and for two values of M_0 (1209 and 1150 Oe). In Fig. 3, the results are compared graphically where the two solid lines are the calculated limits imposed by the nonuniformity of the sample dimensions. Considering that none of the parameters have been adjusted for the fit, the overall agreement in Fig. 3 (within $\pm 3\%$) is considered quite good. However we make note of the following

TABLE I. A comparison of the measured and calculated resonance fields H_r for various modes for the following parameters: $b = 2.26$ mm, $c = 2.55$ mm, $s = 196$ μm, $g = 2$, and $\omega/2\pi = 9.170$ GHz.

Mode (n_y, n_z)	Observed H_r (Oe)	Calculated H_r (Oe) $M_0 = 1209$ Oe	$M_0 = 1150$ Oe
(0,0)	...	926	956
(1,1)	771(s)	795	835
(1,3)	1539(s)	1707	1698
(1,5)	1954	1973	1978
(1,7)	2251	2226	2231
(1,9)	2493	2443	2448
(1,11)	2731	2636	2640
(1,13)	2899	2807	2809
(1,15)	3064	2959	2958
(1,17)	3187	3093	3088
(1,19)	3290	3210	3203
(1,21)	3383	3312	3302
(1,23)	3451	3402	3390
(1,25)	3513	3481	3466
(1,27)	3568	3550	3533
(1,29)	3613	3611	3591
(1,31)	3652	3665	3643
(1,33)	3685	3713	3689
(1,35)	3714	3755	3730
(1,37)	3735	3793	3766
(1,39)	3758	3827	3798
(1,41)	3777	3857	3827
H_e	3892	4246	4148

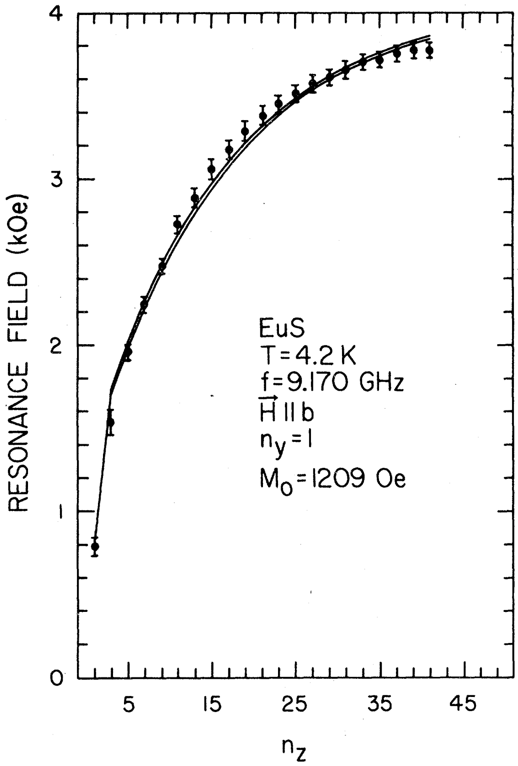

FIG. 3. Comparison of the observed (circles) and calculated (solid lines) resonance fields for various modes $(1,n_z)$ for $\vec{H} \parallel b$. The two solid lines bracket the calculated values, with allowance for the nonuniformity of the sample dimensions.

discrepancies: First, a discrepancy of about 10% in $H_r(1,3)$ and second, tendency of H_r for higher modes to be lower than the calculated values, resulting in a discrepancy in H_e.

