Infrared studies of magnetic surface modes on antiferromagnets (invited)

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In contrast to ferromagnets, where low frequency surface excitations typically have frequencies in the 10 GHz range, surface excitations in antiferromagnets are often in the several hundred GHz to few THz range. Theoretical predictions for surface spin waves on antiferromagnets indicate that they should be highly nonreciprocal, i.e., the properties of a wave with wave vector $+k$ would be very different from those with a reversed wave vector of $-k$. Surface spin waves on antiferromagnets have recently been measured using a high resolution Fourier transform infrared spectrometer. The results show evidence of both true surface modes and surface resonances. The nonreciprocal features of the surface modes are seen in a dramatic nonreciprocal reflection. For example, the reflectivity can be 80% for one orientation, but when the incident and reflected waves are reversed the reflectivity drops to near zero. While the initial measurements were done on a bulk antiferromagnet, we also present calculations showing the results for thin films. [S0021-8979(98)36811-5]

The properties of antiferromagnets have recently received renewed attention for a variety of reasons. Many antiferromagnets are insulators and therefore have very different properties from the thoroughly studied ferromagnetic metals of Fe, Ni, and Co. For example, anisotropy fields in antiferromagnets are often in the 100 kG range compared to the 1 kG or less found in the transition metals. Also, antiferromagnets play an important role in the exchange biasing[1] of ferromagnetic films, a feature of current importance in magnetoresistive reading heads.[2] Again in contrast to ferromagnets, antiferromagnets can have long-wavelength spin excitations in the infrared (IR) frequency regime. This makes antiferromagnets of interest for signal processing in the infrared.

In this paper we report on theoretical and experimental studies of the infrared reflectivity from a bulk antiferromagnet sample of FeF_2. We concentrate in particular on the surface spin waves that propagate in this structure. In contrast to the bulk waves, the surface waves show significant nonreciprocity[3] in that reversing the wave vector can significantly change the frequency of the excitation when the system is in the presence of an external magnetic field. In a reflectivity experiment this corresponds to interchanging the incident and reflected waves, and a nonreciprocal reflectivity is also observed.

We also indicate the possibility of IR studies of thin antiferromagnetic films by theoretical calculations. In very thin ferromagnetic films, it is the surface waves which will have the lowest frequencies and which are most easily measured. This is likely to be true in antiferromagnets as well, and our initial calculations show that it should be possible to see signals from antiferromagnetic films with thicknesses in the 100–1000 Å range.

In the long-wavelength limit the reflectivity of a magnetic insulator is governed primarily by the frequency dependent permeability tensor. For the antiferromagnet, a calculation of this tensor begins with the equations of motion for the spins on the two sublattices:

$$\frac{d\mathbf{M}_1}{dt}=\gamma(\mathbf{M}_1\times\mathbf{H}_1^{\mathrm{eff}}), \tag{1}$$

and

$$\frac{d\mathbf{M}_2}{dt}=\gamma(\mathbf{M}_2\times\mathbf{H}_2^{\mathrm{eff}}). \tag{2}$$

In the above equations $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetizations on the two sublattices, γ is the gyromagnetic ration, and $\mathbf{H}^{\mathrm{eff}}$ is an effective field acting either on sublattice 1 or 2.

The effective field is composed of a number of contributions. For example the effective field acting on sublattice 1 is given by

$$\mathbf{H}_1^{\mathrm{eff}}=\mathbf{H}_1^{\mathrm{exchange}}+\mathbf{H}_1^{\mathrm{anisotropy}}+\mathbf{H}_0+\mathbf{h}_{\mathrm{dipolar}}, \tag{3}$$

with a similar expression for the effective field on sublattice 2. Here H_0 is the applied field. The exchange field is typically the largest of all the fields above with a magnitude on the order of 100–1000 kG. A key point to notice is that the exchange field acting on sublattice 1 comes primarily from the magnetization on sublattice 2, i.e.,

$$\mathbf{H}_1^{\mathrm{exchange}}=\lambda\mathbf{M}_2, \tag{4}$$

where λ is the exchange coupling constant. Even in the long-wavelength limit, the two sublattices do not have to be par-

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allel and as a result the large exchange field produced by sublattice 2 influences the motion of sublattice 1 through Eq. (1).

