Nonreciprocal reflection by magnons in FeF_2: A high-resolution study

D. E. Brown, T. Dumelow, and T. J. Parker
Department of Physics, University of Essex, Colchester CO4 3SQ, United Kingdom

Kamsul Abraha
Department of Physics, University of Essex, Colchester CO4 3SQ, United Kingdom
and Jurusan Fisika, Universitas Gadjah Mada, Yogyakarta 55281, Indonesia

D. R. Tilley
Department of Physics, University of Essex, Colchester CO4 3SQ, United Kingdom
(Received 15 July 1993)

We present oblique-incidence frequency scans of an antiferromagnetic-resonance reflectivity spectrum in the presence of an applied magnetic field H_0. Results are shown for the c axis of an FeF_2 single crystal both parallel and perpendicular to H_0, which is normal to the plane of incidence. In both cases the s-polarization spectra show nonreciprocal behavior with respect to H_0.

Long-wavelength properties of ordered magnetic systems involve coupling of spins via the electromagnetic field as well as via exchange interactions. Quantities involving both a surface and a magnetic field $\mathbf{H}_0$ may display the property of nonreciprocity;[1] i.e., they change when $\mathbf{H}_0$ is reversed.

Nonreciprocal propagation has been thoroughly studied in the magnetostatic limit $k \gg \omega/c$ by means of Brillouin scattering from ferromagnetic surfaces and films.[2] For the retarded region $k \sim \omega/c$ where the full form of Maxwell's equations must be applied, however, there is a considerable theoretical literature[3–5] but very little experimental work. This is because the spectral features associated with the antiferromagnetic resonance are extremely narrow, of the order of 0.01 cm^{-1}, compared with 1 cm^{-1} typical of much far-infrared spectroscopy. In addition, a magnetic field and variable temperature are required for substantial investigations.

Apart from early zero-field reflectivity studies on the antiferromagnets FeF_2 and CoF_2,[6,7] there is to our knowledge just one relevant paper.[8] This reports oblique-incidence reflectivity measurements off the surface of the uniaxial antiferromagnet MnF_2. The radiation source was at fixed frequency and the reflectivity R was measured as a magnetic-field scan through the antiferromagnetic-resonance frequency. The results confirm that the surface reflectivity R is nonreciprocal in the sense that $R(-H_0) \neq R(H_0)$. However, due to the restriction of their instrument to a single frequency, Remer *et al.*[8] were able to observe only one of the Zeeman-split lines. Up till now, however, their paper has represented the only experimental evidence we know of for this form of nonreciprocity.

The purpose of this paper is to describe an investigation of the magnetic-field dependence of the reflectivity from an antiferromagnet making use of frequency scans through the resonances. The present studies are of FeF_2, which is a uniaxial antiferromagnet, isomorphic to MnF_2, with the resonance frequency having a value near 50 cm^{-1}. For the c axis along the magnetic field, we show unequivocal evidence of nonreciprocity and track the resonance frequency and linewidth up to $T \sim 0.5 T_N$. We have also measured the reflectivity with the c axis transverse to the field. In this case, the sublattice magnetizations are canted away from the easy axis toward the field, and resonant reflections appear in both s and p polarization.

A specialized far-infrared Fourier-transform spectrometer has been constructed for these measurements. Further details are given elsewhere.[9] The instrument has two optical channels, one for the far-infrared beam of 50 mm diameter, and the other for a single-mode He-Ne laser beam of 0.5 mm diameter passing along the optical axis. The moving mirror is scanned at constant speed over a distance up to 1 m, giving a resolution limit of 10^{-2} cm^{-1}. The laser fringes are scanned continuously and used to monitor the optical path difference. The far-infrared beam is modulated by vibrating the "fixed" interferometer mirror and detected with a phase-sensitive detector. This arrangement combines the advantages of phase-sensitive detection for the infrared beam and precise measurement of the optical path difference using the laser.

The output beam from the interferometer is focused, at angle of incidence $\phi = 45°$ and plane of incidence horizontal, on the sample located between the poles of a 7-T vertical-field magnet. The reflected beam is subsequently focused onto a liquid-He-cooled Si bolometer.

