Brillouin scattering from thermal magnons in a thin Co film

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We report a room-temperature Brillouin scattering study of surface and standing spin-wave modes in a 100-nm film of polycrystalline hcp cobalt. The film was prepared by vapor deposition of Co metal onto a single-crystal sapphire substrate with the crystalline c axis in the substrate plane. X-ray measurements show the film is predominantly aligned with the [001] axis normal to the film plane. Measurements were made with a five-pass plus four-pass tandem vernier Fabry-Perot interferometer with an instrumental full width of 700 MHz. The magnetic field H_0, $150 \leq H_0 \leq 3500$ Oe, was applied in the plane of the film, perpendicular to the scattering plane. Fitting the theoretical expressions for surface and standing spin-wave dispersion relations to the experimentally determined frequency shift versus applied field gives values for the gyromagnetic ratio and saturation magnetization in satisfactory agreement with literature values obtained from the application of other experimental techniques. However, the exchange stiffness constant D obtained from analysis of the light scattering measurements is significantly smaller than that deduced from both neutron scattering and magnetization measurements.

INTRODUCTION

The development of high contrast, multipass Fabry-Perot interferometers has made backscattering measurements in opaque materials experimentally accessible.[1,2] Consequently, both the experimental technique and the theory of Brillouin scattering in metals have undergone rapid development in recent years. One particularly fertile area of research in Brillouin scattering spectroscopy is the investigation of thermal acoustic magnetic excitations (magnons) in thin ferromagnetic metallic films.[3]

In a conducting film, where the optical skin depth of the specimen δ is small compared to the free-space wavelength of the incident radiation λ, i.e., $\delta \ll \lambda$, the components of the wave vector of the scattered and incident light *normal* to the film plane are not well defined within the specimen. Therefore, only momentum components parallel to the sample surface are conserved in the scattering process,[4] and the selection rules are given by

$$k_s'' = k_i'' + Q'' , \tag{1}$$

$$\omega_s = \omega_i + \Omega . \tag{2}$$

Here k_s'', k_i'', Q'' are the projections of the wave vector of the scattered light, incident light, and magnon, parallel to the sample surface, and ω_s, ω_i, and Ω are the respective quasiparticle energies expressed as angular frequencies. All spin-wave excitations (surface and bulk modes) characterized by the same value of Q'' scatter the incident radiation in the same final direction. Consequently, a single measurement can provide information on a large number of spin-wave excitations.

For a homogeneous thin film with the magnetic field $\vec{H}_0$, applied perpendicular to the scattering plane and in the plane of the sample, the bulk magnon dispersion is given by[5,6]

$$\omega_b = \gamma[(H_0 + DQ^2)(H_0 + DQ^2 + 4\pi M_b)]^{1/2} ,$$
$$Q^2 = Q_{\parallel}^2 + Q_{\perp}^2 , \tag{3}$$

where ω_b is the magnon angular frequency, γ is the gyromagnetic ratio, D is the exchange stiffness, M_b is the *bulk* saturation magnetization and $Q_{\parallel}, Q_{\perp}$ are the components of the magnon wave vector parallel and perpendicular to the sample plane, respectively. In a film, $Q_{\perp}$ can only take on discrete values, i.e., $Q_{\perp} = n\pi/L$, where L is the sample thickness and $n = 0, 1, 2, \ldots$. Therefore, one can experimentally tune the energy splitting between modes, e.g., ω_n and ω_{n+1}, simply by altering the sample thickness. This can be readily seen from Eq. (3) by comparing the squared frequency shifts of the $(n+1)$ and n modes, i.e., $\omega_{n+1}^2 - \omega_n^2$, which in the limit $Q_{\parallel} = 0$ and $DQ^2 \ll 4\pi M_b$ gives

$$\omega_{n+1}^2 - \omega_n^2 = \gamma^2(2n+1)(2H_0 + 4\pi M_b)\frac{D\pi^2}{L^2} . \tag{4}$$

Experimental resolution of these discrete standing spin-wave modes depends in detail on the fundamental constants [Eq. (4)], magnon lifetime, spectrometer resolution, etc. For our ferromagnetic films with $L \leq 250$ nm the modes are well separated (by a few Ghz) for moderate applied fields.

