

# Frequency and temperature dependence of the ferromagnetic resonance linewidth in single crystal platelets and pulsed laser deposited films of barium ferrite

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The half power ferromagnetic resonance (FMR) linewidth  $\Delta H$  has been measured from 8 to 300 K for a nominal frequency of 61 GHz and from 50 to 75 GHz at room temperature for normally magnetized single crystal platelets and pulsed laser deposited (PLD) films of barium ferrite. The platelet linewidth versus temperature data show a peak value of 27 Oe at 25–30 K, a dip to 21 Oe at 75 K, structure related to line merging at 130–160 K, and a region of slow increase at 240–295 K. The 240–300 K data extrapolate to a 0 K linewidth of about 8 Oe. The corresponding film data show a peak value of 240 Oe at 20 K and a gradual decrease at higher temperatures. Both the platelet and film linewidths show a weak frequency dependence at about 0.3 Oe/GHz at room temperature (RT). The film data also show several linewidth spikes due to interference effects between the FMR response and dielectric resonances in the substrate. The RT linewidth zero frequency intercepts for the platelet and the film were 9 and 30 Oe, respectively. The frequency dependence of the platelet linewidth was also examined at 230–295 K. The frequency responses and intercepts for these data were affected by the line merging at lower temperatures and showed high temperature limit values of 0.35 Oe/GHz and 9.5 Oe, respectively. The linewidth frequency response in both cases is attributed to conductivity. The linewidth versus temperature peaks are attributed to impurities. The linewidth dip at 75 K for the platelet results from the drop in  $\Delta H$  on the tail of the temperature peak and an increase associated with iron ion hopping motion. © 2002 American Institute of Physics. [DOI: 10.1063/1.1450057]

## I. INTRODUCTION

Barium ferrite is a promising material for millimeter wave applications because of the large uniaxial magnetocrystalline anisotropy. The large uniaxial anisotropy allows these materials to be used at millimeter wave frequencies without the need for large static applied fields. The main problem with these materials is the large microwave loss. Reference 1 provides an indication of the current situation and previous works on the various loss processes which apply to M-type barium ferrite (BaM). Recent advances in pulsed laser deposited (PLD) barium ferrite thin film technology make the microwave loss issue even more important. The PLD technique is compatible with monolithic microwave integrated circuits technology. Integrated barium ferrite film microwave devices offer the possibility of unique applications in radar and high frequency communications.

The two main control parameters for the investigation of the ferromagnetic resonance (FMR) linewidth and relaxation mechanisms are frequency and temperature. Previous data on linewidth versus frequency and temperature for single crystal barium ferrite provide no consistent picture of the phenom-

enology or the physical relaxation mechanisms. Four groups report a linear frequency dependence for the linewidth.<sup>1–5</sup> In Ref. 2, Röschmann *et al.* measured linewidth versus frequency for a wide variety of barium ferrite compositions. They found a linear variation in linewidth with frequency with frequency responses of 0.4 Oe/GHz and higher, along with extrapolated zero frequency intercepts of 7–8 Oe or higher. In Ref. 5, Silber and Wilber measured the linewidth from 52 to 64 GHz at 60–300 K and extracted a frequency dependence of 0.2 Oe/GHz at 300 K and 0.04 Oe/GHz at 60 K. This response was attributed to the Kasuya–LeCraw (KLC) processes.<sup>6</sup> Reference 3 gives a linewidth change with a frequency of 0.5 Oe/GHz for pure BaM and 0.3 Oe/GHz for Mn doped BaM. The difference was attributed to conductivity effects. On the other hand, El Rayess *et al.*<sup>7</sup> report field swept linewidths of 21–32 Oe over the frequency range 48–105 GHz, with no clear correlation between linewidth and frequency.

Linewidth versus temperature data by Silber and co-workers<sup>5,8</sup> for BaM indicate a linear increase which is attributed to KLC processes and a combination of phonon-magnon and two magnon processes. For Mn-substituted BaM, Silber and co-workers report two different results, a linewidth peak<sup>5</sup> at about 130 K and a temperature independent linewidth.<sup>8</sup>

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There has been a substantial amount of work on the fabrication of PLD barium ferrite films. See Ref. 9 and other references therein. These works also include some limited results on microwave properties. One report by Carosella *et al.*<sup>10</sup> gives linewidth versus frequency. The data show a decrease in the half power linewidth from about 150 Oe at 86 GHz to 90 Oe at 50 GHz. There have been no reports on the linewidth versus temperature for PLD barium ferrite films.