The surface of the crystal is taken as the x-z plane with z in the direction of H_0. For the first set of measurements the crystal is oriented with the c axis vertical. The magnetic permeability tensor $\overleftrightarrow{\mu}$ then has components[10]

$$\mu_{xx} = \mu_{yy} = 1 + \gamma^2 H_a M_s \left[\frac{1}{\omega_r^2 - (\omega + i\Gamma - \gamma H_0)^2} + \frac{1}{\omega_r^2 - (\omega + i\Gamma + \gamma H_0)^2} \right] \quad (1)$$

and

$$\mu_{xy} = -\mu_{yx} = i\gamma^2 H_a M_s \left[\frac{1}{\omega_r^2 - (\omega + i\Gamma - \gamma H_0)^2} - \frac{1}{\omega_r^2 - (\omega + i\Gamma + \gamma H_0)^2} \right] . \quad (2)$$

All other components are zero, except $\mu_{zz} = 1$. H_a is the anisotropy field, M_s the sublattice magnetization, $\omega_r^2 = \gamma^2(2H_a H_e + H_a^2)$ the antiferromagnetic-resonance frequency, γ the gyromagnetic ratio, and Γ the damping. For the magnetic-resonance frequency to appear in the reflectivity, the optical H field must lie in the x-y plane, i.e., the magnetic mode is seen in s polarization. The reflectivity is $R = |(1-\rho)/(1+\rho)|^2$, where

$$\rho = \frac{(\varepsilon\mu_v - \varepsilon_m \sin^2\phi)^{1/2} - (\mu_{xy}/\mu_{xx})\varepsilon_m^{1/2}\sin\phi}{\varepsilon_m^{1/2}\mu_v \cos\phi} . \quad (3)$$

Here ε is the dielectric constant of FeF_2, $\varepsilon_m(=1)$ the dielectric constant of the medium of incidence, and $\mu_v = \mu_{xx} + \mu_{xy}^2/\mu_{xx}$ is the Voigt permeability. The nonreciprocity of R, $R(-H_0) \neq R(H_0)$, is evident from (3) since it contains the product $\mu_{xy}\sin\phi$, which is odd in both H_0 and ϕ.

We first show results for $H_0 = 0$, in which case $\overleftrightarrow{\mu}$ is diagonal. Figure 1 shows the spectra for a range of temperatures. They are dominated by a single strong magnetic resonance line at about 52 cm^{-1}. This broadens and decreases in frequency with temperature. The signature away from the main resonance line is due to reflections from the front and back surfaces of the sample. The background level of R is determined by ε. However, R is calculated from the ratio of the intensities obtained from reflection first off the sample and then off an aluminized mirror. Since two spectra, obtained at different times, are needed, the overall level of the calculated reflectivity is sensitive to small variations in experimental conditions. Hence ε is not accurately determined.

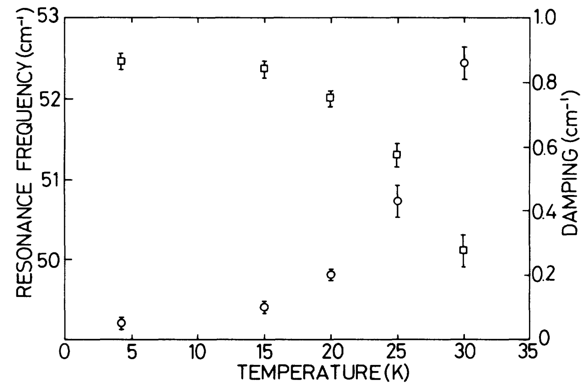

FIG. 2. Temperature dependence of the resonance frequency (□, left-hand axis) and damping (○, right-hand axis) determined by a fit of theory to the data of Fig. 1.

A least-squares fit to the spectra was used to determine the temperature dependence of ω_r and Γ shown in Fig. 2. Our technique is effectively a refined version of antiferromagnetic resonance (AFMR) and the results in Fig. 2 are similar to those obtained by Ohlmann and Tinkham[11] in their AFMR study of FeF_2. The temperature dependence of ω_r is also similar to that observed in Raman scattering.[12]

The effect of applying a field at 4.2 K is shown in Fig. 3; the zero-field line Zeeman splits into two resonances.

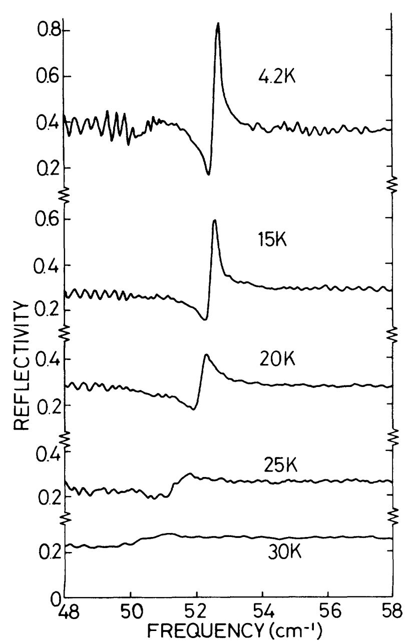

FIG. 1. Frequency scans of 45° reflectivity in s polarization through magnetic resonance in zero field for a range of temperatures. The crystal c axis is along the magnetic field, i.e., vertical. Resolution: 0.25 cm^{-1} at 30 K, 0.125 cm^{-1} at other temperatures.

