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Experimental determination of magnetic polariton dispersion curves in FeF_2

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A series of high-resolution attenuated total reflection and reflectivity measurements in the far infrared are used to obtain experimental dispersion curves for bulk and surface magnetic polaritons in uniaxial antiferromagnets. In addition to bulk and surface modes, the individual spectra also allow identification of, to the best of our knowledge, previously unseen surface resonances. The spectra also show huge nonreciprocity in infrared reflectivity at low fields, a feature unique to magnetic systems. [S0163-1829(97)06205-X]

In recent years the study of antiferromagnetic material has received increasing attention. This resurgence of interest is based in part on improved growth techniques which have produced high-quality bulk samples, and have allowed fabrication of ultrathin antiferromagnetic films and multilayers.[1–3] These structures allow the exploration of properties that are central to the fundamental physics of magnetism—the exchange interaction and magnetic configurations—in systems with truly localized spins, in contrast to studies on metallic ferromagnets. For example, superlattices of FeF_2/MnF_2 and CoO/NiO demonstrate strong exchange coupling between the two magnetic systems at the interfaces.[1–3] Most recently, there have been studies of very thin antiferromagnetic films which appear to have a magnetic structure different from that of the bulk and which have a net magnetic moment due to an uneven number of spin planes.[4]

Antiferromagnets may also play an important technological role in the near future. First, antiferromagnets are being used for biasing the spin-valve structures used in the giant magnetoresistance effect.[5] Second, and in direct connection to the results in this paper, antiferromagnets have excitation frequencies that typically lie in the far infrared. These are well above the frequencies available in ferromagnets. Thus, antiferromagnets may play an important role in signal processing in the far infrared. This is particularly true in regard to the signal processing features that are unique to magnetic systems—tunable nonreciprocal behavior.

One of the most fundamental methods for probing a magnetic system is to look at the spin excitations since these provide information on the microscopic parameters, exchange, and anisotropy fields that govern the spin system. For larger wave vectors this can be done by inelastic neutron scattering,[6] but surface excitations are not generally seen. At very small wave vectors, the spin excitations can couple to the electromagnetic field to produce a magnetic polariton. The appropriate tool for measurement is then far-infrared reflectivity. Thus it is somewhat surprising that there exists no complete experimental study of magnetic polaritons in antiferromagnets, in spite of many theoretical works.[7,8]

The reason for this lack is that such experiments are quite difficult. Magnetic excitations have a narrow linewidth—on the order of $0.05\ \mathrm{cm}^{-1}$. Thus, observation requires very-high-frequency resolution. As a result, the few earlier measurements of bulk magnetic polaritons concentrated on working with a laser at a single frequency.[9,10] Magnetic surface polaritons have only recently been directly observed.[11]

We note that magnetic surface polaritons have a special property of nonreciprocity where the frequency of the surface mode depends on the sign of the wave vector.[12] Thus, reversing the direction of propagation can lead to a different frequency, i.e., $\omega(k)\neq\omega(-k)$. Similarly, we find a nonreciprocal reflectivity where the reflection coefficient depends on the direction of propagation. In our study of reflectivity, we find an enormous nonreciprocal reflectivity where reversing the propagation direction changes the reflectivity from nearly 80% to near zero. This kind of behavior, which occurs at small applied fields, is unique in the infrared region and should be of great interest for signal processing in the infrared.

In this paper we present a complete experimental study of bulk and surface magnetic polariton dispersion curves for a uniaxial antiferromagnet. In addition, we also obtain features that are indicative of surface resonances which have been discussed theoretically in the literature,[13] but have not been previously identified. We note that the dispersion curves for surface modes are likely to be the ones most influenced by interface exchange coupling, and thus this is an important step in obtaining accurate values for interface exchange in antiferromagnets coupled to other magnetic materials. Finally, we note that since we examine the entire range of relevant frequencies and wave vectors, we can trace out the nonreciprocal *group* velocity of the surface modes. Such information is vital for possible device applications in delay lines, phase shifters, and isolators.

The FeF_2 single crystal used was grown at Bell Laboratories.[14] The crystal is placed with its reflecting surface in the xz plane. The c axis (easy axis) is parallel to the external magnetic field in the z direction. The measurements were carried out using s-polarized light with the plane of incidence in the xy plane. The resolution has been increased from $0.06\ \mathrm{cm}^{-1}$, which was used in the previously reported observation of the magnetic surface polariton[11] using ATR (attenuated total reflection), to about $0.02\ \mathrm{cm}^{-1}$. The higher resolution is required to resolve the surface modes in the reststrahl regions from the bulk edges.

