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Antiferromagnetic-Resonance Linewidths in MnF$_2$†

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The fundamental relaxation mechanisms by which the uniform and magnetostatic modes decay in an antiferromagnet are identified by antiferromagnetic-resonance linewidth observations in MnF$_2$ at 4.2°K in fields of 85 kOe. The dependence of the linewidth—0.2 Oe in the narrowest instance—on sample geometry, surface preparation, and impurity concentrations makes quantitative comparison with the Loudon-Pincus theory possible. The predominant relaxation process is surface pit scattering into the degenerate spin-wave manifold in all but the most impure crystals.

While the linewidth and relaxation of magnetic resonance modes in ferromagnetic and ferrimagnetic insulators have been both well studied and explained,[1] little attention has been given to the problem of antiferromagnetic-resonance (AFMR) linewidth, despite the existence of a quantitative theory[2] for the nonthermal relaxation processes. Here we report the results of an extensive study of the AFMR linewidths of the uniform and magnetostatic modes in pure and impurity-doped MnF$_2$ at 4.2°K.[3] The various mechanisms (e.g., pit scattering and spatial variations in the local field) that contribute to the linewidths have been isolated and identified.

The AFMR frequencies in an easy axis, uniaxial antiferromagnet (AFM) with the external field H_0 applied collinear with the spin moments are, at 0°K,[4]

$$\begin{aligned}&\omega/\gamma=(H_C^2+2H_AN_\perp M_S)^{1/2}\pm H_0,\\&H_0<H_C=(H_A^2+2H_AH_E)^{1/2};\end{aligned}\tag{1}$$

here H_A and H_E are the anisotropy and exchange fields, respectively, M_S the sublattice magnetization, and $N_\perp$ the perpendicular demagnetizing factor. In MnF$_2$ $H_E\simeq 515$ kOe, $H_A\simeq 8.4$ kOe, $H_C\simeq 93$ kOe, and $M_S\simeq 600$ Oe. To minimize the effects of rf field variations across the sample, the downgoing AFMR branch was studied at high fields ($H_0\sim 85$ kOe) and relatively low frequencies (~ 23 GHz). The samples were cut cylindrically symmetric with respect to the c axis, highly polished, and strain-free mounted. Because of the strong angular dependence of the AFMR at high fields[5] the c axis was made collinear with the field to better than 0.1° by tilting the sample, *in situ*, while observing the resonance.

The following qualitative features associated with the uniform mode were noted immediately. Resonance half-widths ΔH are smallest in flat disks ($c\perp$ plane)—$\Delta H\simeq 5.0$ Oe—and the line profiles are Lorentzian as expected for relaxation broadening. These widths are nearly two orders of magnitude smaller than were previously found in AFMR studies of MnF$_2$.[6,7] Small misalignment in the field causes severe broadening and, with increasing misalignment, the uniform mode splits up into many distinct modes. The linewidth is temperature independent below 8°K. It can be increased by an order of magnitude by roughening the surface of the samples. The substitution for Mn of 1% Fe, Zn, or Ni impurities causes negligible linewidth changes despite resonance-field shifts of as much as 7 kOe. Only in samples with Co impurities ($\sim 1\%$) does the linewidth increase fourfold.

The experimental results strongly indicate that the relaxation of the AFMR is dominated by its decay into the degenerate manifold of $k\neq 0$ magnons via surface pit scattering. This mechanism has been found to be an important source of linewidth in ferromagnets[1] and has been theoretically explored for the AFMR by Loudon and Pincus (LP).[2] LP showed that in a finite sample volume dipolar fields cause the AFM spin-wave spectrum to become anisotropic. The dispersion relation for long-wavelength magnons $[r_0^{-1}\ll k\ll (H_A/2H_E b^2)^{1/2}]$ is, assuming $2\pi M_S\ll H_E$ and

$2\pi M_S H_A/H_C \ll H_0$, $(H_C - H_0)$,

$$\omega/\gamma = (H_C^2 + 4\pi M_S H_A \sin^2\theta_k + 2H_E^2 b^2 k^2)^{1/2} \pm H_0; \qquad (2)$$

here $b = az^{-1/2}$, a being the nearest-neighbor distance to spins on opposite sublattices, and z their number; θ_k is the angle between the c axis and the direction of propagation of spin wave $\vec{k}$; and r_0 is the sample radius.[8] Hence from a comparison of Eqs. (1) and (2) we see that except for an infinitely flat disk, there are always $k \neq 0$ spin-wave modes degenerate with respect to the uniform mode into which it can decay, provided there exists a non-k-conserving perturbation. LP have considered several such scattering mechanisms. Extending their calculations to finite fields within the above limits, the following results are obtained.