In the limit that $Q_{\perp} = 0$, the surface magnon dispersion is given by[5,6]

$$\omega_s = \gamma(H_0 + \xi DQ^2 + 2\pi M_s) , \tag{5}$$

where ξ equals 2 from analytic model theory[6] and 1.76 from numerical calculation,[7] and M_s is the *surface* saturation magnetization. We distinguish between M_s and M_b because experiments show that these quantities are a function of sample condition.[1]

In the following we report a Brillouin scattering study of thermal acoustic magnons in a polycrystalline thin film of hcp cobalt with a nominal thickness $L=100$ nm. In addition to the surface mode, discrete standing spin-wave modes characterized by $Q_\perp = n\pi/L$ with $1 \leq n \leq 4$ were observed. The frequency shifts, for all modes, were measured in applied fields H_0 for $150 \leq H_0 \leq 3500$ Oe. Best agreement fits of model theory expressions for surface and bulk magnon dispersion relations, Eqs. (5) and (3), to the measured frequency shift yield values of γ, M_s, and M_b in good agreement with values determined by other experimental techniques. However, the D value extracted from these measurements is significantly smaller than that determined by other experimental methods.

EXPERIMENT

The sample was prepared by vacuum deposition of Co metal onto single-crystal sapphire with the crystalline c axis in the substrate plane. The film thickness, as determined from a calibrated quartz-crystal deposition monitor, was 100 ± 10 nm. X-ray measurements with the scattering vector normal to the film plane (θ-2θ scan) showed the sample was preferentially oriented with respect to the substrate.[8] The reflections observed correspond to the hexagonal close-packed phase, with 90% of the scattering intensity arising from the (002) and (100) reflections. No measurements were made to ascertain the extent of alignment within the plane of the film. We estimate that approximately 60% of the crystallites are oriented with the [001] axis normal to the film plane and the remainder with the [100] axis normal to the film plane.

The experimental geometry employed in the light scattering studies is illustrated in Fig. 1. The magnetic field H_0 was applied in the plane of the film, perpendicular to the scattering plane. Approximately 300 mW of single mode 514.5-nm Ar^+ laser radiation at an angle of incidence $\theta_i = 42°$ was applied to the sample and the backscattered light at $\theta_s = 30°$ was collected by the optical system and analyzed with a five-pass plus four-pass tandem vernier Fabry-Perot interferometer.[2] All measurements were made at ambient temperature. The magnetic field was measured with a calibrated Hall-effect probe in proximity to the sample and the interferometer was operated with a single etalon free spectral range of 50 GHz yielding an instrumental full width of approximately 700 MHz.

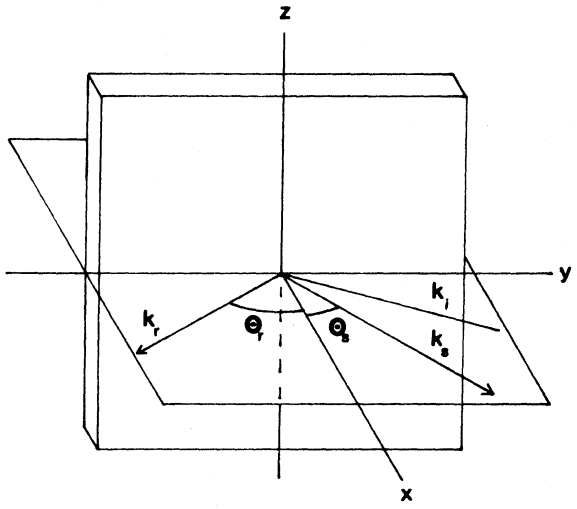

FIG. 1. Geometry of the Brillouin scattering measurements. The magnetic field H_0 is applied in the plane of the sample (y-z) and perpendicular to the scattering plane defined by k_i and k_s. The measurements were made with $\theta_i = 42°$, $\theta_s = 30°$.

In the geometry of Fig. 1, Brillouin scattering from magnons is easily distinguished from scattering from other fundamental acoustic excitations since the magnon scattering is depolarized. Additionally the surface mode is readily identified since, for a given magnetic field direction, it appears only in the Stokes or anti-Stokes scattering. In all cases the surface magnon was readily observed in the first few scans of the interferometer. However, light scattering from the bulk modes was considerably weaker, and accurate determination of the frequency shift of the standing spin-wave modes required considerable signal averaging. Consequently, several hours were spent in accumulating the data at each magnetic field value.

