Observation of the high frequency spin wave branch and the uniform precession mode in $FeBO_3$ by Brillouin scattering[a)]

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Brillouin scattering has been used to observe the two spin wave branches of the weak ferromanget $FeBO_3$. The high frequency, quasiantiferromagnetic branch is seen by this method for the first time. The temperature dependence of this branch is measured and compared with previous FMR measurements. An asymmetry is found between the Stokes and anti-Stokes scattering intensities for this mode which is dependent on the incident polarization. This is attributed to an interference between linear and quadratic magneto-optic effects. Scattering from the low frequency, quasiferromagnetic branch at very small wave vectors has also been measured. This was done using a forward scattering configuration.

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INTRODUCTION

Iron borate, $FeBO_3$, a canted antiferromagnet has been investigated rather intensively during the last ten years because it exhibits a number of interesting magnetic, optical and magnetoelastic properties. A review of the literature on $FeBO_3$ up to 1981 is given in Ref. [1]. The most striking features of $FeBO_3$ are its Neél temperature above room temperature (T_N = 348 K) and its optical transparency in the green spectral range. $FeBO_3$ is therefore an ideal material for magneto-optical investigations and especially for inelastic light scattering studies [2,3].

The spin wave spectrum of $FeBO_3$ consists of two branches which are nondegenerate at the center and degenerate at the edge of the Brillouin zone. The low energy spin wave branch has been studied by Brillouin scattering [2,3]. The high energy branch has been measured by far infrared absorption [1,4] and ferromagnetic resonance (FMR) in the mm wave region [5,6]. The present paper complements these earlier investigations. The Brillouin scattering spectrum of the high energy magnon branch has been seen for the first time. Furthermore, the low energy branch has been measured at very small wavevectors ($\sim 10^3$ cm^{-1}) using a forward scattering configuration.

The intensity of the light scattered from the upper spin wave branch is two orders of magnitude smaller than the intensity scattered from the lower branch. This is very similar to recent findings in the Raman scattering from orthoferrites by White et al. [7]. It can be qualitatively explained by analyzing the precession of the sublattice magnetizations of the two spin wave branches taking into account the linear and quadratic magnetooptical effects that contribute to the light scattering cross section.

THEORY

$FeBO_3$ has a rhombohedral (calcite) crystal structure with space group $R\bar{3}c$. The magnetic Fe^{3+}-ions are octahedrally coordinated by 6 O^{2-}-ions. The magnetic order of the spins is predominately antiferromagnetic, but the two magnetic sublattices, with magnetizations $\bar{M}_1$ and $\bar{M}_2$ as in Fig. 1, are slightly canted due to antisymmetric exchange. These moments, as well as the weak resultant magnetization $\bar{M}_s$ are strongly confined to the plane perpendicular to the [111] c-axis due to easy plane anisotropy.

The spin wave spectrum of $FeBO_3$ consists of two magnon branches which are degenerate for large values of the wavevector k. For k=0 one finds two uniform precession modes, one low lying "quasi-ferromagnetic" mode,

$$(\omega_1/\gamma)^2 = 2H_E(H_a+H_{me}) + H_D(H+4\pi M_s(N_y-N_x)), \quad (1a)$$

and one high frequency "quasi-antiferromagnetic" mode,

$$(\omega_2/\gamma)^2 = 2H_EH_A + H_D^2 . \quad (1b)$$

Here, γ is the gyrotropic ratio ($\gamma/2\pi$ = 2.8 GHz/kOe), H_E, H_D, H_{me} and H are the symmetric and antisymmetric exchange, magnetoelastic interaction, and applied fields, respectively. H_a and H_A are the in-plane and out-of-plane anisotropy fields, respectively. N_x and N_y are demagnetization factors. The approximations $H_E >> H_D$ and $H_A >> H_a$ were used to obtain Eq. (1a,b).

The spin precession in the two modes is illustrated in Fig. 1. For the ω_1-mode, the precessions of $\bar{M}_1$ and $\bar{M}_2$ have an ellipticity,

$$|\bar{M}_x/\bar{M}_z| = 2\gamma H_E/\omega_1 \approx H_E/(HH_D)^{1/2} ,$$

with the major axis nearly parallel to the field axis (x-direction). The ellipticity of $\bar{M}_s$ is given by

$$|\bar{M}_{sy}/\bar{M}_{sz}| = \gamma H_D/\omega_1 \approx (H_D/H)^{1/2} .$$

The precession ellipse of the ω_2 mode has its major axis along the c-axis; with

$$|\bar{M}_z/\bar{M}_x| = 2\gamma H_E/\omega_2 \approx 2\, H_E/(2H_EH_A + H_D^2)^{1/2}$$

In this approximation, the magnetization $\bar{M}_s$ has only an x-component which varies in length at the precession frequency as indicated in Fig. 1. Using room temperature values $H_E \approx 1.6\times10^3$ kOe, H_A = 3 kOe, and H_D = 60 kOe, the above ellipticities range between 10 and 100 for moderate fields below 1 kOe.

