Nonreciprocal Optical Reflection of the Uniaxial Antiferromagnet MnF_2

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Microwave reflectivity measurements in Voigt geometry were performed on a polished surface of MnF_2. We observed nonreciprocal behavior in the reflectivity with respect to the magnetic field reversal. The nonreciprocal behavior of the antiferromagnet's reflectivity gives clear evidence for the existence of nonreciprocal dipolar antiferromagnetic surface spin-wave excitations. The results are quantitatively explained. They give very accurate values for antiferromagnetic resonance and spin-wave damping.

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The influence of magnetic dipolar fields on ferromagnetic resonance, magnetostatic modes, and long-wavelength spin waves in ferromagnets and ferrimagnets is well documented. It is less well known that similar effects can be observed also in antiferromagnets. This has been shown in particular by Kotthaus and Jaccarino[1] for the uniaxial antiferromagnet MnF_2 using antiferromagnetic (AF) resonance techniques.

For the corresponding surface-wave excitations (surface spin waves, magnetic surface polaritons) the situation for the two types of magnets is even more drastic. While dipolar surface spin waves for ferromagnets and ferrimagnets have been observed for a number of substances,[2] in the case of antiferromagnets there exist only a few calculations but no experimental evidence so far.[3]

In this Letter we present indirect evidence for the existence of antiferromagnetic dipolar-type surface spin waves by showing microwave reflectivity experiments in MnF_2 for a particular geometry of applied magnetic field and of plane of incidence. The key feature of the experiment is the nonreciprocity of the surface-wave excitation and of the reflectivity itself, in the presence of a magnetic field. Thus if either the direction of the incident and reflected waves is reversed, or the direction of the applied field is reversed, the reflection coefficient is changed. In the case of Voigt geometry a general symmetry argument[2,4] states that the energy degeneracy of a surface-wave excitation is raised, i.e., $\omega(k_x,B_y)\neq\omega(k_x,-B_y)$ and $\omega(k_x,B_y)\neq\omega(-k_x,B_y)$ with ω the excitation frequency, k_x the propagation direction in the plane, and B_y the magnetic field in y direction also in the surface plane. Such nonreciprocal effects have been observed for dipolar ferromagnetic spin waves,[5,6] for surface polaritons in semiconductors,[7] and for surface acoustic waves in metals.[8] In addition, nonreciprocal reflection of electromagnetic waves has been observed for InSb.[9]

In analogy to the case of a semiconductor,[9] we present in Fig. 1 the calculated antiferromagnetic dipolar surface spin waves,[3] the corresponding antiferromagnetic polaritons,[10] and the calculated reflectivity for an angle of incidence of 45°, using the parameters of MnF_2 for $t=4.2$ K taken from Ref. 3 (anisotropy field $H_a=0.787$ T, exchange field $H_{ex}=55$ T, sublattice magnetization $M=0.06$ T, zero-field AF resonance field $\omega_0/\gamma=9.337$ T, $\epsilon_{yy}=5.5$).

If we confine our considerations to polaritons with **k** parallel to the x axis and $E=(0,E_y,0)$, we obtain the dispersion relation for AF surface polaritons:

$$\mu_v[k^2-\omega^2/c^2]^{1/2}+[k^2-\mu_v\epsilon_{yy}\omega^2/c^2]^{1/2}+(i\mu_{xz}k/\mu_{xx})=0 \quad (1)$$

with

$$\mu_v=\mu_{xx}+\mu_{xz}^2/\mu_{xx};$$
$$\mathscr{K}=-[\epsilon_{yy}(\mu_{xx}+\mu_{xz}^2/\mu_{xx})-\sin^2\alpha]^{1/2}.$$

μ_{xx}, μ_{xz} are the diagonal and off-diagonal tensor components of the permeability tensor. They refer to dipolar spin excitations of antiferromagmetic systems[3,10]:

$$\mu_{xx}=1-4\pi(\gamma/\omega_0)^2MH_a[A+B],$$
$$\mu_{xz}=i4\pi(\gamma/\omega_0)^2MH_a[A-B] \quad (2)$$

with

$$A=[(\omega/\omega_0+H(\gamma/\omega_0)+i/\omega_0\tau)^2-1]^{-1},$$
$$B=[(\omega/\omega_0-H(\gamma/\omega_0)+i/\omega_0\tau)^2-1]^{-1},$$