The large exchange field is a significant difference between the ferromagnet and the antiferromagnet and explains why antiferromagnet excitations lie in the infrared while long-wavelength ferromagnetic spin wave frequencies are in the few GHz region. In the ferromagnet—in the long wavelength limit—the exchange field is simply proportional to the magnetization:

$$\mathbf{H}^{\text{exchange}} = \lambda \mathbf{M}. \tag{5}$$

As a result the contribution of the exchange field in the equations of motion is zero since $\mathbf{M} \times \lambda \mathbf{M} = 0$. So even though the exchange field is very large, it does not influence the motion of the spins. In the antiferromagnet, however, the exchange field does play a role, and the resulting frequencies are much higher.

Using the expressions for the exchange field and any external and anisotropy fields, the coupled equations of motion for the two sublattices can be solved to relate the dipolar driving fields to the fluctuating magnetization. We assume a time dependence of $\exp(-i\omega t)$ for all the dynamic fields and obtain the frequency dependent susceptibility tensor defined through the equation

$$\mathbf{M} = \boldsymbol{\chi}(\omega)\mathbf{h}_{\text{dipolar}}. \tag{6}$$

The frequency dependent permeability is then found through the usual definition

$$\boldsymbol{\mu}(\omega) = 1 + 4\pi\boldsymbol{\chi}(\omega). \tag{7}$$

The explicit form for the permeability can be found in Ref. 4. Having found the permeability, the electromagnetic modes for the antiferromagnet may be found in the usual way.[5] One looks for wavelike solutions which satisfy Maxwell's equations inside and outside the antiferromagnet. These solutions are then matched at the surface of the antiferromagnet and this results in the dispersion relation. The reflectivity may also be calculated similarly.[6]

We note that a number of different structures and geometries have been considered theoretically in the literature. Both easy plane and uniaxial antiferromagnets have been studied, and general geometries with arbitrary directions for the applied magnetic field and for the direction of propagation have been examined. Much of this work is summarized in the review article found in Ref. 7.

We consider a geometry where the surface of the antiferromagnet lies in the xz plane. The results take a particularly simple form for a uniaxial antiferromagnet where the easy axis and the external field are both along the surface and parallel to each other (along the z axis), and the direction of propagation is perpendicular to the external field, i.e., in the xy plane. We look for electromagnetic waves with the electric field parallel to z and the magnetic field in the xy plane (s polarized). With no external field, one may find that the permeability tensor is given by

$$\boldsymbol{\mu}(\omega) = \begin{pmatrix} \mu_1 & 0 & 0 \\ 0 & \mu_1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \tag{8}$$

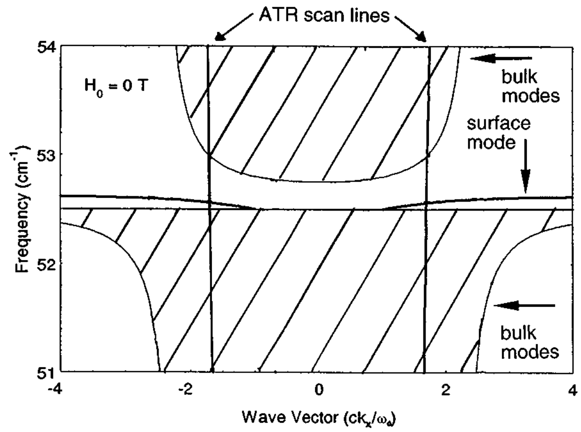

FIG. 1. Dispersion relations for bulk and surface polaritons in FeF_2. The frequency, $\omega/2\pi c$, is given in wave numbers. The applied field is zero and so both bulk and surface modes are reciprocal, i.e., $\omega(+k_x) = \omega(-k_x)$. Propagation is perpendicular to the easy axis.

where

$$\mu_1(\omega) = 1 + \frac{8\pi H_A M}{\omega_0^2 - \omega^2}. \tag{9}$$

Here ω_0 is the resonance frequency given by

$$\omega_0 = \gamma[H_A(2H_E + H_A)]^{1/2}, \tag{10}$$

and M is the saturation magnetization of one of the sublattices. In FeF_2 the anisotropy field $H_A = 197$ kG and the exchange field $H_E = 533$ kG, and $M = 0.56$ kG. With a gyromagnetic ratio of $\gamma = 0.105\ \text{cm}^{-1}$/kG this gives a resonance frequency of $52.4\ \text{cm}^{-1}$ or about 1500 GHz.