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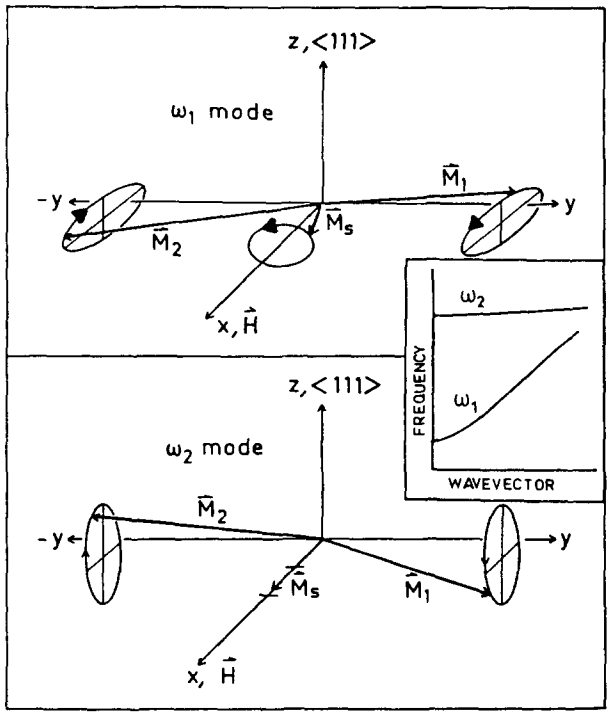

Fig. 1. Spin precession of the two spin wave modes of $FeBO_3$ from Ref. 1. Inset: schematic spin wave dispersions for small wavevectors.

The spin wave dispersions for low, but nonzero k-values are given by

$$(\omega_1(k)/\gamma)^2 = 2H_E(H_a+H_{me}) + H_D(H+4\pi M_s \cdot \cdot \sin^2\theta_k \cos^2\phi_k) + (2\pi/\gamma)^2 E(\theta_k,\phi_k)k^2 \quad (2a)$$

$$(\omega_2(k)/\gamma)^2 = 2H_E H_A + H_D^2 + (2\pi/\gamma)^2 E(\theta_k,\phi_k)k^2 \quad (2b)$$

where θ_k,ϕ_k are the polar and azimuthal angles of k and $E(\theta_k,\phi_k)$ is an exchange parameter of the order of 10^{-8} GHz^2 cm^2. The two spin wave branches are drawn schematically in the inset of Fig. 1.

EXPERIMENT

The Brillouin scattering experiments were performed in both backscattering and forward scattering configurations in order to probe excitations with a range of wavevectors. The green argon-laser line at 514.5 nm and a power level of about 20 mW was used. The scattered light was analyzed with a recently developed multipass tandem Fabry-Perot interferometer (FPI) [8], in which the light is passed four times through the first and two times through the second interferometer. An overall peak transmission of 10 percent and a contrast of about 10^{12} was achieved.

Figure 2 shows examples of spectra for a 1 mm thick platelet with the c-axis in-plane. Figure 2a is a logarithmic plot over a free spectral range of 375 GHz or 12.5 cm^{-1}, for H parallel to k and the plate normal (x-direction in Fig. 1). The phonon and magnon lines are superimposed on a wide central mode type background which is close to exponential in shape. The low energy magnon at about 60 GHz and the high energy magnon at 260 GHz are labeled M1 and M2, respectively. The (M1) lines are the M1 lines in the neighboring orders, suppressed by a factor of 100 due to the tandem operation.

Figure 2b shows the spectrum in the neighborhood of M1 taken with higher resolution and for a different magnetic field direction. The spectrum is similar to one shown in Ref. [2] with three acoustic phonons and the M1 magnon peaks. One interesting new feature is the M1F peak. This is the M1 magnon in forward scattering reflected at the back surface of the crystal plate. The magnetic field dependence of the M1

