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Brillouin light-scattering intensities for thin magnetic films with large perpendicular anisotropies

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The intensity of light scattered from thermal magnetic waves in thin magnetic films with large perpendicular uniaxial anisotropies was calculated. The anisotropies were large enough to pull the magnetization out of the sample plane. The intensity of light scattered from the surface magnetic mode in a 3.0-monolayer-thick fcc Fe film grown on Cu(001) was measured as a function of the applied in-plane dc magnetic field. The measured intensities were found to be in excellent agreement with those calculated using the magnetic parameters determined from the measured values of the surface-mode frequency. Both the frequency and intensity data indicated that the magnetization of the Fe film was oriented perpendicular to the sample plane in zero applied field.

One of the most intriguing properties of very thin magnetic films is the presence of large perpendicular uniaxial anisotropies which tend to pull the magnetization out of the plane of the film. This effect was inferred from spin-polarized photoemission measurements[1] and has been predicted theoretically[2,3] and observed experimentally using various techniques: ferromagnetic resonance (FMR),[4,5] Brillouin light scattering (BLS),[5,6] and surface magneto-optic Kerr effect (SMOKE).[7,8] In the previous BLS work,[5,6] perpendicular anisotropy was studied by the measurement of the resonant frequencies of the magnetic films. In the present paper, we show that the effects of perpendicular anisotropy can also be observed by the measurement of the scattered light intensities.

Cochran and Dutcher[9,10] have calculated the normal mode frequencies of a thin ferromagnetic film, and the intensity of light scattered from these modes. The effects of exchange, magnetic damping, and metallic conductivity were included. In the calculation, a dc magnetic field H was applied in the plane of the film and the calculation was valid for films with the magnetization M_s lying in the plane of the film for all values of H. We have extended the calculation to allow M_s to tilt out of the sample plane toward the sample normal; in the calculation, H is still assumed to be applied in the plane of the film. The tilting of M_s is assumed to be due to large uniaxial anisotropies with the easy axis along the sample normal. The uniaxial anisotropy energy can be written as

$$E_u = -K_u^{(1)} \sin^2\alpha - K_u^{(2)} \sin^4\alpha \,, \qquad (1)$$

where α is the angle between M_s and the sample plane. The effective magnetization $(4\pi M_s)_{\rm eff}$ is defined as

$$(4\pi M_s)_{\rm eff} = 4\pi M_s - 2K_u^{(1)}/M_s \,. \qquad (2)$$

If $(4\pi M_s)_{\rm eff} > 0$, M_s lies in the film plane ($\alpha = 0$) for all values of H. If $(4\pi M_s)_{\rm eff} < 0$, M_s is oriented perpendicular to the film plane ($\alpha = 90°$) for $H = 0$ and, as H is increased from zero, M_s is pulled into the film plane. For $H > H_c$, where $H_c = -(4\pi M_s)_{\rm eff}$, M_s lies in the film plane ($\alpha = 0$). The calculation of Cochran and Dutcher[9,10] was valid only for $(4\pi M_s)_{\rm eff} > 0$; the present calculation is valid for both positive and negative $(4\pi M_s)_{\rm eff}$.

Because of the complexities introduced into the calculation by the tilting of M_s with respect to the crystal axes, the effects of exchange have been neglected. The neglect of exchange has two consequences. First of all, the calculation applies only to the surface magnetic mode of ultrathin magnetic films for which the spins are necessarily parallel to one another. Standing spin-wave modes, for which the spins are not necessarily parallel, are not calculated. However, for ultrathin magnetic films, the standing spin-wave modes have very high frequencies [$\sim$23 000 GHz for the first-order standing spin-wave mode for a three-monolayer (ML) thick Fe film] and cannot be measured using BLS. Secondly, there is a slight shift in the surface mode frequency for ultrathin magnetic films when exchange is included, but this shift is small ($\sim$0.5 GHz).[9]

In the calculation, magnetic damping of the Gilbert form was included. An in-plane anisotropy term was not included since the surface mode frequency was found to be independent of the direction of the magnetic field in the sample plane.