When the applied field is zero the dispersion relations have relatively simple forms. The dispersion relation for the bulk polaritons in zero field is given by the usual relation.

$$k_x^2 + k_y^2 = \epsilon\mu_1\omega^2/c^2, \tag{11}$$

where k_x is the component of the wave vector parallel to the surface and k_y is the wave vector component perpendicular to the surface. The dispersion relation for surface polaritons is given by an implicit dispersion relation

$$k_x^2 = \left(\frac{\epsilon - \mu_1}{1 - \mu_1^2}\right)\mu_1\omega^2/c^2. \tag{12}$$

The results for the bulk and surface polaritons in FeF_2 with zero applied field are shown in Fig. 1. We see two frequency regions which represent the bulk excitations. Between the bulk bands we see a surface mode which is reciprocal, i.e., the frequency does not depend on the sign of the wave vector.

A very different dispersion curve is found when there is an external magnetic field as can be seen in Fig. 2. Now there are three bulk bands. While the bulk modes are reciprocal, the surface modes are clearly nonreciprocal. For example, one mode which exists at higher frequencies for $-k_x$ has no counterpart for $+k_x$ in the same frequency range. We note that for the geometry considered here reversing the applied field is equivalent to a reversal of the propagation direction. Thus one may determine nonreciprocal propagation and reflectivity characteristics by leaving the optical setup un-

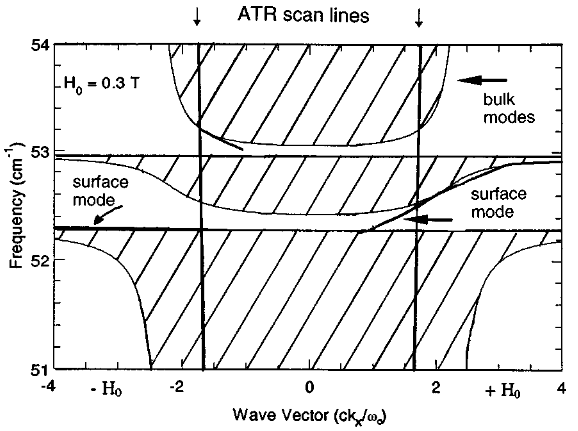

FIG. 2. Dispersion relations for bulk and surface polaritons in FeF_2 with an applied field of 3 kG. In contrast to the $H=0$ case there are now three bulk bands and the surface modes are strongly nonreciprocal.

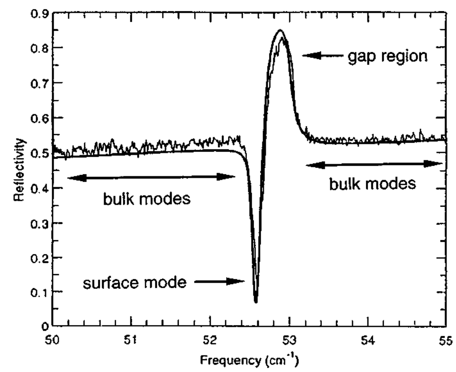

FIG. 4. Experimental and theoretical ATR reflectivity as a function of frequency for FeF_2 in zero field. The broad regions of depressed reflectivity represent losses to bulk modes. The sharp dip in reflectivity represents the loss to the surface mode. The angle of incidence in the prism is 30° and the gap between the prism and the antiferromagnet is 17 μm.

changed and simply reversing the applied field. This is a much simpler procedure experimentally and we use this method.

Since these excitations are in the infrared, it is natural to use infrared radiation as a probe. We note that this has been done in the past using a laser at a single frequency,[6,8] and also with a Fourier transform infrared (FTIR) system.[9] However, none of these experiments truly identified the surface modes. There are several reasons for this. First, the frequency gap between the bulk and the surface modes is quite small, requiring a system with high frequency resolution. Second, an ordinary reflectivity measurement is normally sensitive to bulk modes and not to surface modes.