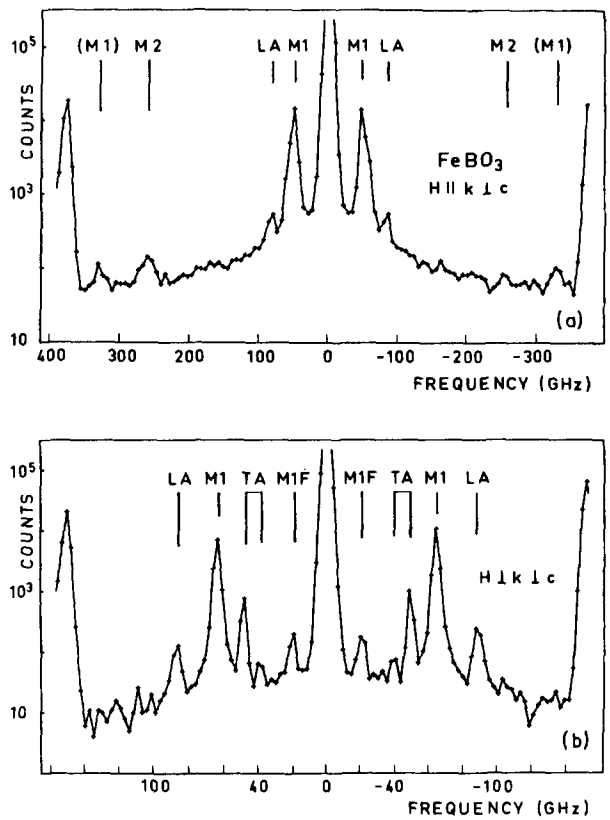

Fig. 2. Representative 180° backscattering Brillouin spectra of $FeBO_3$ at H = 760 Oe for a free spectral range of (a) 375 GHz and (b) 150 GHz. For (a), the laser power was 25 mW and the sample temperature was approximately 313 K. For (b), the laser power was 15 mW and the sample temperature was approximately 306 K.

and the M1F peak are in accordance with previous results and with the theory. The wavevector in backscattering is given by

$$|k| = (n_o+n_e)2\pi/\lambda_o = 5.4\times10^5 \text{ cm}^{-1} .$$

For forward scattering

$$|k| = (n_o-n_e)2\pi/\lambda_o = 7.0\times10^3 \text{ cm}^{-1} .$$

Here, n_o and n_e are the ordinary and extraordinary refractive indices, respectively. A forward scattering configuration with $\bar{k}$ parallel to the c-axis reduces $|k|$ to even smaller values because ordinary and extraordinary refractive indices are then equal. In this configuration, therefore, one is essentially looking at the thermally excited uniform precession magnon. For H = 54 Oe, a frequency of 6.1 GHz was observed which is close to the 6 GHz value found by conventional FMR [3].

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DISCUSSION

The temperature dependence of the high frequency magnon branch, 300-340 K, is shown in Fig. 3. The temperature was calibrated from the well known temperature dependence of the low frequency branch [2]. The solid curve labeled ν_2 is based on the FMR data of Velikov et al. [5] in the mm wave regime. The agreement is fairly good, but the Brillouin frequencies are slightly lower than the FMR values. Measurements should be extended to higher and lower temperatures in order to investigate this small discrepancy.

The intensity of the M2 line is 2 orders of magnitude smaller than that of the M1 line as evident from the logarithmic plot in Fig. 1a. The anti-Stokes M2 line is a factor of 3 stronger than the Stokes line. Rotating the polarization of the incident light by 90 degrees shifts the stronger line to the Stokes side. Such behavior is well known in light scattering from magnons. It is attributed to an interference of linear and quadratic magnetooptic effects in the scattering cross section [9].

Recently White et al [7] have given a detailed analysis of magnon spectra of $YFeO_3$ by Raman scattering, in which similar effects were observed. This analysis can also be applied to $FeBO_3$, keeping in mind the differences in crystal structure and magnetic anisotropy.

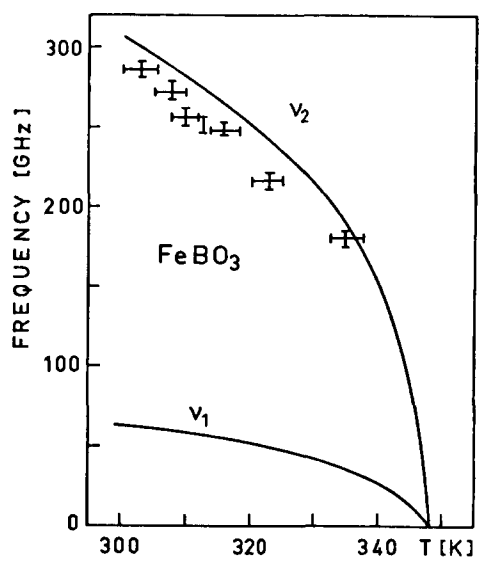

Fig. 3. Temperature dependence of the high frequency spin wave branch of $FeBO_3$. Solid lines indicate data from Ref. 2 (ν_1) and Ref. 5 (ν_2).

The scattered intensity is proportional to the square of a dynamic dielectric tensor

$$\Delta\varepsilon_{ik} = K_{ik\ell}M_\ell \pm G_{ik\ell m}M_\ell M_m^o \qquad (3)$$

where K and G are the real linear and quadratic magnetooptic tensor elements which are closely related to Faraday rotation and magnetic linear birefringence, respectively. The M_ℓ are components of the dynamic magnetization. The M_m^o are components of the static sublattice magnetization and the ± refer to Stokes and anti-Stokes scattering, respectively.

Taking into account the canted spin configuration and the ellipticities of the precession in the two modes, White et al. argue that the ellipticity of the ω_2 mode increases the difference between K- and G-contributions, while for the ω_1-mode, the ellipticity suppresses this interference. Therefore, the Stokes-anti-Stokes intensity difference will show up only for the M2 mode. This is in qualitative agreement with our observations in $FeBO_3$.

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