Low power nonlinear effects in the ferromagnetic resonance of Zn_2Y and MnZnY hexagonal ferrites

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The room temperature ferromagnetic resonance (FMR) absorption at 8.9–11.2 GHz in two planar hexagonal ferrites, Zn_2Y and MnZnY, has been found to exhibit strong nonlinear behavior at low power levels corresponding to microwave field amplitudes on the order of 10 mOe. For somewhat larger amplitudes, 20–50 mOe, the asymmetric resonance curves show pronounced foldover effects. With increasing static field, the absorption increases gradually to a steep cutoff at a field several hundred Oe above the position of the low power FMR. With decreasing field, a sharp spike occurs coincident with the high field tail of the low power FMR line, followed by a gradual decrease which tracks the upsweep absorption. The detailed shapes of these curves depend strongly on sweep rate. Steady-state absorption curves at the highest powers employed, with 350 mW incident power and a microwave field amplitude of 58 mOe, result only for very slow sweep rates of 20 Oe/min or less.

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INTRODUCTION

Nonlinear microwave phenomena have been reported in single crystal hexagonal ferrites from the first studies of such materials. Dixon[1] observed saturation of the ferromagnetic resonance (FMR) in planar $Ba_2Zn_2Fe_{12}O_{22}$ (Zn_2Y) spheres at microwave field amplitudes of 100 mOe–2 Oe for materials with FMR linewidths of 100–200 Oe. The threshold was found to increase sharply with frequency, from 0.16 Oe at 8.6 GHz to 1.8 Oe at 16.6 GHz, and then level off at higher frequencies. This behavior was attributed to the observation that, for spheres, first-order low threshold spin-wave instability processes are allowed at the lower frequencies due to the availability of spin waves at half the pump frequency, while such processes are not possible at the higher frequencies.[2]

Additional resonance saturation data for Mn–ZnY were reported by Savage *et al.*[3] For the narrowest linewidth samples, 3.8 Oe at 9 GHz and 4.9 Oe at 17.3 GHz, the onset for saturation at 17.3 GHz occurred at 100 mOe microwave field amplitude. Spin-wave instability data were also reported for Mn–ZnY under parallel pumping at 17.2 GHz, with the microwave field parallel to the static magnetic field.[4] The minimum critical field was below 500 mOe (extrapolated from the data of Ref. 4). More recently, nonlinear effects at very low microwave powers have also been reported for uniaxial $Ba_3Zn_2Fe_{24}O_{41}$ (Zn_2Z), with a 35 GHz threshold field of 30 mOe.[5]

The results of Dixon[1] and Goldberg[5] included high power FMR absorption curves which exhibited so-called "foldover" effects. Because of the possible dependence of the FMR frequency on sample shape, magnetization, anisotropy, and g factor, under certain conditions that frequency can change with the level of microwave power exciting the resonance. This leads to a reentrant or bistable resonance absorption curve depicted schematically in Fig. 1 for the case of a shift to higher field. Upon traversing the resonance with increasing field (upsweep), one would, in principle, measure curve (a). One would measure curve (b) for a decreasing or downsweep field.

Theoretical investigations of foldover due to sample shape,[6] spin-wave interactions,[7] and magnetocrystalline anisotropy[8] have also been reported. These works were primarily concerned with the saturation of the FMR absorption observed first by Damon[9] and Bloembergen and Wang[10] and explained in terms of spin-wave instability processes by Suhl.[11] Weiss reported distinct bistable behavior and jitter on the FMR curve at high power for single crystal Mn–ferrite,[12] but the dominant effect was simply a broadening of the FMR absorption curve. A pronounced bistable behavior was observed for high power FMR in thin spinel ferrite disks[13] and, more recently, in uniaxial Zn_2Z as noted above.[5]

On the whole, none of these foldover effects were as spectacular as the standard schematic absorption curve used to illustrate the phenomenon, as shown in Fig. 1. The present

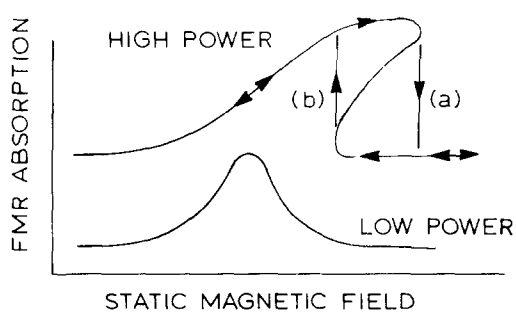

FIG. 1. Schematic FMR absorption curves at low and high power. Foldover is illustrated by the upper reentrant absorption curve. An FMR recorder trace would exhibit an abrupt discontinuity at (a) for increasing field and at (b) for decreasing field.