The purpose of this work was to perform precise linewidth measurements from 50 to 75 GHz and 8 to 300 K on high quality single crystal platelets and PLD thin films of BaM. The linewidth temperature dependences for the platelet and the film are completely different. The platelets show a slowly decreasing background linewidth with decreasing temperature which is decorated with several peaks. The films show a strong linewidth increase with decreasing temperature to a peak value of about 240 Oe at 20 K. This temperature peak response is superimposed on a background which scales with the magnetization and is attributed to two magnon processes.

The linewidth versus frequency data for the platelet at high temperature, away from the temperature peak range, show a linear response of about 0.35 Oe/GHz and a zero frequency intercept of about 9 Oe. The linear frequency dependence is due to conductivity. The extrapolated zero frequency linewidth of 9 Oe is taken as intrinsic. This value matches the extrapolation of the decreasing background linewidth versus temperature to 0 K. The best PLD films show almost the same frequency response but a much higher zero frequency intercept of 30 Oe.

Section II describes the materials and measurement procedures. The linewidth measurement technique is particularly critical because of the sensitivity of the response to the exact frequency operating point. Section III presents the experimental results and analysis. In addition to basic results on linewidth versus temperature and frequency in Secs. III A and III B, Sec. III B also gives a complete analysis of the combined frequency and high temperature data for the platelet. This analysis leads to a clear and self-consistent determination of the intrinsic platelet linewidth. Section IV gives key conclusions.

## II. MATERIALS AND PROCEDURE

The single crystal barium ferrite materials were prepared by standard flux-melt growth methods at Purdue University.<sup>11</sup> The actual sample for the FMR measurements was a rectangular  $250\ \mu\text{m} \times 800\ \mu\text{m}$ ,  $15\ \mu\text{m}$  thick virgin cleaved platelet with the hexagonal  $c$  axis perpendicular to the surface. The use of an as-cleaved platelet avoided the problems associated with the usual pit and crack defects produced by polishing. This sample was selected from a collection of about 40 cleaved or polished platelets, based on surface quality and a symmetric and narrow FMR absorption. The small thickness ensured a highly uniform internal magnetic field. This factor is important for a narrow and intrinsic linewidth.

The PLD films were deposited on (0001) oriented  $\text{Al}_2\text{O}_3$  substrates. The target was conventionally sintered  $\text{BaFe}_{12}\text{O}_{19}$

polycrystalline material from Praxair Surface Technologies. The pulsed KrF 248 nm wavelength excimer laser was operated at an incident energy density of  $2.5\ \text{J}/\text{cm}^2$  per pulse at the target and a 10 Hz repetition rate. The depositions were made at an ambient oxygen partial pressure of 300 mTorr with a heated substrate at  $900\ ^\circ\text{C}$ . The nominal film thickness was  $0.3\ \mu\text{m}$ . After deposition, the films were cooled at  $5\ ^\circ\text{C}/\text{min}$ , removed from the chamber, and then annealed in flowing oxygen at  $1100\ ^\circ\text{C}$  for 20 min. The heating and cooling rate during annealing was maintained at  $\pm 20\ ^\circ\text{C}/\text{min}$ , respectively.

The FMR absorption versus field derivative profiles were obtained by a standard field-swept shorted waveguide technique with ac field modulation and lock-in detection. The cw microwave source consisted of an HP8341B synthesized sweeper, an HP8349B microwave amplifier, and an HP83557A frequency multiplier. The microwave power was always below 5 dBm. Data were obtained from 50 to 75 GHz. The platelet or film sample was glued at the center of the inside face of a WR-15 waveguide shorted section which was mounted on the cold finger of a Janis closed cycle refrigerator system. The cryostat system was positioned with the sample in the middle of the gap of a 12 in. Varian magnet with the field perpendicular to the plane of the film or platelet. A Lake Shore DRC82C temperature controller and cold finger heater were used to obtain temperatures from 8 to 300 K. The temperature stability was better than 0.5 K. A MicroNow 8320A field controller with a resolution of 1 Oe was used to step the field.