To overcome the difficulties outlined above, we have used a specially designed FTIR system with a resolution on the order of 0.01 cm^{-1}. This requires the scanning arm of the interferometer to be about 1 m long. Additional information on the interferometer may be found in Ref. 10. In addition we use the attenuated total reflection (ATR) technique which allows the external radiation to couple to the surface modes effectively.[11] This technique is illustrated in Fig. 3. External light is incident on a Si prism with dielectric constant $\epsilon=11.57$. Because of the high index of the prism, the light is totally internally reflected at the base of the prism and the reflectivity, in the absence of a sample, would be unity. However, there is an evanescent wave below the prism base which can couple to electromagnetic modes in the sample. When this occurs, the reflectivity is reduced from unity since some of the energy is transferred to the sample.

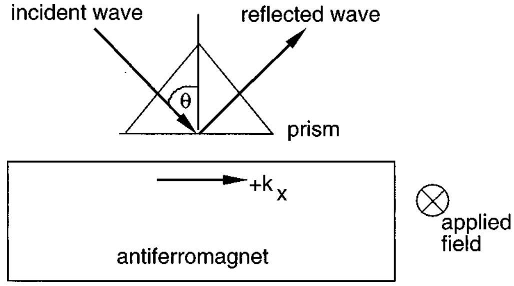

FIG. 3. ATR reflectivity geometry. Reversing the incident and reflected waves reverses the direction of k_x.

To understand the ATR curves, it is helpful to plot the dispersion curve representing the incident light in the prism on top of the magnetic polariton dispersion curves. When we write the dispersion relation in terms of the component of the wave vector parallel to the surface we obtain

$$k_x=(\omega/c)\sqrt{\epsilon}\sin\theta. \tag{13}$$

This gives the straight lines shown in Figs. 1 and 2 for positive and negative k_x. Where the dispersion curve (or scan line) of the incident light crosses the bulk or surface modes of the antiferromagnet, there can be a loss of energy from the incident wave to the modes of the antiferromagnet and a corresponding reduction in reflected intensity. Thus a broad region of depressed reflectivity corresponds to the existence of bulk bands, while a sharp dip at one particular frequency corresponds to a surface mode.

In Fig. 4 we plot the ATR reflectivity for the case of zero field. The temperature here and in Fig. 5 is 1.7 K. In this plot we see two broad regions of depressed reflectivity corresponding to the two bulk bands of Fig. 1. In between there is a sharp dip in reflectivity, corresponding to the surface mode, and then an increase in reflectivity corresponding to the gap between the bulk modes. It is clear that the experimental results are in very good agreement with both the theoretical calculations for ATR reflectivity and for the dispersion relations calculated in Fig. 1.

In Fig. 5 we again plot ATR reflectivity presently for a field of ±3 kG. From the dispersion curve in Fig. 2, we expect a total of three regions of reduced reflectivity corresponding to the three bulk bands. In addition we expect sharper dips representing the surface modes. These features are all present in Fig. 5. A key feature to note is that the surface modes appear at different frequencies depending on the sign of applied magnetic field. This clearly demonstrates the expected nonreciprocity of the surface modes. It may be possible to exploit this nonreciprocity for signal processing in the infrared. For example the ATR reflectivity near 52.5 cm^{-1} is close to zero for positive field. In contrast, the wave traveling in the reversed direction (equivalent to reversing the applied field) has a reflectivity of close to 80%. Thus this system might be used as the basis for a nonreciprocal isolator in the infrared.