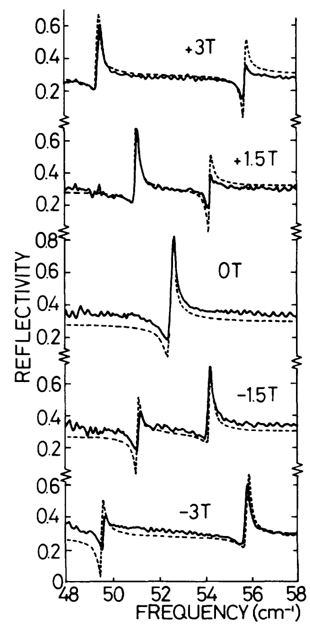

FIG. 3. Magnetic-field splitting of the 4.2-K spectrum in the geometry of Fig. 1. —— experiment, – – – theory. Resolution: 0.125 cm^{-1}. The theoretical curves were drawn with $\varepsilon = 5.5$, $\omega_r = 52.45$ cm^{-1}, as determined from the 0-T spectrum, and $\Gamma = 0.05$ cm^{-1}.

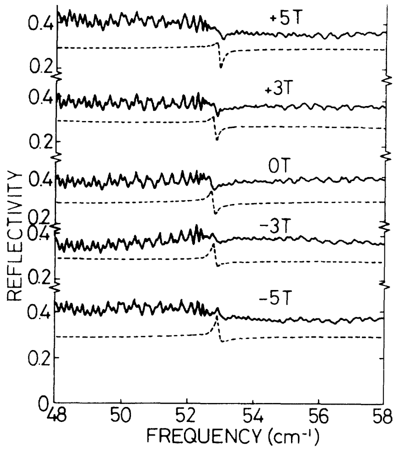

FIG. 4. Spectra at various fields in s polarization for the crystal c axis transverse to the magnetic field, i.e., horizontal. —— experiment, — — — theory. Resolution: 0.063 cm^{-1}, fitting parameters as for Fig. 3.

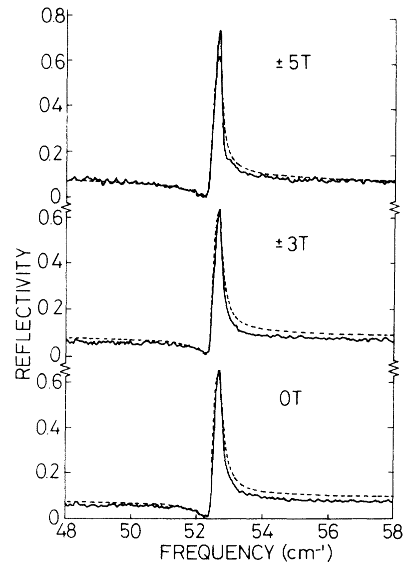

FIG. 5. Spectra at various fields in p polarization for the crystal c axis horizontal. —— experiment, — — — theory. Resolution: 0.063 cm^{-1}, fitting parameters as for Fig. 3.

The nonreciprocity between the two is seen in a very clean and striking fashion. The parameters for the computed curves were chosen to given the closest resemblance to the experimental spectra. The field dependence of the line splitting is linear as follows from (1) and (2), and the field gradient is given by the standard value of γ. We have also measured spectra in p polarization for this geometry and as expected they are featureless.

For the second set of measurements the crystal was turned through 90° so that the c axis was horizontal. In this case, as pointed out by Almeida and Mills,[13] the sublattice magnetizations cant through an angle α toward H_0 in the equilibrium configuration so that the permeability tensor takes a different form. Thus, rotating an antiferromagnet is more complicated than rotating a uniaxial dielectric, since in the latter case one would be dealing with rotation of a *fixed* permittivity tensor. The permeability tensor components μ_{xx}, μ_{xy}, μ_{yx}, and μ_{yy} now contain a single resonance $\omega_\perp^2=(\omega_r^2\cos^2\alpha+2\gamma^2H_0H_e\sin\alpha)$. μ_{zz} contains a separate resonance $\omega_\parallel^2\equiv\omega_r^2\cos^2\alpha$; all other components are zero. The explicit expressions are given in Ref. 13.