For the platelet data, all measured lines were symmetric and undistorted. The modulation field was kept well below the width of the FMR peak. For each peak-to-peak derivative linewidth determination, the actual derivative absorption curve was field scanned at least three times and averaged. These derivative linewidth values were then multiplied by  $\sqrt{3}$  to obtain the half power linewidth results given below. The  $\sqrt{3}$  factor is valid for a Lorentzian absorption profile. These derived linewidths were checked for selected profiles by direct numerical integration. The overall error in the linewidth determinations was less than 1 Oe.

The FMR profiles for the PLD films were more complicated. For the narrow linewidth films of interest here, the derivative profiles for the main resonance were distorted by the presence of higher order spin wave modes on the low field side. For this reason, linewidths were obtained from field spacing of the derivative zero crossing and the high field extrema.

For both the linewidth versus frequency and the linewidth versus temperature measurements, it was necessary to exercise care in setting the exact frequency for the linewidth determination. The interference between the microwave signal reflected from the waveguide short and the microwave leakage signal from the source caused a periodic variation in the output response with frequency with a repetition period of about 100–200 MHz. In order to obtain the correct linewidth, one must operate only at the local maxima for this

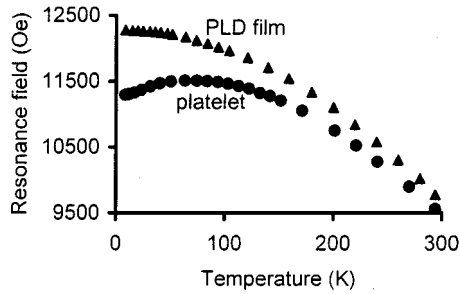


FIG. 1. Ferromagnetic resonance (FMR) field vs temperature for the BaM platelet and the pulse laser deposited film. The operating point FMR frequencies for the platelet and the film were 60.95 and 61.25 GHz, respectively.

response. For this reason, the frequency was always fine tuned to obtain the minimum measured linewidth.

### III. RESULTS AND DISCUSSION

#### A. Linewidth temperature dependence

Figure 1 shows data on the resonance field versus temperature for a nominal frequency of 61 GHz. The platelet data show a small maximum at about 75 K. The film data show a smooth monotonic decrease in the resonance field with temperature. These different responses are related to the temperature dependences of saturation induction  $4\pi M_s$  and uniaxial anisotropy field  $H_A$ .

The uniform mode FMR field  $H_{\text{res}}$  for a BaM film or thin platelet is given by

$$H_{\text{res}} = 2\pi f / |\gamma| - H_A + 4\pi M_s, \quad (1)$$

where  $f$  is the frequency and  $\gamma$  is the gyromagnetic ratio. For ferrites,  $|\gamma|/2\pi$  is close to 2.8 GHz/kOe. If  $4\pi M_s$  is a smoothly decreasing function of temperature,<sup>12</sup> then the different  $H_{\text{res}}(T)$  profiles in Fig. 1 may be attributed to different  $H_A(T)$  temperature dependences for the platelet and the film. From the small increase in  $H_A$  with  $T$  at low temperature for BaM single crystals from Ref. 12, one would not expect the maximum in  $H_{\text{res}}(T)$  shown in Fig. 1. Such a maximum is

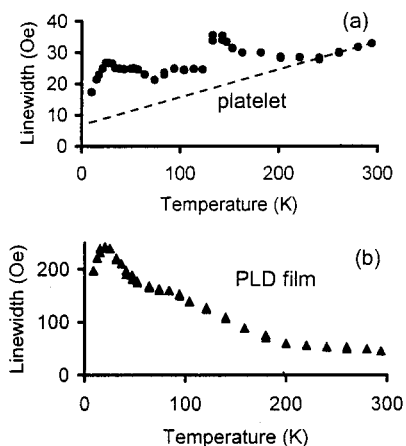


FIG. 2. Half power ferromagnetic resonance linewidth vs temperature for (a) the BaM platelet and (b) the pulse laser deposited film. The nominal frequency was 61 GHz.

consistent, however, with similar data on planar ZnY hexagonal ferrite materials by Dorsey *et al.*<sup>13</sup> and unpublished data from Petrov.<sup>14</sup> For the PLD film, the smooth decreasing  $H_{\text{res}}(T)$  profile indicates a somewhat different  $H_A(T)$  response. Here, one may have a different intrinsic anisotropy as well as a temperature dependent strain contribution which yield the result shown in Fig. 1.