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paper presents new FMR results on two planar hexagonal ferrites, Zn_2Y and Mn–ZnY, in which dramatic foldover effects are shown. The bistable jump corresponding to curve (a) in Fig. 1 is shifted above the FMR position by 300–500 Oe at the highest powers available (350 mW incident, 58 mOe microwave field amplitude). The downsweep trace corresponding to curve (b) exhibits a steep, sharp cusp prior to rejoining the low-field tail of curve (a). Finally, transient effects, with time constants on the order of seconds, are apparent.

EXPERIMENT

The measured FMR absorption curves were obtained at room temperature and 8.9 GHz, with some additional data for frequencies up to 11.17 GHz. Data were obtained on two compositions, $Ba_2Zn_2Fe_{12}O_{22}$ (Zn_2Y) and $Ba_2Zn_{1.375}Mn_{0.625}Fe_{12}O_{22}$ (Mn–ZnY). The samples were rounded platelets with a diameter to thickness ratio of approximately five. The FMR absorption was obtained by monitoring the microwave power reflected from a x-band waveguide short. For most of the measurements, samples were mounted on a thin dielectric rod in the center of the waveguide, one-half wavelength from the shorting plate. To study possible heating effects, a series of measurements was also made with samples glued securely in the center of the shorting plate. The power incident upon the shorted section was controlled by a precision variable attenuator following the klystron source. The peak power at 8.9 GHz was 350 mW. A second precision attenuator in the detector arm of the spectrometer was used in a complimentary fashion, so that all FMR recorder traces were normalized to approximately the same off-resonance detector power. The microwave and dc magnetic fields were mutually perpendicular and both in the plane of the platelets, which was also the easy magnetic plane. Data were obtained by sweeping the dc magnetic field at a fixed rate and monitoring the reflected microwave power versus field with a conventional diode detector.

Figure 2 shows ten sets of 8.9 GHz resonance curves for the Zn_2Y platelet at a sweep rate of 400 Oe/min. The incident power in db below 350 mW and the microwave field amplitude, rounded off to mOe, at the sample are indicated for each pair of sweeps. At -20 db, nonlinear effects are just beginning to appear, in the form of a small asymmetry between the curve for increasing dc field, labeled (I), and the curve for decreasing field, labeled (D). At lower powers, the traces correspond to conventional FMR with a linewidth of approximately 45–50 Oe. The small displacements between the two curves will be discussed at the end of this section.

As the power is increased above -15 db, the FMR for increasing field (upsweep) broadens, develops a pronounced asymmetry, and exhibits a peak shift to higher field. At the highest powers, the steep high-field side of the FMR evolves into a sharp shoulder shifted above the low field FMR position by approximately 350 Oe. The evolution of the FMR curve for decreasing field (downsweep) with increasing power is rather different. At moderate powers, this curve also develops a pronounced asymmetry, but shows no appreciable broadening. At the highest powers, the peak has more the