If pits of radius R uniformly cover the surface of a spherical sample of radius r_0, a resonance half-width $\Delta H \simeq 30(R/r_0)M_S H_A/H_C$ is predicted. The matrix element for surface pit scattering peaks sharply at $kR \simeq 2$, and it is assumed that $2R^{-1} \ll k_m$, where k_m is the maximum wave vector in the degenerate spin-wave manifold. For a nearly spherical ellipsoid with $r_0 \simeq 0.4$ mm, $R/r_0 \simeq 10^{-2}$, $\Delta H \simeq 17$ Oe is observed, which compares well with the predicted one of $\Delta H \simeq 16$ Oe. For a flat disk of thickness d and radius r_0 the pit-scattering half-width is

$$\Delta H = \frac{8\pi^2}{3}\frac{R}{d} M_S \frac{H_A}{H_C}\left(\frac{N_\perp}{2\pi}\right)^2 \left(1 - \frac{N_\perp}{2\pi}\right)^{-1/2}; \qquad (3)$$

here $N_\perp$ is the average demagnetizing factor, experimentally obtained by measuring the resonance-field shift of the disk with respect to an ellipsoid with known $N_\perp$. For a flat disk with $d \simeq 0.05$ mm, $r_0 \simeq 1.5$ mm, $R/d \simeq 2\times10^{-2}$, and $N_\perp \simeq 1$ we observe $\Delta H \simeq 5$ Oe, while Eq. (3) predicts $\Delta H \simeq 0.8$ Oe. We believe the difference between experimental and theoretical half-widths to arise from inhomogeneous scattering, as will be discussed below.

Exchange-coupled impurities (with $S = S'$) give rise to a half-width of

$$\Delta H = (J - J')^2 \frac{S^2 H_A^2}{\pi \hbar^2 \gamma^2 H_E^3}\left(\frac{2N_\perp M_S}{H_E}\right)^{1/2} z^{7/2} f;$$

here J and J' are the host and impurity exchange integrals, respectively, and f is the concentration of impurities. For a flat disk of 1.4% Fe:MnF_2 with $N_\perp \simeq 1$ the experimental contribution to the half-width due to the presence of the impurity is $\Delta H \lesssim 2$ Oe, while we predict a contribution of $\Delta H \simeq 0.2$ Oe. For a sample with as much as ~5% Zn:MnF_2 the observed impurity contribution to the linewidth is $\Delta H \simeq 6$ Oe, while the theory predicts $\Delta H \simeq 1$ Oe. Part of the disagreement here might arise from impurity magnon interactions that are neglected in the theory. The comparably large change in linewidth for ~1% Co-doped MnF_2 is attributed to the strong spin-orbit coupling of the Co^{2+} ion. From all of the above results we conclude that the linewidth contribution from any impurities in the relatively pure samples must be totally insignificant.

We calculated the linewidth resulting from a spatially varying z component of H_0 that has a mean square deviation of $(\delta H)^2$, when averaged over the sample. This is analogous to the calculation of Geschwind and Clogston[9,1] for the ferromagnet. We find a half-width $\Delta H = (\delta H)^2 (H_C/4M_S H_A) \times (1 - N_\perp/2\pi)^{-1/2}$ and that the effect of the inhomogeneous field is "narrowed" as long as $\delta H \ll 4M_S \times H_A/H_C$, which for MnF_2 requires $\delta H \ll 220$ Oe. When several samples were deliberately placed into an inhomogeneous field region with $\delta H \simeq 10$ Oe, no noticeable broadening of the resonance linewidth occurred, in agreement with the theoretical prediction.