Regarding the (1,3) mode, first we note that using the condition on η discussed in Sec. III, the (1,3) [as well as the (1,1) mode] are surface modes, whereas all the other observed modes are volume modes. The transition given by H_0 [Eq. (8)] occurs at 1648, close to H_r for the (1,3) mode. Also, from Fig. 2, it is evident that there are several other modes superposed on the (1,3) mode, near the boundary between the volume and the surface modes. From intensity considerations,[5] we have identified the line with the largest intensity as the (1,3) mode. However, the interference by the other lines makes $H_r(1,3)$ somewhat uncertain. Calculations show that the modes (3,3), (3,5), and (5,5) have H_r of 1680, 1767, and 1750 Oe, respectively, all in the neighborhood of $H_r(1,3)$ and H_0. A more positive identification of these lines is not possible, because of the lack of resolution. One may also conjecture that these lines are in some way related to the transition between the volume and the surface modes, since they appear close to the field position of this boundary. In any case we believe that the interference by these lines is the major source of the discrepancy between theory and experiment for $H_r(1,3)$. For the (1,1) mode, the unusual shape (Fig. 2) may be related to the fact that the magnetization is not saturated at $H_r(1,1)$ (see Fig. 1). The slight discrepancy between the theory and experiment at higher mode numbers, as well as in the position of the band edge H_e (Table I), may be related to the onset of the breakdown of the magnetostatic conditions for higher wave numbers; however, we cannot provide a more quantitative explanation. Studies by Storey *et al.*[5] were limited to only the first few modes, although Tittman[4] in his studies on YIG claimed to have observed the position of H_e within about 2% of the theoretical prediction, whereas our discrepancy is about 8%. The major difference between the two cases is that in EuS, corrections for the demagnetizing fields are necessary, since the magnetization is about an order of magnitude larger. This would tend to accentuate the discrepancy. Another noteworthy observation is the effect of the change in the magnetization on H_r of various modes. In Table I, we have given calculated H_r for $M_0 = 1209$ and 1150 Oe for the various modes. Up to mode (1,15) the decrease in magnetization increased H_r, whereas for higher modes, the effect is just the opposite. The only exception to this rule is

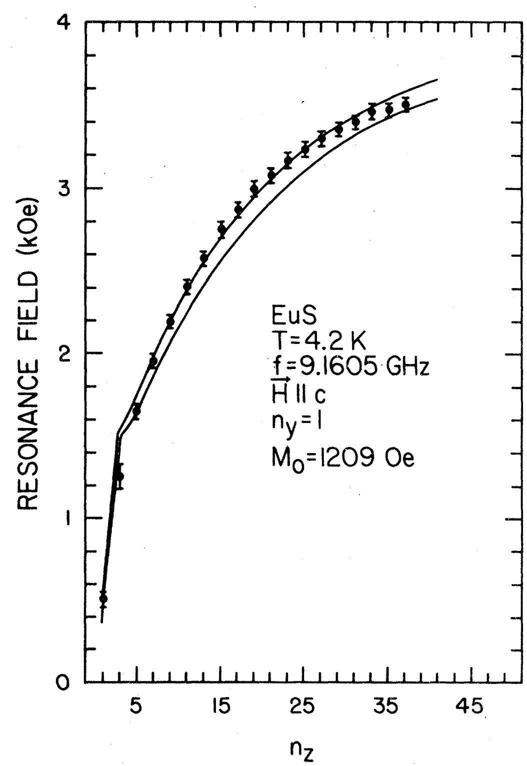

FIG. 4. Comparison similar to Fig. 3, for $\vec{H} \parallel c$.

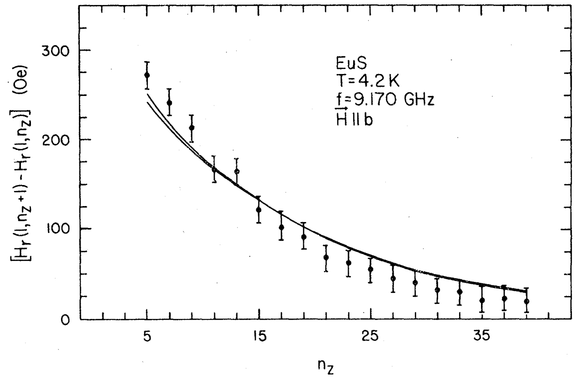

FIG. 5. Magnetic field separation between the consecutive modes vs mode number for $\vec{H} \parallel b$. The two solid lines bracket the theoretical predictions.

the mode (1,3). Further theoretical work is needed to explain these observations.