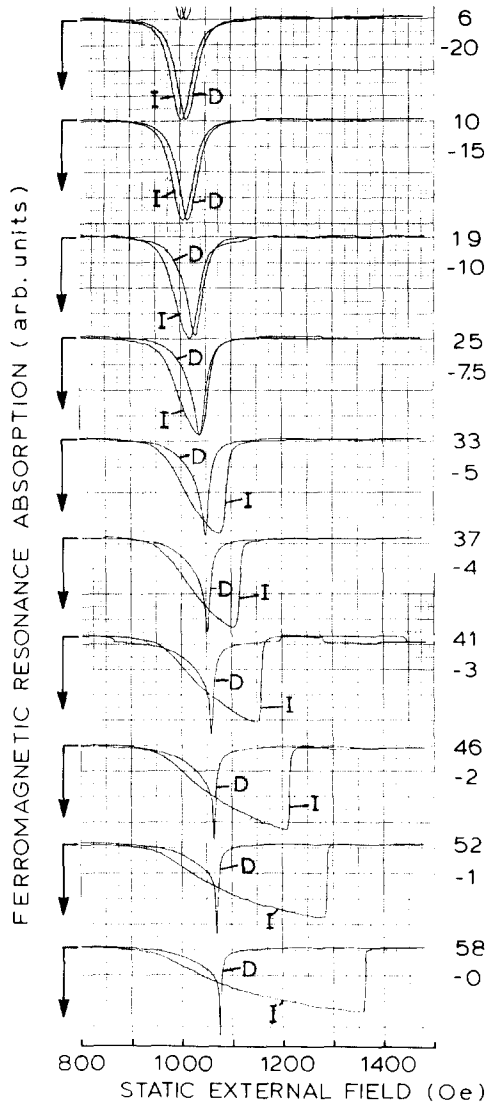

FIG. 2. Upsweep (I) and downsweep (D) FMR absorption vs applied static field for a thin platelet sample of Zn_2Y hexagonal ferrite at room temperature and 8.9 GHz. The pair of numbers to the right of each set of curves indicate the microwave field amplitude in mOe (upper) and incident power in db relative to 350 mW (lower). The microwave and static fields were both in the slab plane. The sweep rate was 400 Oe/min.

appearance of a sharp cusp, shifted by 60–70 Oe from the low power FMR position.

The power at which the sharp shoulder for upsweep and sharp cusp for downsweep occurs is reduced if the sweep rate is reduced. This effect is shown in Fig. 3. Pairs of FMR curves are shown for a fixed incident power of -5 db, as a function of the dc magnetic field sweep rate. The third pair of curves at 400 Oe/min in Fig. 3 corresponds to the fourth pair in Fig. 2. It is seen that the rounded character of the curves for the faster sweep rates evolve into the sharp shoulder and cusp-like features at the slower sweep rates. The ratio of the shoulder width to sweep rate converts to a time constant of 5–10 sec.

Figure 4 shows similar data at maximum power, again for the Zn_2Y platelet. As the sweep rate is decreased, the high-field shoulder appears to be converging to a "steady state" dc field location of 1375 Oe. In addition, the low-field

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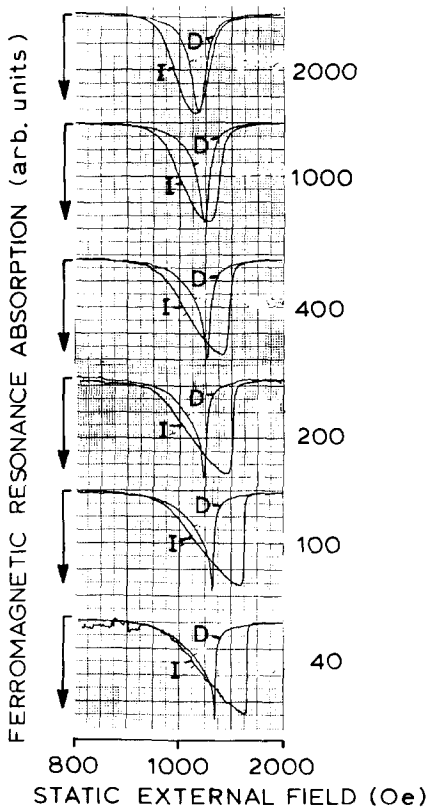

FIG. 3. Upsweep (I) and downsweep (D) FMR absorption curves as in Fig. 2 but as a function of sweep rate at a fixed incident power of 110 mW or 33 mOe microwave field amplitude. The sweep rates are indicated on the right side of each set of curves, in Oe/min.