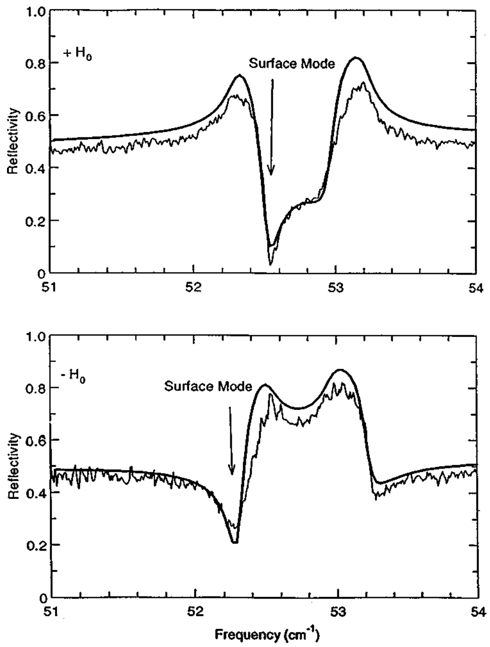

FIG. 5. ATR reflectivity as a function of frequency for FeF_2 with an applied field of 3 kG. The angle of incidence in the prism is 30° and the gap between the prism and the antiferromagnet is 17 μm. Note the large nonreciprocity in frequency at 52.5 cm^{-1}. The smooth curves are the theoretical results and the thinner, jagged lines are the experimental results.

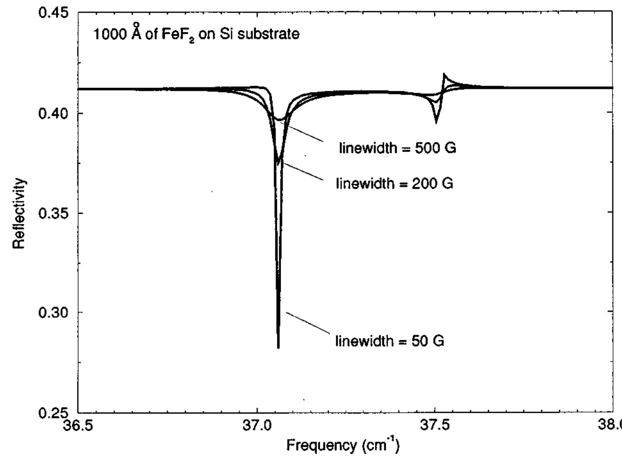

FIG. 6. Ordinary reflectivity from a thin FeF_2 film on a Si substrate for different linewidths. The applied field is 1.5 kG, and the angle of incidence is 45°.

By doing measurements with different angles of incidence inside the prism, one can trace out the behavior of the edges of the bulk band and the position of the surface mode. In addition one can also use ordinary reflectivity. The results[12,13] are in excellent agreement with the theoretical calculations shown in Figs. 1 and 2. Other recent reflectivity studies on FeF_2 in a different geometry also show excellent correspondence between theory and experiment.[14]

The properties of thin antiferromagnetic films, or of antiferromagnetic films coupled to ferromagnets in an exchange biasing configuration may be quite different from those of bulk antiferromagnets. It is therefore of interest to see whether the infrared studies of bulk materials can be extended to thin films. In Fig. 6 we plot the ordinary infrared reflectivity (not ATR reflectivity) seen from a 1000 Å FeF_2 film on a Si substrate. If the linewidth is on the order of 500 G, the signal is small. However, if the linewidth is only 50 G the signal is quite substantial with a change in reflectivity on the order of 15%. Linewidths in antiferromagnets can vary substantially depending on the quality of the crystal. Our bulk sample showed a linewidth on the order of 400–500 G at low temperatures. However some samples have been reported with linewidths an order of magnitude or more lower.[15]

The current FTIR system is sensitive to about a 1% change in reflectivity. If an antiferromagnetic film has a linewidth on the order of 50 G, this would lead to the conclusion that one should be able to observe a signal from an antiferromagnetic film with a thickness of about 100 Å. Improvements in the signal to noise ratio might therefore easily allow much thinner films to be studied. Alternatively, one could form a superlattice where the unit cell contained, say, a 100 Å antiferromagnetic film. Ten repeats of this structure would give 1000 Å of antiferromagnet. In this case a signal should be observable even with the higher linewidths.

What could be expected from studies of thin antiferromagnetic films? We know that in ferromagnets surface anisotropy fields or interfacial exchange fields can substantially change the surface spin wave frequency.[16] Measurements of this frequency can then be used to determine these surface and interface parameters. Initial calculations[17] show that the spin wave modes in antiferromagnets will also be sensitive to surface and interfacial properties. Thus IR probes, either with the FTIR system or with a narrow linewidth laser of thin antiferromagnetic films may be helpful in understanding the exchange biasing effect.

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