We obtain the dispersion curves, frequency vs wave vector, from the oblique incidence reflectivity and ATR spectra. The spectra show the response of the crystal as a variation in the reflectivity as a function of frequency, and it is by examination of these variations that we identify the various magnetic modes in the dispersion curves. The wave vector for the bulk and surface modes is defined by the scattering

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geometry. For the reflectivity measurements, the component of the wave vector parallel to the surface is given by $q = (\omega/c)\sin\phi$. Here ω is the frequency of the radiation and c the speed of light in a vacuum, and ϕ is the angle of incidence.

In the ATR measurements a silicon prism is placed above the sample, in front of the incident beam. The incident light is totally internally reflected off the base of the prism, but an evanescent wave extends into the antiferromagnet. The scan line in this case is given by $q = (\omega/c)\epsilon_p^{1/2}\sin\phi$ where ϵ_p is the dielectric constant of the prism and the incident angle is measured inside the Si prism.

Three different silicon prisms, $\epsilon_p = 11.56$, with angles of incidence of 30°, 45°, and 50° were used. With the reflectivity measurements, this gives four different scan lines. Reversing the external field is equivalent to reversing the sign of q giving a total of eight scan lines mapping out the dispersion curve for each external field $|H_0|$. The 30° prism has virtually overcome the problem of interference fringes introduced by the 45° prism.

When the frequency and wave vector of the incident radiation match those of the magnetic excitations in the sample, there is a transfer of energy from the incident radiation to the internal modes of the antiferromagnet. This coupling is observed as a reduction in the measured reflectivity. The calculation is given in Refs. 8 and 9. The different kinds of modes excited can be identified by key features in the spectra. The bulk bands appear as broad regions with a nearly constant reduction in the measured reflectivity. The bulk bands are separated by regions in which no bulk waves can propagate. These are referred to as reststrahl regions and show up against the bulk background as regions of high reflectivity. Magnetic surface modes appear as sharp dips and generally show stronger coupling to the incident light, and therefore the measured reflectivity drops below the reflectivity of the bulk bands. The surface excitations are either damped surface resonances within the bulk bands or surface modes that lie in the reststrahl regions between the bulk bands.

A sample reflectivity spectrum for $H_0 = \pm 0.15$ T is shown in Fig. 1. Reading the spectra from the lower-frequency end, we observe a region of reduced reflectivity indicative of a broad bulk band. Just under 52.5 cm^{-1}, the reflectivity changes suddenly. Both spectra show a further reduction in the reflectivity. This is attributed to interactions with nonreciprocal surface resonances just within the bulk band. Both spectra clearly show the dramatic increase in the reflectivity due to the reststrahl band between the lower and the middle bulk bands. The middle bulk band is clearly visible in the 0.15 T spectrum as a broad dip. Comparing the $+H_0$ curve with the reverse field, we see a sharp dip just below 53 cm^{-1} that is again due to a surface resonance. Both spectra show an increase in reflectivity above 53 cm^{-1} followed by a decrease indicating a gap between the middle and the upper bulk bands.

The theoretical curves are calculated using the previously published parameters.[11] The parameters used are the exchange field $H_E = 53.3$ T, the anisotropy field $H_A = 19.7$ T and the sublattice magnetization $M_S = 560$ G. The linewidth is 0.05 cm^{-1} at $T = 1.6$ K. The gyromagnetic ratio γ has been changed very slightly from 1.05 to 1.0525 cm^{-1}/T to produce a better fit. Also included in the calculation is the anisotropic g-factor reported by Ohlmann and Tinkham.[15] The inclusion is justified since the previous data were at insufficient resolution and quality for such a small effect to be visible. However, comparison of the experimental data with theory shows a marked improvement when the known g-factor anisotropy is included. Due to some surface defects of the crystal and minor variations of the measured signal, small corrections to the experimental data have been carried out. This affects only the relative reflectivity and it should be stressed that the frequency scale remains unaltered.