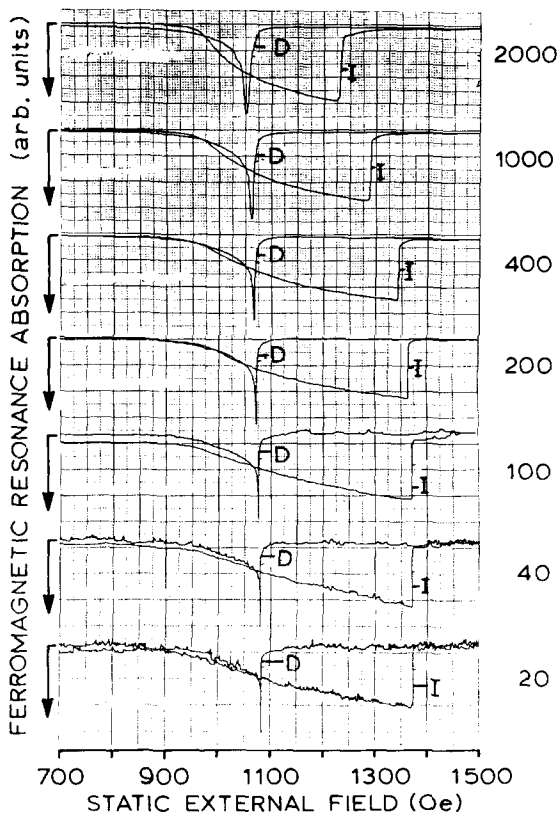

FIG. 4. Upsweep (I) and downsweep (D) FMR absorption curves as in Fig. 3 but as a function of sweep rate at a maximum power, 350 mW incident or 58 mOe microwave field amplitude. The sweep rates are indicated on the right side of each set of curves, in Oe/min.

tail of the downsweep FMR curve appears to converge with the low-field portion of the upsweep FMR curve. The vertical displacements between these curves for the bottom three pairs of traces is due to vertical offset drift. At 20 Oe/min sweep rate, the downsweep cusp is extremely sharp.

Figure 5 shows a series of FMR upsweep and downsweep absorption curves for the platelet of Mn–ZnY as a function of incident power for a sweep rate of 400 Oe/min, the same as in Fig. 2. The same general comments apply as for Fig. 2, but with several significant differences. First, the high field shoulder for upsweep FMR appears to be converging to a dc field around 1400 Oe at the highest powers for this sweep rate, while the shoulders in Fig. 2 for this same sweep rate are still moving up in field at the highest powers. Second, the convergence of this shoulder position to 1400 Oe is accompanied by a pronounced decrease in the peak amplitude of the FMR absorption. No such decrease is evident from Fig. 2. The low power FMR linewidth for this Mn–ZnY platelet is 30–35 Oe, somewhat less than that for the

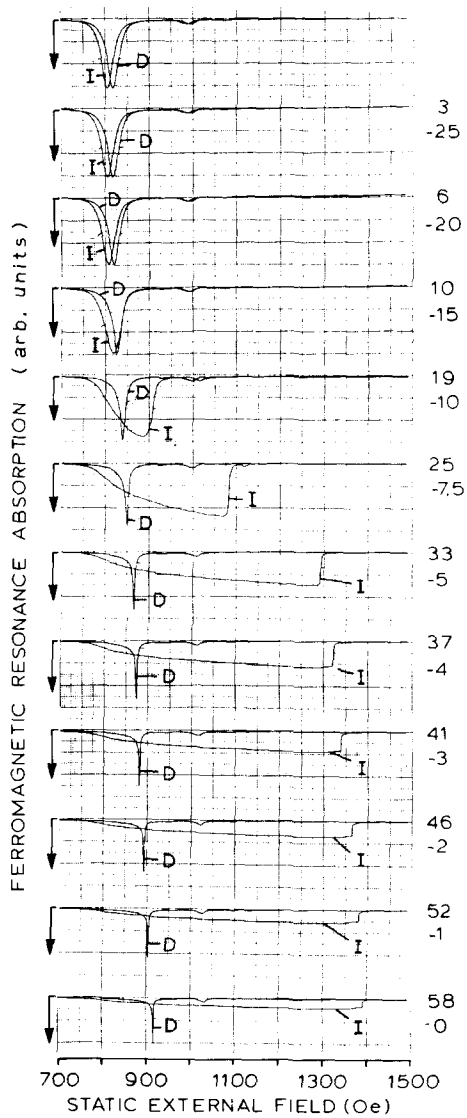

FIG. 5. Upsweep (I) and downsweep (D) FMR absorption curves as in Fig. 2 but for a thin platelet of Mn–ZnY hexagonal ferrite.