Figure 2 shows data on the half power linewidth versus temperature for (a) the platelet and (b) the PLD film. The nominal frequency was 61 GHz. The platelet linewidth versus temperature profile in graph (a) shows four characteristic regions. (1) There is a low temperature maximum around 25–30 K. (2) There is a small dip at 75 K. (3) There is a discontinuous jump at 130 K. (4) Above about 240 K, the linewidth shows a gradual increase with temperature.

The low temperature maximum is attributed to the well known valence exchange or impurity temperature-peak process.<sup>6</sup> The 4 Oe dip at 75 K, while small, is well above experimental error and is completely reproducible. This dip is attributed to competition between the tail of the temperature peak response noted above and a linewidth contribution due to the iron ion hopping mechanism which comes in at about 80 K.<sup>15</sup> While the iron hopping mechanism was initially proposed by Tsantes and Silber in Ref. 15 and discussed by several authors.<sup>16,17</sup> Up to now, there has been no direct experimental evidence for this process.

The discontinuous jump in linewidth at 130 K and the gradual drop from 130 to 180 K is the result of line merging. A detailed examination of the actual FMR profiles shows a small secondary peak which appears above the main FMR line at about 50–60 K. With increasing temperature, this peak shifts toward the main resonance and produces an abrupt increase in the measured peak-to-peak derivative linewidth at about 130 K. As the temperature is increased further, this peak merges into the main FMR line.

The dashed line in Fig. 2(a) shows an extrapolation of the linear temperature response above 240 K down to 0 K. This extrapolated 0 K linewidth is about 8 Oe. The linewidth versus frequency data presented in Sec. III B will give zero frequency extrapolations which are close to this same value. These results, considered together, indicate that this common extrapolated value has a single origin. This may correspond to an intrinsic linewidth for the platelet.

The PLD film linewidth versus temperature profile in graph (b) of Fig. 2 has a different character from the linewidth profile for the platelet. The film shows a temperature peak at 20 K which appears to match the 25–30 K peak in graph (a). At higher temperatures, however, the film shows a linewidth which simply decreases monotonically with increasing temperature. The small shoulder at about 80 K may correspond to a second broad temperature peak.

The gradual decrease in linewidth above 200 K is most likely due to a combination of two magnon scattering and conductivity. The decrease scales approximately with the drop in magnetization with temperature, and this is the usual signature of two magnon scattering from pores, pits, or defects in a film.<sup>18</sup> It is important to note that two magnon processes are allowed here, even though the film is magnetized perpendicular to the surface. For very thin films, as is

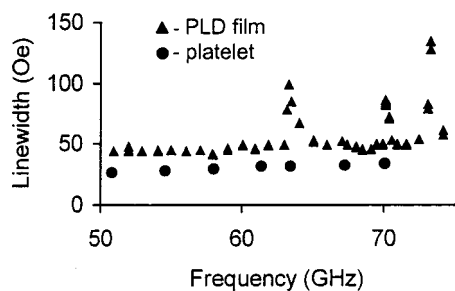


FIG. 3. Room temperature half power ferromagnetic resonance linewidth vs frequency for the BaM platelet and the pulse laser deposited film.

the case here, the main branch dipole exchange spin wave dispersion curve of frequency versus wave number has a very small initial slope. Because of the nonzero linewidth of the FMR mode, scattering is allowed to other near degenerate modes on this same branch. The role of conductivity will be considered below, in connection with the linewidth versus frequency results.

### B. Linewidth frequency dependence

Figure 3 shows room temperature data on linewidth versus frequency. The platelet linewidth had a frequency response of 0.36 Oe/GHz and a zero frequency extrapolation value of 8.6 Oe. Apart from the three spikes at 63, 70, and 73 GHz, the PLD film linewidth also shows an increase with frequency, with a response close to 0.3 Oe/GHz. The three spikes in linewidth for the PLD film are a few hundreds MHz in width. These spikes are attributed to dielectric resonances in the  $\text{Al}_2\text{O}_3$  substrate and interference effects for the FMR response in the shorted waveguide.