Likewise a variation $\delta N_\perp$ of the demagnetizing factor across a nonellipsoidal sample will produce a spatial variation of the resonance field $\delta H \simeq \delta N_\perp M_S H_A/H_C$. This acts as does an inhomogeneous z-directed field of the same magnitude and produces a linewidth

$$\Delta H = (\delta N_\perp)^2 (M_S H_A/4H_C)(1 - N_\perp/2\pi)^{-1/2}. \qquad (4)$$

By cutting a nearly spherical ellipsoid in half we observe an effective change in linewidth of $\Delta H \simeq 20$ Oe, while we predict, with $\delta N_\perp \approx 1$, $N_\perp \approx 4$, a broadening of $\Delta H \approx 23$ Oe. In a flat disk with $d \simeq 0.05$ mm, $r_0 \simeq 15$ mm, the variation of the demagnetizing factor is $\delta N_\perp \approx 0.3$; the resulting broadening will be $\Delta H \approx 2$ Oe. This partially explains the difference between the observed and calculated widths from pit scattering in flat disk samples, as alluded to above.

Magnetostatic modes have been well studied in ferromagnets,[10] but observed only once in a canted AFM.[11] Theoretical work on the magnetostatic modes in an AFM is scant.[12] We observed magnetostatic modes in all of our flat disk samples. A typical spectrum is shown in Fig. 1. In this sample magnetostatic modes were observed on the low- and high-field sides of the uniform mode.

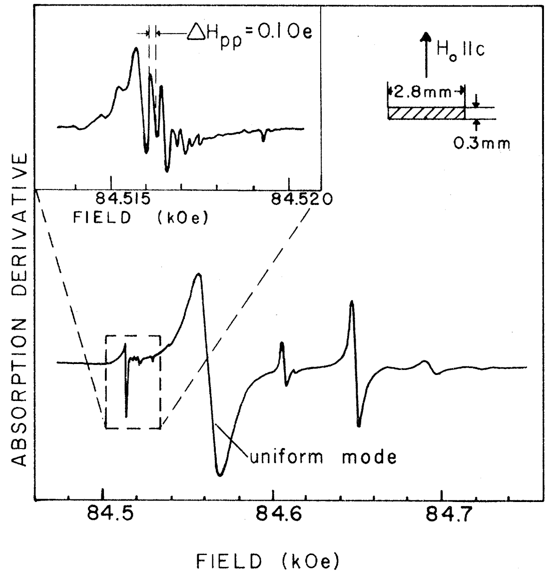

FIG. 1. AFMR uniform mode and magnetostatic modes versus the field in a flat disk of MnF_2 at $T = 4.2°K$, $\nu = 23.1$ GHz. The inset shows a blowup of the low-field lines with decreased field modulation (~ 0.1 Oe).

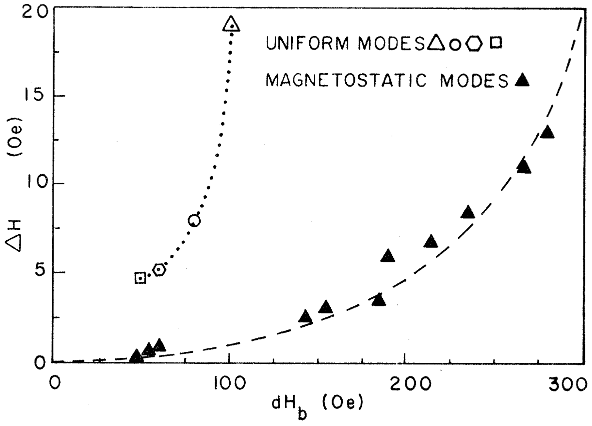

FIG. 2. Half-width ΔH of the magnetostatic modes of sample shown in Fig. 1 and the uniform modes of various disks as a function of their spacing dH_b from the bottom of the spin-wave band. The dashed line is a theoretical prediction of the pit-scattering–induced linewidth [Eq. (3)] with $R/d \simeq 6\times 10^{-3}$ and an effective $N_\perp = (dH_b)H_C/M_S H_A$, which is obtained from Eq. (1). The dotted line is a "best fit" to the available uniform-mode linewidth data, which increase monotonically with disk thickness. The origins of these widths are discussed in the text.

Those below the uniform mode are extremely narrow as is shown in the Fig. 1 blowup. Tilting the sample in the field splits the uniform mode into several modes, indicating that there are magnetostatic modes nearly degenerate with the uniform mode even at optimum allignment. These results suggest that the uniform mode is broadened partially by excitation of nearly degenerate magnetostatic modes and partially by inhomogeneous demagnetization which allows the uniform mode to relax via unexcited, almost degenerate magnetostatic modes. This is illustrated in Fig. 2. Here the linewidth of the uniform mode, for disks with different $N_\perp$, and of the magnetostatic modes for the particular disk shown in Fig. 1, is plotted versus their spacing dH_b from the bottom of the spin-wave band. As seen from Eq. (1) this spacing is proportional to $N_\perp$. Notice that the uniform-mode widths increase more rapidly with dH_b than do the magnetostatic-mode widths. We believe the magnetostatic modes are less affected by inhomogeneous demagnetization, as their transverse magnetization varies across the sample and tends to partially cancel inhomogeneous effects. Hence their width is more suitably compared with the theoretical pit-scattering result of Eq. (3), which is shown by the dashed line in Fig. 2. Deviation of the experimental data from the line itself might be partially due to inhomogeneous effects.