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Zn_2Y sample. It is not unreasonable, therefore, for the nonlinear behavior to start at lower power levels and saturate at lower power levels than for the Zn_2Y. The narrow linewidth is probably responsible for the resolved magnetostatic mode at 990 Oe. This mode shifts position by about 40 Oe at the highest powers.

The small but distinct relative shifts between the upsweep and downsweep FMR curves at the lowest powers, evident in Figs. 2 and 5, are partly but not completely due to instrumentation time constants. Somewhat smaller shifts were observed at high sweep rates for FMR in yttrium iron garnet (YIG) polycrystals of comparable linewidths, with shifts of 15 Oe at 2000 Oe/min compared to 30 Oe for Zn_2Y, for example. The upsweep and downsweep absorption curves for the YIG, however, were found to converge as the sweep rate was decreased below 100 Oe/min, while the Zn_2Y FMR curves were still displaced by about 10 Oe at this sweep rate. It is to be noted that the upsweep FMR curve peaks at a lower field than does the downsweep curve. All obvious sources of lag, such as eddy currents, magnet induction, or hysteresis, would produce the opposite effect.

DISCUSSION

The experimental results presented above are noteworthy for both fundamental and practical reasons. Fundamentally, a particularly striking resonance foldover effect and long time-constant transient behavior are evident, with foldover shifts on the order of magnitude of the field for resonance. Practically, the nonlinear behavior is seen to occur at low waveguide power levels on the order of 10 mW. These effects might therefore be very useful for certain applications, such as limiters at millimeter wave frequencies, or very detrimental, as in high power millimeter wave filters. While a satisfactory explanation of these effects has not yet been found, some comments on possible origins of the behavior are in order.

Foldover effects in ferromagnetic resonance, as discussed in the introduction, are generally related to a shift in resonance field from the theoretical small signal value as the microwave pump field amplitude is increased. The low power FMR resonance condition applicable to planar anisotropy platelets magnetized in the easy plane is

$$(f/g)^2 = [H_0 + (N_y - N_z)4\pi M_0] \times [H_0 + (N_x - N_z)4\pi M_0 + H_k], \quad (1)$$

where f is the resonance frequency, g the applicable g-factor, H_0 the applied dc magnetic field, N_z, N_y, and N_z the applicable demagnetizing factors along the c axis (perpendicular to the plane of the platelet), the microwave field direction, and the static field direction, respectively; $4\pi M_0$ is the saturation induction, and H_k the anisotropy field. For literature values of the above material parameters,[14,15] $g = 2.62$ GHz/kOe, $4\pi M_0 = 2.850$ kG, and $H_k = 9$ kOe, and $N_x = 0.6$, $N_{y,z} = 0.2$ for the Zn_zY platelet, one calculates an FMR field value of 1032 Oe, in reasonable agreement with the observed value of 1010 Oe.

From the shift of the upsweep absorption curves as the power is increased, as evident in Figs. 2 and 5, it is apparent that the shift in FMR with power for planar ferrites is to higher fields. From Figs. 2 and 4, the shift for Zn_2Y at 350 mW is approximately 360 Oe. From Fig. 5, the shift for Mn–ZnY at 350 mW is 600 Oe. It is perhaps misleading, however, to describe the nonlinear behavior in terms of a simple shift. It is evident from the data that the entire FMR curve becomes distorted at the higher powers. The high field shoulder or cutoff represents the maximum shift. Possible origins of such shifts will be considered shortly.