It is noteworthy that both sets of data in Fig. 3 give a linewidth frequency response of 0.3–0.35 Oe/GHz. This result is consistent with the 0.5 Oe/GHz response for pure barium ferrite found by Karim *et al.* in Refs. 1 and 3 from room temperature measurements. Karim *et al.* also found that 0.1 Mn atom per f.u. (1) reduced the conductivity of their particular series of samples, (2) gave a linewidth increase, and (3) caused the frequency response to decrease by about a factor of 2. The extrapolated linewidth at zero frequency was close to zero for the pure BaM sample and about 20 Oe for the Mn doped sample. Additional data on the high field effective (HFE) linewidth for the Mn doped sample gave values which were significantly smaller than the FMR linewidth, but which also had a slightly higher change with frequency. The zero frequency extrapolation of the HFE linewidth was about 4 Oe. These responses were taken as an indication of (1) an “intrinsic” origin of the linewidth change with frequency in the 0.3–0.4 Oe/GHz and (2) an inhomogeneity origin for residual linewidth at zero frequency.

The present data, in combination with further numerical considerations, provide a perspective on the physical origin of these linewidths. First, the observed 0.3–0.5 Oe/GHz linewidth frequency response from Refs. 1–3 and the present data give an overall linewidth change of tens of Oe over the

50–90 GHz frequency range. The magnitude of this change is much too big to be explained by intrinsic processes such as KLC relaxation, for example. An “intrinsic” KLC process would contribute only about 0.05 Oe/GHz to the linewidth frequency response.<sup>16</sup> Second, a simple calculation<sup>19</sup> for a 200  $\mu\text{m}$  diam BaM sphere with a V-band conductivity of 0.1  $\Omega^{-1}\text{cm}^{-1}$  (Ref. 20) gives a linewidth which changes from 30 Oe at 50 GHz to 42 Oe at 70 GHz, for an estimated frequency response of 0.6 Oe/GHz. This estimate is consistent with the present and the previous data. Note that the Mn substitutions in Ref. 3 would serve to reduce the conductivity somewhat, and this would reduce the linewidth frequency response coefficient, just as observed experimentally.

The above conclusion is also consistent with the decrease in linewidth with temperature over the temperature range from 230 to 295 K shown in Fig. 2(a). A conductivity linewidth scales with both frequency and conductivity. Insofar as ferrites generally show a semiconductor-like decrease in conductivity with temperature, this linewidth decrease may well be due to conductivity as well. The fact that the linewidth data extrapolated to zero frequency and the dashed line extrapolation to zero temperature in Fig. 2(a) give nearly the same 9 Oe value gives further support to this conclusion.

The BaM linewidth analysis in Ref. 5 by Silber and Wilber was in terms of a operational linewidth of the form  $\Delta H = AfT + BT + C$ , where  $f$  is the frequency and  $T$  is the temperature. These authors suggested that the  $A$  coefficient originates in KLC processes, the  $B$  coefficient derives from two magnon and phonon magnon processes, and the  $C$  coefficient is related to two magnon processes at  $T=0$ . The experimentally determined value of  $C$  was 10.8 Oe, very close to the 9 Oe value indicated above. The experimentally determined values of  $A$  and  $B$  were  $7.3 \times 10^{-4}$  Oe/K GHz and 0.014 Oe/K, respectively. While, these numerical values are consistent with the present data, the specific mechanisms suggested in Ref. 5 do not appear to be correct. The data in Ref. 5 can be explained exclusively in terms of conductivity losses, in the same way as described above. Reference 5 also reports results for Mn substituted BaM materials, with a reduction in the experimentally determined  $A$  coefficient. This reduction is consistent with a reduction in the conductivity, the same as noted above for the Ref. 3 data.

Further measurements of the platelet linewidth versus frequency, similar to the data in Fig. 3, were made over the temperature range from 230 to 295 K. This high temperature range corresponds to the linear part of the temperature response shown in Fig. 2(a) unaffected by other complications. For lower temperatures, the linewidth increases due to the line merging effect discussed in Sec. III A. These data were then analyzed in terms of a linewidth response equation of the form

$$\Delta H = C(T)f + D(T), \quad (2)$$

where  $C(T)$  represents the slope of the  $\Delta H$  versus  $f$  response and  $D(T)$  gives the zero frequency intercept. At the low end of the 230–295 K temperature range, these coefficients are affected by the line merging effect. The extrapolation of these  $C(T)$  and  $D(T)$  coefficients to high temperature give the limit values in the absence of line merging.