For $\vec{H} \parallel c$, observations similar to the ones noted above, for $\vec{H} \parallel b$, have been made. In Fig. 4, the observed H_r for various modes is compared with the calculated values allowing for the variation in the c axis. The two solid lines are for $c = 2.55$ and 2.65 mm, respectively, the measured minimum and maximum values of c. The agreement is good within experimental errors for modes higher than the (1,3) mode. There is again considerable discrepancy for the (1,3) mode. Overall, the observations for b and c directions are similar. This is not surprising, since $b \simeq c$.

Another critical check on the theory can be made by comparing the difference in H_r between the consecutive modes. This comparison is shown in Figs. 5 and 6 for $\vec{H} \parallel b$ and $\vec{H} \parallel c$, respectively, for modes higher than the (1,3) mode. The two solid lines are the calculated values, allowing for the nonuniformity in the crystal dimensions. Within the experimental uncertainties, the agreement is quite good, although the calculated values tend to be a bit higher than the average experimental values for the higher mode numbers. This is mostly due to the discrepancy already noted in Figs. 3 and 4 for the higher mode numbers.

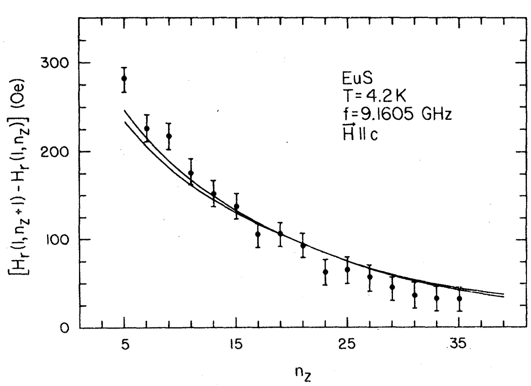

FIG. 6. Comparison similar to Fig. 5, for $\vec{H} \parallel c$.

D. Angular variation of the resonance field

For $\vec{H} \perp$ platelet, the calculated H_0 [Eq. (8)] and H_e [Eq. (7)] with $M_0 = 1209$ Oe, are 14 and 16.7 kOe, respectively. These values are well outside the range of our magnet. Therefore, it is no surprise that we did not observe the MS modes for this configuration. We were, however, able to follow several of the modes as the magnetic field is rotated out of the plane of the platelet, and the results are shown in Fig. 7. The solid lines are the curves $H_r(\theta = 0)/\cos\theta$, where θ is the angle between the applied field $\vec{H}$ and the plane of the platelet. The good agreement of Fig. 7 suggests that the magnetization is still within the plane of the platelet for these values of $\vec{H}$ and θ, and, consequently, the component of $\vec{H}$ parallel to the platelet ($= \vec{H}\cos\theta$) is able to excite the parallel MS modes. Since $4\pi N_z M_0 \simeq 13.4$ kOe for $\vec{H} \perp$ platelet, a value considerably larger than the resonance fields observed in Fig. 7, the above observation is quite understandable.

E. Linewidths of the MS modes

We have carefully measured the linewidth ΔH (peak-to-peak separation in the absorption derivative)