Such an upward shift is consistent with the behavior of the downsweep absorption curves. From Fig. 2, there is no appreciable FMR absorption upon downsweep until the dc field falls below about 1100 Oe, the field at which the absorption on the high-field tail of the low power FMR absorption starts to increase. As the downsweep field is further decreased, the sharp cusp occurs, followed by a gradual decline which coincides with the upsweep absorption as in the lower traces of Figs. 4 and 5. The sharp cusp is probably a result of the shift of the FMR position to high field as soon as the downsweep field gets low enough (1100 Oe) to excite the tail of the FMR. The cusp occurs as the FMR position, in effect, sweeps through the applied field.

The literature on nonlinear and foldover effects in ferromagnetic resonance provides few clues to explain the present results. The most extensively treated mechanism involves the parametric excitation of spin waves, which leads to nonlinear behavior at high power levels.[16] The theory was extended to planar anisotropy materials by Schlömann *et al.*[2] For platelets, the FMR condition and spin-wave dispersion preclude the parametric excitation of spin waves at half the x-band pump frequency so that first-order instability processes are not allowed. The excitation of spin waves at the pump frequency is allowed and the corresponding second-order spin-wave instability process can therefore give rise to nonlinear behavior. From Fig. 9 of Ref. 2, the present threshold field of 15–30 mOe in Zn_2Y implies a spin-wave linewidth of 1–10 mOe. Such an intrinsic linewidth seems unrealistically small; even for ultrapure single crystal YIG at room temperature spin-wave linewidths exceed 200 mOe. More realistic linewidths, two or three orders of magnitude larger, would imply threshold fields of 100 mOe at 1 Oe, much larger than those apparent from the present data.

In Refs. 1 and 3, spherical samples were used so that first-order instability processes could be a possible source of the nonlinear microwave behavior. Based on this assumption, Ref. 3 estimates the spin-wave linewidth in Mn–ZnY to be about 0.74 Oe at 17.3 GHz for samples with an x-band FMR linewidth of 3.8 Oe. Such values are not unreasonable. In order to check the possibility of a first-order process in the present experiments, the measurements were extended to 11.17 GHz. If a first-order process were operative, one would expect to see an increase in the power required to achieve nonlinear behavior as the frequency is increased. No such change was observed. In summary, then, the interpretation of the microwave threshold for nonlinear FMR in terms of spin wave instability is unclear. The present data imply spin wave linewidths for second-order processes which are unrealistically low. The data of Savage *et al.*[3] im-

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ply reasonable linewidths for first-order processes. Further work on samples of different shapes versus frequency, and for parallel pumping as well, is needed to resolve these conflicts.

Foldover due to magnetocrystalline anisotropy has also been proposed, but quantitative calculations have only been made for ferrites of cubic symmetry.[8] From Ref. 8, the critical microwave for foldover due to cubic anisotropy is 0.5–1.0 Oe, and similar numbers would be expected for hexagonal ferrites with planar anisotropy. These numbers are far larger than observed in the present work.

One possible explanation of an upward resonance field shift is sample heating. Heating would reduce $4\pi M_0$ and thereby increase the external field H_0 required to satisfy Eq. (1) at fixed frequency. If it is assumed that $4\pi M_0$ decreases by 5–6% for a 10 °C temperature increase above room temperature, as appears reasonable from the literature,[14] one obtains a field shift of 25 Oe. It is apparent, therefore, that an appreciable temperature increase approaching the Curie temperature of 400 °K would be required to account for the 300–600 Oe shifts observed experimentally.

An additional check on heating effects was made by bonding the platelet directly on the copper waveguide short and generating FMR curves similar to those in Figs. 2 and 5. Apart from a shift in resonance position due to imaging effects, the onset of nonlinear behavior and the evolution in the upsweep and downsweep curves with increasing power were no different than already described. If heating was an important factor in the results for samples mounted on the dielectric rod, one would expect some change in the response with the sample bonded to the metal short, a reasonable heat sink. It is concluded, therefore, that heating can not account for the nonlinear behavior or the foldover effects reported herein.

CONCLUSION

Nonlinear microwave behavior has been observed for easy plane hexagonal ferrites at very low microwave power levels. This behavior is comprised of very large foldover effects, with FMR field shifts by hundreds of Oe, and transient behavior, with time constants of 1–10 sec. No existing theory appears capable of explaining these effects at present.

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