RESULTS

In Fig. 2 we show a typical Brillouin scattering spectrum of the scattered intensity, as a function of frequency

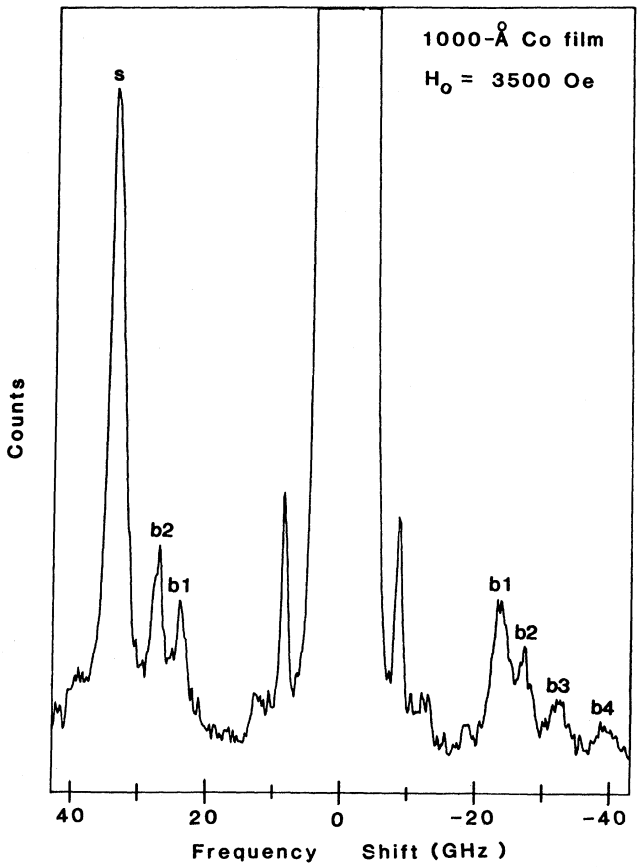

FIG. 2. Scattering intensity vs frequency shift for the 100-nm Co film at room temperature for $H_0 = 3500$ Oe, $\theta_i = 42°$, $\theta_s = 30°$, $\lambda = 514.5$ nm. The large peak labeled S at ~ 35 GHz is the surface magnon, the smaller peaks labeled by b's which occur at both positive and negative frequency shifts are scattering from standing spin-wave modes of the film.

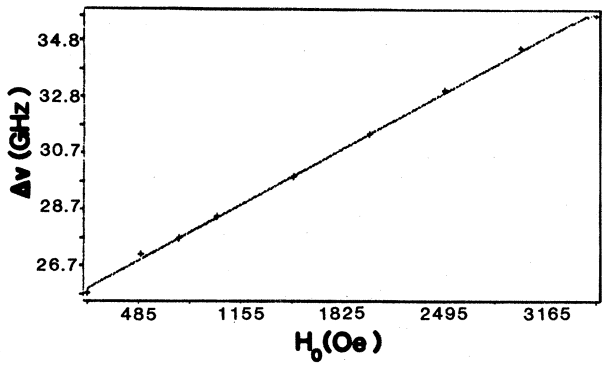

FIG. 3. Frequency shift vs applied field for the surface magnon. The crosses are measured values, and the solid curve is a weighted least-squares fit of Eq. (5) to the measured values. The best fit gave $g=2.16\pm0.02$ and $M_s=1339\pm50$ Oe.

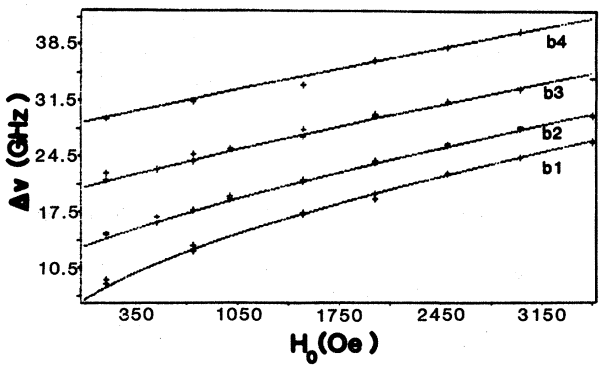

FIG. 4. Frequency shift vs applied field for the first four standing spin-wave modes. The crosses are the absolute value of the measured frequency shifts for both the Stokes and anti-Stokes data. The solid curves are determined as described in the text using the value of g obtained from Fig. 3. Taking $Q_0=\pi/L$ yields $D=(2.7\pm0.05)\times10^{-9}$ Oe cm^2 with $M_b=1325\pm50$ Oe.

shift, for a 100-nm Co film at an applied field of 3500 Oe. The most prominent feature is the surface magnon which is shifted approximately 35 GHz on the anti-Stokes side of the spectrum. The other resolved features are discrete standing spin-wave modes of the bulk ($n=1,2,3,4$) which appear in both the Stokes and anti-Stokes scattering, corresponding to the creation or annihilation of a spin wave, respectively. Note that there is a marked asymmetry in the Stokes—anti-Stokes intensity ratios; however, in all cases where the standing spin wave is well resolved from the surface mode, the frequency shifts of both branches are identical within the experimental errors.