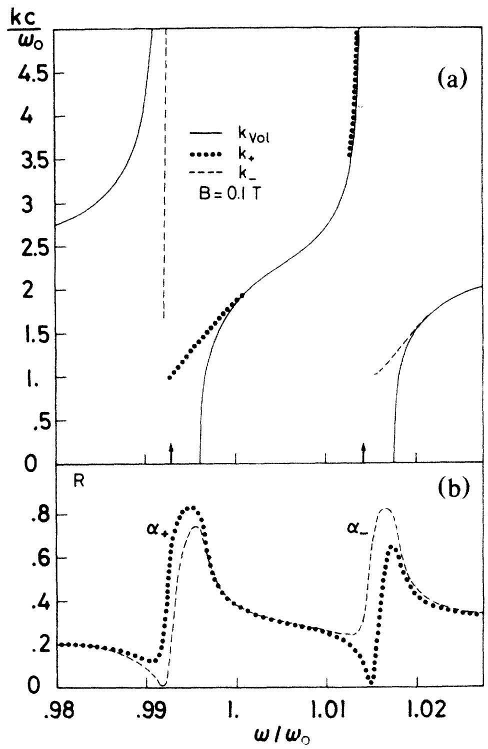

FIG. 1. (a) Dispersion relation of AF volume polaritons (full lines) and AF surface polaritons (dashed lines and dotted lines, corresponding to k^- and k^+; $B=B_y$ lying in the surface perpendicular to k^- and k^+, respectively). Damping is neglected. The frequencies of the unretarded magnetostatic surface modes are indicated by the two arrows. (b) Calculated reflectivity for $\alpha = \pm 45°$ with $(\omega_0\tau)^{-1} = 0.0005$.

and H = applied field.

The results of Fig. 1(a) are obtained by solving Eq. (1) without damping and with the material parameters for MnF_2 listed above. Introduction of finite τ leads to bend-back effects which are necessary for real-photon–surface-plasmon coupling.[11] Therefore the calculation of the reflectivity is carried out with an appropriate value of τ.

The calculation of the reflectivity on a half space for the geometry mentioned above reads

$$r = \frac{1-\beta}{1+\beta} \text{ with } \beta = \frac{(\mu_{xx}\mathscr{H} - \mu_{xz}\sin\alpha)\cos\alpha}{\sin^2\alpha - \epsilon_{yy}\mu_{xx}}. \quad (3)$$

Equation (3) exhibits nonreciprocity in the linear term $\mu_{xz}\sin\alpha$. $R = |r|^2$ is plotted as a function of ω/ω_0 in Fig. 1(b). Nonreciprocal features in R occur in exactly the same frequency region as the analogous nonreciprocal features of the AF polaritons of Fig. 1(a). This is exactly analogous to the case of a magnetoplasma.[9] The difference between reflection of electromagnetic waves from a semiconductor[9] and the reflection from an antiferromagnet is in the geometry of the experiment. In the present case for magnetic dipolar excitations in antiferromagnets, we have the E vector parallel to the plane of the surface and the B vector of the microwave in the plane of incidence. For the semiconductor or in reflection of visible light from ferromagnets[12,13] (equatorial Kerr effect) the B vector is parallel to the surface and the field E is in the plane of incidence.

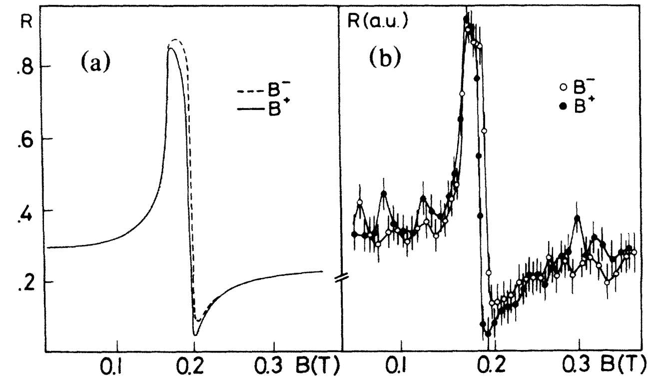

FIG. 2. (a) Calculated reflectivity with $(\omega_0\tau)^{-1} = 8\times10^{-5}$ and $\omega_0(4.2\ \text{K}) = 1.63$ THz for both directions of the applied field B. (b) Measured reflectivity for B^+ and B^- at 4.2 K.