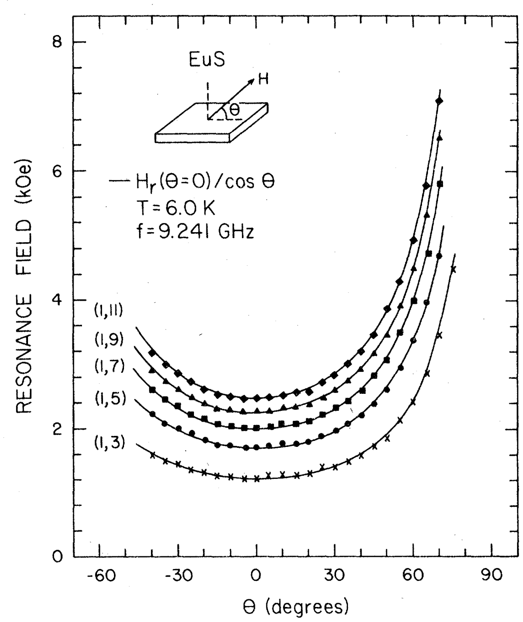

FIG. 7. Angular variation of the resonance field H_r for several modes as the magnetic field is rotated out of the plane of the platelet.

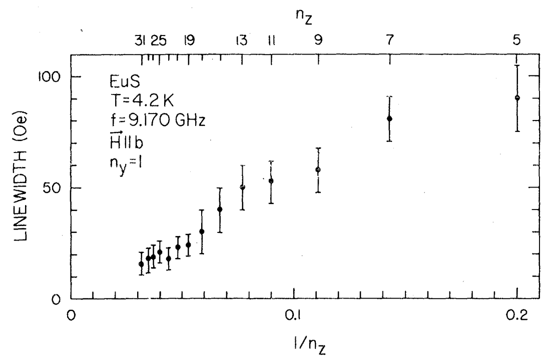

FIG. 8. Linewidth of the modes vs inverse mode number.

of the various MS modes by a slow sweep of the field through each mode on an expanded scale. The results, in the form of ΔH vs n_z, for $\vec{H} \parallel b$ and at 4.2 K, are shown in Fig. 8. It is seen that ΔH varies approximately as $(1/n_z)$. In many of the earlier studies[3–5,7,8] no attention has been paid to the linewidth of the modes. In the case of C(n)Cu, we have crudely estimated the linewidth of the volume MS modes from the experimental trace given in the paper by Reimann and Waldner.[8] It is found that ΔH does decrease with increase of n_z, somewhat similar to the situation in Fig. 8.

In a discussion of an earlier theory by Clogston *et al.*,[15] White[16] argues that the linewidth of the MS modes should increase as their distance from H_e increases if the linewidth is due to random magnetic inhomogeneities. (Such inhomogeneities could result, for example, if the sample is not highly polished—the situation in our case.) Our observations are in qualitative agreement with this prediction. However, Nemarich[17] in a later work, has demonstrated that the magnitudes of the linewidths and even their functional dependence on the mode number are affected if the sample is highly polished. So the situation is far from certain. It should be noted that studies by White[11] and Nemarich[17] were carried out on YIG and Mn-Zn ferrite samples in the form of *spheres*, whereas our studies are in a thin platelet. The magnitudes and even their functional dependence on n_z may be quite different in the two cases, because of the influence of the shape effect. Thus, whether the functional dependence of $\Delta H \sim (1/n_z)$ observed in EuS (Fig. 8) is a general phenomenon characteristic of thin platelet needs to be verified in other ferromagnets and investigated theoretically.

V. CONCLUDING REMARKS

In this paper, the first observation of well-resolved MS modes in a thin platelet of a Heisenberg ferromagnet (EuS) has been presented and the results are compared with the resonance fields predicted by the Damon-Eshbach theory, modified for a finite platelet and to include the demagnetization fields. Overall, good agreement is obtained between the observed and calculated values. However, there are two areas where need for further work is indicated by this study. First, the trend towards increasing deviation between the observed and calculated values for higher modes, leading to about 8% discrepancy in the position of the absorption edge, needs to be better understood, although we suggest that this may be due to breakdown of the magnetostatic conditions for higher wave numbers. Second, further theoretical and experimental work is needed to understand the variation of the linewidth with the mode number in a thin platelet. We hope that this paper will stimulate further studies along these lines.

ACKNOWLEDGMENTS

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