The resonance at $\omega_\parallel$ is seen in p polarization whilst the resonance at $\omega_\perp$ is seen in s polarization as before. The expressions quoted mean that $\omega_\parallel$ decreases and $\omega_\perp$ increases with H_0, the shifts being proportional to H_0^2 for accessible fields. Numerically, $\delta\omega_\parallel=-(1.6\times10^{-3})H_0^2$ and $\delta\omega_\perp=(7.2\times10^{-3})H_0^2$ for H_0 in T. The reflectivity for each polarization can be calculated from the permeability expressions as before. Reflectivity spectra at 4.2 K for a range of applied fields are shown in Figs. 4 and 5. The background value is smaller for p than for s polarization as is generally the case for reflection off a dielectric material.

The s-polarization resonances (Fig. 4) are relatively weak. This is because the diagonal component μ_{yy} is the only term in $\vec{\mu}$ coupling in s polarization that is nonvanishing for $H_0=0$; the other relevant terms are small for accessible values of H_0. However, due to refraction the component H_y of the ir H field is small. The upward shift $\delta\omega_\perp$ with increasing $|H_0|$ can be seen as a corresponding shift in the resonant spectral feature. Although $\delta\omega_\perp$ is the same for $\pm H_0$, the shape of the feature is nonreciprocal.

In p polarization the ir H field is vertical both outside and inside the sample. Consequently, it is almost transverse to both spin-sublattice directions and couples to the large resonant term in μ_{zz}. As seen in Fig. 5, the magnetic feature is therefore much larger than in s polarization. The downward shift $\delta\omega_\parallel$ with $|H_0|$ is very small, however, so that the shift of the resonant feature is only just discernible. In this case the whole spectrum is invariant to the sign of H_0.

In this paper we have presented experimental results for the retarded regime of surface magnetic interactions which demonstrate very clearly nonreciprocal behavior in a magnetic field. FeF_2 was chosen for our first investigation since it is a well-understood antiferromagnet with a Neel temperature and resonance frequency easily accessible with our instrument. The predicted nonreciprocity of the reflectivity is very clearly displayed in Fig. 3, and the results are extended for $\theta=90°$ in Figs. 4 and 5. These results establish a powerful technique for investigating magnetic materials, which we shall now apply to more complicated antiferromagnets, ferrimagnets, and, in due course, to magnetic superlattices.

We have benefited from helpful discussions with Nilson Almeida, Bob Camley, Mike Cottam, Stephen Smith, and Bob Stamps. K. A. is supported by Proyek PS2PT DEPDIKBUD Indonesia and D. E. B. by SERC. This work is supported by SERC through Grant GR/G/54139.

[1]R. E. Camley, Surf. Sci. Rep. **7**, 103 (1987).
[2]M. G. Cottam and D. J. Lockwood, *Light Scattering from Magnetic Systems* (Wiley, New York, 1986).
[3]D. R. Tilley, in *Linear and Nonlinear Spin Waves in Magnetic Films and Superlattices,* edited by M. G. Cottam and A. Slavin (World Scientific, Singapore, in press), Chap. 4.
[4]R. E. Camley and D. L. Mills, Phys. Rev. B **26**, 1280 (1982).
[5]N. S. Almeida and D. R. Tilley, Solid State Commun. **73**, 23 (1990).
[6]K. M. Häussler, A. Lehmeyer, and L. Merten, Phys. Status Solidi B **111**, 513 (1982).
[7]K. M. Häussler, J. Brandmüller, L. Merten, H. Finsterhölzl, and A. Lehmeyer, Phys. Status Solidi B **117**, 225 (1983).
[8]L. Remer, B. Lüthi, H. Sauer, R. Geick, and R. E. Camley, Phys. Rev. Lett. **56**, 2752 (1986).
[9]T. Dumelow, D. E. Brown, and T. J. Parker, in *Eighteenth International Conference on Infrared and Millimeter Waves,* edited by J. R. Birch and T. J. Parker, SPIE Proc. Vol. 2104 (SPIE, Bellingham, WA, 1993), p. 633.
[10]D. L. Mills and E. Burstein, Rep. Prog. Phys. **37**, 817 (1974).
[11]R. C. Ohlmann and M. Tinkham, Phys. Rev. **123**, 425 (1961).
[12]D. J. Lockwood, M. G. Cottam, V. C. Y. So, and R. S. Katiyar, J. Phys. C **17**, 6009 (1984).
[13]N. S. Almeida and D. L. Mills, Phys. Rev. B **37**, 3400 (1988).