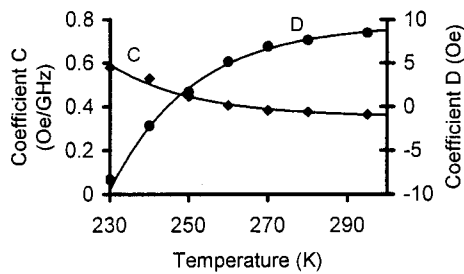


FIG. 4. The temperature dependences of coefficients  $C$  and  $D$  and their exponential fitting curves.

Figure 4 shows the results of the measurements and analysis. The solid points show the experimental  $C$  and  $D$  values obtained from fits of the actual linewidth data to Eq. (2) as a function of temperature. The solid curves were obtained from exponential fits to the data points and are shown as a guide to the eye only. It is clear from the data in Fig. 2 that the  $C(T)$  and  $D(T)$  values at the lower temperatures are affected by the line merging and other processes which cause the linewidth to increase at low temperature. More importantly, the trend of the  $C(T)$  and  $D(T)$  determinations for temperatures increasing from about 250 K give values of these parameters which apply to the conductivity and the intrinsic losses, respectively. The limit  $C$  and  $D$  values from the exponential fits are 0.35 Oe/GHz and 9.5 Oe, respectively.

The conductivity origin of  $C(T)$  suggests a form  $C(T) \propto M_s(T)\sigma(T)$ , where  $M_s$  is the magnetization and  $\sigma$  is the conductivity.<sup>19</sup> The change in the magnetization over the temperature range 270–295 K is estimated to be a few percent at most. The semiconductor-like conductivity should be an increasing function of temperature. The data suggest that  $C(T)$  is a slightly decreasing function of temperature. This could be a residual effect of the line merging. Further data at higher temperature would be needed to clarify these processes.

Turn now to the  $D$  parameter. Recall that the zero temperature extrapolated linewidth from the dashed line in Fig. 2 was 8 Oe. The fact that the zero frequency extrapolated linewidth at high temperature, i.e.,  $D \sim 9.5$  Oe, and the zero temperature extrapolated linewidth are about the same provide additional support for the conductivity origin for the frequency and temperature dependent part of the linewidth. The actual origin of this conductivity free “intrinsic” linewidth is unknown.

The film data in Fig. 3 give a 0.3 Oe/GHz increase in linewidth with frequency, and this is consistent with conductivity losses in the same way as discussed above. Apart from the temperature peaks at 20 and 80 K, the extrapolated film linewidth increases from 45 Oe at 300 K to about 80 Oe at 10 K can be ascribed to a combination of conductivity losses and two magnon processes. Both processes scale with magnetization.<sup>6,19</sup> However, the conductivity losses are estimated to give a change from 300 to 10 K of +7 Oe if the conductivity is temperature independent. Because of the semiconductorlike decrease in conductivity with decreasing temperature for ferrites, it is likely that this change is actu-

ally less, or even negative. Most of the increase in the extrapolated film linewidth from 300 to 10 K, therefore, must be attributed to two magnon processes. This is also consistent with previous room temperature FMR data on PLD spinel and garnet films.<sup>21</sup>

#### IV. CONCLUSION

The FMR linewidth versus temperature and frequency has been measured for normally magnetized single crystal platelet and PLD film samples of BaM at 50–75 GHz and from 8 to 300 K. The data reveal three important results. First, both the platelets and the film have frequency dependent linewidths with a response that is consistent with a conductivity mechanism.

Second, the temperature data for the platelets give a small but distinct dip in the linewidth at around 75 K. This dip provides experimental verification of an iron ion hopping motion contribution to the linewidth.

Third, the PLD film linewidth data versus temperature show a gradual increase which scales with the magnetization and provides an indication of two magnon processes. This result shows that two magnon processes can be realized even for perpendicular magnetized films, a configuration for which two magnon processes are usually excluded. Two magnon scattering is allowed here because of the nonzero linewidth of the FMR mode in combination with the nearly flat character for the lowest order dispersion branch in very thin films.

Additional results include (1) evidence for temperature peak processes in both the platelets and the films and (2) high temperature linewidth extrapolations to  $T=0$  for the platelet which match the zero frequency extrapolations at high temperature, indicating an “intrinsic” BaM linewidth of about 8–9 Oe. The most important remaining question concerns the origin of this “intrinsic” linewidth.

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