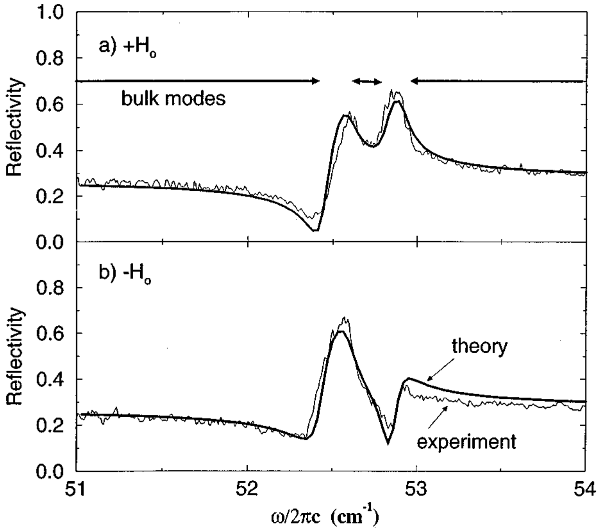

FIG. 1. Reflectivity as a function of frequency from FeF$_2$ ϕ=45°. The applied field is $H_0 = \pm 0.15$ T.

A representative ATR spectrum is shown in Fig. 2. This is obtained using the 30° Si prism for $H_0 = \pm 0.15$ T. The gap between the base of the prism and the sample is around 17 μm. Again, bulk modes can be generally identified by broad regions of reduced reflectivity, and surface modes are indi-

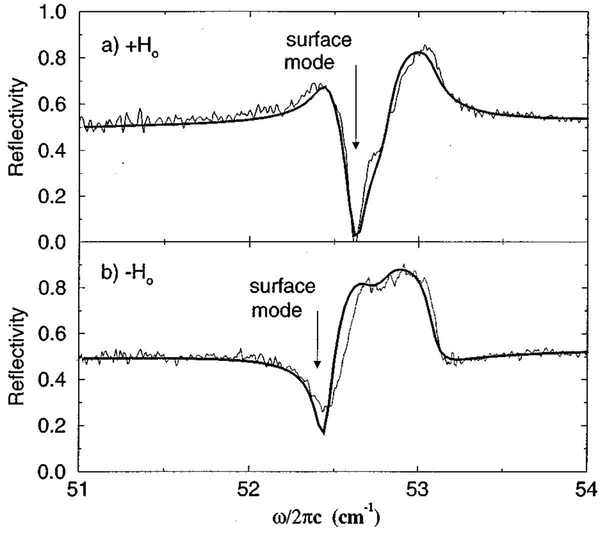

FIG. 2. ATR reflectivity as a function of frequency from FeF$_2$. ϕ=30°; the gap between the prism and the antiferromagnet is 17 μm. Note the large nonreciprocity in frequency at 52.6 cm^{-1}.

cated by sharp dips or sharp reductions in reflectivity. The agreement between experiment and theory for both the ATR and the ordinary reflectivity measurements is excellent, with even small features in the theory properly reflected in the experimental results. Additional measurements (not shown here) at 0.3 T and higher fields show equally good agreement between theory and experiment.

The nonreciprocity between positive and negative fields is very striking. At the surface mode frequency for $+H_0$(52.6 cm^{-1}), the reflectivity of the two spectra varies from nearly zero for positive field to about 80% for negative field. Similarly, at the surface mode frequency for $-H_0$, the reflectivity changes from about 70% to about 25% in reversing the applied field. This large nonreciprocity in a small field is a unique effect in the far infrared (FIR). The nonreciprocity in the reflectivity near 53.1 cm^{-1} is attributed to the surface resonance just inside the bulk band present only for $H_0 = -0.15$ T.

We obtain points for the dispersion curve in the usual manner. For example, for positive applied field, the edge of the lower bulk band is clearly marked by an increase in reflectivity around 52 cm^{-1}. The surface mode in the reststrahl band is clearly resolved from the bulk band. Reversing the field brings the surface mode to a lower frequency. It is still in the reststrahl region, but this time it is too close to the lower bulk band to be resolved due to the damping in the crystal.