We have reduced the data in the following manner: First we fit Eq. (5) to the surface mode, ignoring exchange contributions to the frequency shift. This is justified since D is of order 10^{-9} Oe cm^2, and $Q_{||}\sim10^5$ cm^{-1} so that $DQ_{||}^2\sim10$ Oe, which is negligible compared to $2\pi M_s$. With this approximation, the frequency shift of the surface magnon is a linear function of H_0 with slope $\gamma=g\mu_B/\hbar$ and intercept $\omega(H_0=0)=2\pi\gamma M_s$. In Fig. 3 we show the least-squares fit of Eq. (5) to the measured frequency shift from which we obtain $g=2.16\pm0.02$ and $M_s=1339+50$ Oe.

In analyzing the bulk standing spin-wave modes, we have taken explicit account of the discrete character of the spectrum and have fit Eq. (3), in the approximation $Q=Q_\perp=nQ_0$ ($n=1,2,3,4$), to all of the modes simultaneously. Using γ obtained from the surface mode, D and M_b were adjusted to minimize the summed squared deviations between the theoretical expression of Eq. (3) and the measured frequency shifts of all of the modes. In Fig. 4 we show the best fit which gave $M_b=1325\pm50$ Oe and $DQ_0^2=270$ Oe. Identifying $Q_0=\pi/L$, where L is the nominal sample thickness gives $D=2.7\pm0.5\times10^{-9}$ Oe cm^2. The error arises from the uncertainty in the sample thickness $L=100\pm10$ nm, not the quality of the fit illustrated in Fig. 4.

DISCUSSION

The values for g and M_b derived from the Brillouin scattering data are in reasonable agreement with those obtained from other techniques. Within the experimental accuracy, our g value, $g=2.16\pm0.02$, is identical with that obtained from ferromagnetic resonance measurements[9] $g=2.18\pm0.01$. The value for M_b, 1330 Oe is approximately 6% less than that obtained from static measurements, $M_0=1420$ Oe, on bulk Co.[10] We note that M_b, as obtained from Brillouin scattering measurements in Fe is approximately 3% smaller than the static M_0, while superior agreement is obtained for Ni.[3] In thin film metglasses M_b and M_0 differ by as much as 12% when measured for the same sample.[11] Our result that $M_s=M_b$ is interesting, since these quantities are observed to differ appreciably in both Fe and Ni films[1] and appear to be sensitive to surface oxidation.

The exchange stiffness constant, $D=2.7\pm0.5\times10^{-9}$ Oe cm$^2=340\pm75$ meV Å^2, extracted from the Brillouin scattering measurements is significantly smaller than that found from neutron scattering[12] (500 ± 10 meV Å^2) and magnetizaton (580 meV Å^2) studies[13] of single-crystal Co. Part of the discrepancy between these results may be attributable to the polycrystalline character of our sample. The D value derived from the light scattering measurements necessarily represents an average exchange stiffness constant, the other measurements obtained D appropriate to the crystalline c axis. However, an unreasonably large anisotropy in the Co exchange interaction would be required to account for the observed differences. We note that D deduced from light scattering and magnetization measurements on the *same* specimen of amorphous $Fe_{80}B_{20}$ varied by a factor of 2.[14]

Camley *et al.* have calculated Brillouin scattering spectra for thin ferromagnetic films and have shown that pinning produces marked effects on the scattered intensity versus applied magnetic field.[4] Pinning most dramatically affects the intensity of the lowest-energy bulk modes in both the Stokes and anti-Stokes scattering. A comparison of our experimental results with these calculations suggests that strong pinning fields are not present in our sample. In general, Brillouin scattering studies of ferromag-

netic films give values of g and M in good agreement with other experimental determinations; however, frequently the exchange stiffness constants derived from light scattering measurements are found to differ significantly from those derived from other experimental techniques. The source of these discrepancies requires further investigation.

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