The MnF_2 sample used for the reflection measurements has an optically polished front surface and an irregular background surface in order to prevent thickness interferences. (See Ref. 9.) The electromagnetic radiation at 264 GHz was generated by a commercially available impatt diode with a fundamental frequency of 88 GHz coupled to a third-harmonic resonator. The experimental setup and geometry is similar to the case of Ref. 9 except for the polarization direction as discussed above. As a detector we used a Golay cell. The experiment was performed at different temperatures in a superconducting magnet.

Figures 2 and 3 show calculated reflectivities and the experimental results at fixed frequency (264 GHz) for different temperatures (4.2 and 28 K) as a function of applied magnetic field. For technical reasons the re-

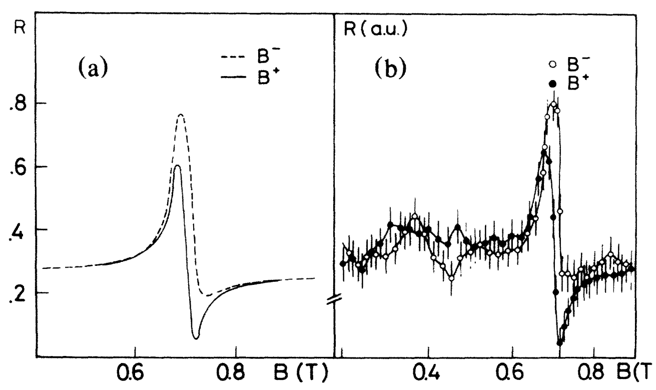

FIG. 3. (a) Calculated reflectivity with $(\omega_0\tau)^{-1} = 6.5\times10^{-4}$ and $\omega_0(28\ \text{K}) = 1.54$ THz. (b) Measured reflectivity for B^+ and B^- at 28 K.

flectivity was measured point by point. One notices rather strong noise in the transparency region outside the resonance peak. This effect arises as follows: For guiding the radiation we use stainless steel tubes acting as an oversized waveguide. In the microwave region then we have single-mode contributions which depend very sensitively on frequency instabilities. For far-infrared radiation (see Ref. 9) the mode density is about a hundred times larger, so that single-mode contributions will average to very small values. The detector noise (bandwidth of 10 Hz) is indicated in the figures by error bars.

These results, especially the ones for 28 K, exhibit clearly nonreciprocal effects with respect to the direction of applied magnetic field; hence they manifest indirectly the nonreciprocal nature of dipolar AF surface polaritons (Fig. 1). The experimental curves look very similar to those calculated with the parameters given above. For a detailed fit the literature values for $H_a(T)$, $H_{ex}(T)$, and $M(T)$ are not accurate enough. Therefore the value of $\omega(B,T)$ was fitted to the experimental curve. This method then allows the determination of $\omega_0(T)$ with high accuracy.

For the same reasons as in the case of semiconductors[9] the nonreciprocal reflectivity disappears for negligible damping of the spin excitations. In order to consider damping we introduce a phenomenological Bloch relaxation time. Again this parameter is fitted to the experimental data of Fig. 2. It is seen that the damping increases with increasing temperature.

With knowledge of $\tau(T)$ we are able to calculate the temperature dependence of the corresponding linewidth of the resonant uniform mode: For $T=28$ K we found $\Delta H = 63 \pm 6$ G, and for $T=4.2$ K $\Delta H < 8$ G, in quantitative agreement with the observed linewidths presented in Ref. 1. Also the nonreciprocal character of the reflectivity appears more significantly with increasing temperature in agreement with the calculations.

Similar reflection measurements on CoF_2 also exhibit nonreciprocal effects with respect to the direction of the magnetic field but we find only qualitative agreement with the corresponding calculations. CoF_2 is not such an ideal uniaxial *S*-type ion antiferromagnet as MnF_2, which exhibits clear-cut nonreciprocal features as shown here. Inspection of Fig. 1 leads to the conclusion that this nonreciprocal feature in the reflectivity implies also the existence of nonreciprocal dipolar antiferromagnetic surface spin waves. Therefore this experiment contributes the first proof for their existence.

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