The experimentally determined dispersion curves for FeF$_2$ are shown in Fig. 3. The thin solid lines mark the edges of the theoretical bulk continua, while the dashed lines indicate the theoretical surface polariton curves.[8] The experimental data are indicated in several different ways. The solid, nearly vertical, lines indicate the frequency regions of reduced reflectivity indicative of the bulk bands. Each solid spot represents an experimental measurement of the edge of a bulk band. If a band edge could not be unambiguously identified, this is indicated by solid lines which do not terminate in an experimental spot. Single spots with no line attached indicate the sharp dips in reflectivity typical of the surface modes. The uncertainty of the bulk edges is estimated to be about ±0.05 cm^{-1}. This uncertainty is simply due to the difficulty of accurately determining the bulk edges due to the damping of the crystal and does not indicate inaccuracies in the experimental spectra. The uncertainty is about ±0.02 cm^{-1} for the sharper surface modes. All of the experimental measurements lie along the predefined scan lines determined by the physical geometry of the instrument and the prisms used.

Figure 3(a) is a plot of the dispersion curve for zero field. It is reciprocal both in the bulk bands and the surface features. The plot shows two bulk bands and a surface mode for $+q$ and $-q$. Due to the damping of the crystal, the upper edge of the lower bulk band could not be accurately determined due to the presence of the strongly coupled surface mode. Despite these uncertainties, the shape of the dispersion curve is clearly defined.

The measurements with an external field are shown in Fig. 3(b). We now see three bulk bands and two gaps as predicted by theoretical calculations. Furthermore, the surface modes are clearly and dramatically nonreciprocal. The surface curve for propagation in the $-q$ direction is squashed down in frequency towards the lower bulk band, while for propagation in the opposite direction, the surface curve shows a significant increase in frequency with an increase in $+q$. Thus the group velocities are very different for propagation in opposite directions. As mentioned previously, this could be important for device applications.

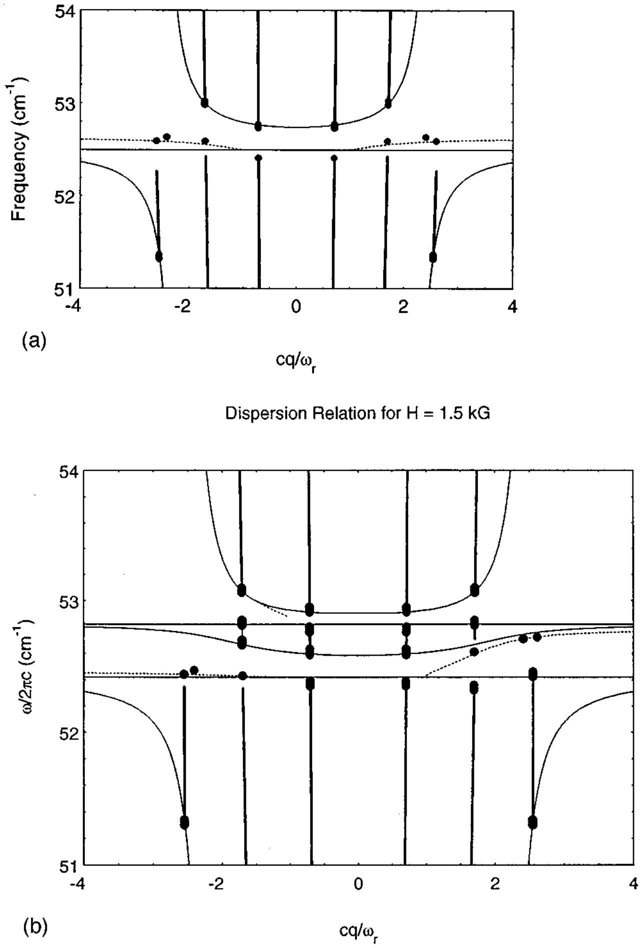

FIG. 3. Experimental dispersion curves for (a) $H_0 = 0$ and (b) $H_0 = 0.15$ T. The solid vertical lines show regions of reduced reflectivity indicating bulk bands. The edge of the bulk bands are marked by large dots indicating the uncertainty in frequency. The better defined surface modes are indicated by smaller dots. Thin solid lines give theoretical edges for bulk bands; thin dashed lines give theoretical dispersion curve for surface waves. ω_T is the antiferromagnetic resonance frequency given by $\gamma(2H_aH_e + H_a^2)^{1/2}$.

In summary, we have obtained experimentally the complete dispersion curves for bulk and surface polaritons on a uniaxial antiferromagnet. The results show excellent agreement with theoretical calculations and open the way for future studies on interface coupling of antiferromagnets with other magnetic materials.[16] The individual reflectivity spectra show a very large nonreciprocity, tunable with a small applied field.

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