The variation of the pit-scattering–induced width with dH_b is more pronounced in an AFM than in a ferromagnet (FM). One may compare the physical quantities in a FM and an AFM which relate to the linewidth contributions caused by decay into the degenerate manifold.[1,2] In an AFM the width of the spin-wave band near $k = 0$ and dH_b are both smaller than in a FM by a factor of H_A/H_C. As the width of the spin-wave band is smaller in the AFM, one might expect an enhancement of the pit-scattering linewidth by the factor H_C/H_A, due to the increased density of states. This is more than compensated for by a factor $(u_0+v_0)^2 \simeq H_A/H_C$ in the scattering matrix element. Here u_0 and v_0 are the transformation coefficients of the antiferromagnetic spin-wave operators[2] in the $k \to 0$ limit. While the pit-scattering linewidth has little sample-shape dependence in the FM, it increases like $\sim N_\perp^2$ in the AFM.[13] The linewidth due to inhomogeneous demagnetization is smaller in the AFM by $H_A/2H_C$, because of the smaller variation of the resonance field with $N_\perp$.

Some preliminary linewidth studies were carried out at 70 GHz in a flat disk in which, for the uniform mode, $\Delta H \simeq 5$ Oe at 23 GHz. At 70 GHz ΔH increases to 39 Oe. Although more magnetostatic modes are excited at the higher frequency, the widths of these modes are comparable to magnetostatic-mode widths at low frequencies and range from only $\Delta H \simeq 4$ to 9 Oe. This suggests that the broadening of the uniform mode at higher frequencies in a flat disk arises from stronger excitation of magnetostatic modes nearly degenerate with the uniform mode, and might explain why narrow resonance lines were not observed in the original 240-GHz AFMR studies.[6]

The line shape and position of the AFMR in MnF_2 is strongly power dependent at moderately high microwave power levels ($H_{rf} \sim 0.05$ Oe).[14] A detailed account of these effects and the temperature dependence of the linewidths will be published elsewhere.

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Observation of Cooperative Nuclear Magnetic Order in $PrCu_2$ below 54 mK

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Magnetic-susceptibility and specific-heat measurements in Van Vleck paramagnetic $PrCu_2$ down to 30 mK reveal that the Pr nuclei enter an antiferromagnetically ordered state below 54 mK. This high nuclear ordering temperature results from magnetic exchange interactions between the Pr ions, which must be close to the critical value necessary for spontaneous electronic magnetic order in this compound.

In the presence of weak exchange interactions between Van Vleck paramagnetic ions, an indirect exchange coupling between the nuclei of these ions results due to second-order hyperfine effects. The physical mechanism of this coupling can be described as an exchange coupling of $4f$ angular moments $\langle J_{4f} \rangle$ which the hyperfine interaction admixes to the $2I+1$ nuclear substates of the singlet ground state. Alternatively, one can also describe it as a Suhl-Nakamura[1]–type mechanism in which a nuclear spin flip at one site excites a collective crystal-field excitation[2,3] (through the hyperfine coupling) which can be reabsorbed by another nucleus at a neighboring site. This coupling is expected to lead to nuclear ferromagnetism or antiferromagnetism at low temperatures.[4-7] If the exchange interactions between the singlet ground-state ions exceed a critical value, the singlet ground state becomes unstable against spontaneous mixing with the higher excited crystal-field states below some (electronic) magnetic ordering temperature.[8] In this case the nuclear moments will align in the local hyperfine field of the exchange-induced ordered moment at low temperatures. We have found experimentally that the Pr nuclei in the compound $PrCu_2$ order antiferromagnetically below 54 mK. This is the first example of an unusually high cooperative nuclear ordering temperate, and we believe that in $PrCu_2$ the exchange interaction between Pr ions must be less than (but close to) the critical value necessary for electronic magnetic order.