The light-scattering calculation followed very closely that of Cochran and Dutcher.[9,10] An outline of the calculation and differences between it and that of Cochran and Dutcher are given below. The explicit details of the calculation have been given by Dutcher.[11] The surface mode frequency of the magnetic film was calculated by combining the Landau-Lifshitz equation of motion for the magnetization, Maxwell's equations for the radio-frequency electric and magnetic fields, and the electromagnetic boundary conditions. The Landau-Lifshitz equation has its simplest form in a coordinate system in which M_s lies along one of the coordinate axes. In this coordinate system, the magnetic permeability tensor was constructed using the linearized Landau-Lifshitz equation for which terms that were second or higher order in components of the magnetization transverse to its equilibrium direction were neglected. The permeability tensor was then transformed into a coordinate system fixed with respect to the magnetic film for the solution of the boundary value problem for the surface mode frequency. The magnetic permeability tensor was not used by Cochran and Dutcher;[9] it was used in the present calculation to facilitate the transformation of the magnetic quantities determined by the Landau-Lifshiftz equation from one coordinate system to the other. With the magnetization tilted out of the magnetic film plane, the boundary value problem consisted of the solution of four equations obtained from the electromagnetic boundary conditions for the electric and magnetic field components tangential to the magnetic film surface. With the magnetization lying in the magnetic film plane, only two electromagnetic boundary conditions were necessary to solve the boundary value problem. A simplified expression for the surface mode frequency, neglecting the effects of magnetic damping and electrical conductivity, was given by Dutcher *et al.*[6]

The relative values of the magnetization components were calculated using the combined Maxwell's equations and the Landau-Lifshitz equation along with the calculated value of the surface mode frequency. The amplitudes of the magnetization components were determined by setting the energy contained in the magnetic mode equal to the equipartition value, $k_B T$. A contribution to the electric polarization is produced which is proportional to the product of the spatial distributions of the transverse magnetization and the electric field set up in the film by the incident optical wave. Only terms linear in the transverse magnetization were retained. The resulting electric polarization was then used to calculate the scattered light intensity, using the optical Green's functions of Cochran and Dutcher.[10] The electric fields in the film due to the incident optical wave and the optical Green's functions are unaffected by the tilting of the magnetization out of the sample plane; only the transverse magnetization components are changed by the tilting of the magnetization.

We have performed BLS measurements in air at room temperature on a sample consisting of a 3.0-ML Fe film deposited on a Cu(001) substrate and covered with a 60-ML Cu film [60-ML Cu, 3.0-ML Fe, Cu(001)]. The preparation of this type of sample has been described previously.[12,13] 140 mW of light from an Ar^+ laser ($\lambda = 514.5$ nm), polarized in the plane of incidence, was focused on the sample surface and 180° back-scattered light, polarized perpendicular to the plane of incidence, was collected and analyzed by means of a high contrast, tandem (4+2 passes) Fabry-Perot interferometer. The angle of incidence of the light with respect to the sample normal was 45°. A dc magnetic field, variable between 0 and 10 kOe, was applied in the plane of the sample and perpendicular to the plane of incidence.

The surface mode peaks for the 60 ML Cu, 3.0 ML Fe, Cu(001) samples were observed easily above the background level in the BLS spectra for applied field values greater than 2 kOe. The peaks had narrow measured linewidths that were determined by the Fabry-Perot instrumental function. In the spectra for similar samples that were studied previously,[6,11] the surface mode peaks were also observed easily above the background level but the peak linewidths were twice that of the instrumental function. These samples were likely less uniform magnetically than the 60 ML Cu, 3.0 ML Fe, Cu(001) sample discussed in the present paper. Figure 1 shows the surface mode frequency as a function of the applied in-plane dc magnetic field H for the 60 ML Cu, 3.0 ML Fe, Cu(001) sample. The crosses are experimental data and the solid curve has been calculated using the magnetic parameters listed in Fig. 1. To obtain the curve, $(4\pi M_s)_{\text{eff}}$ and the g factor were chosen to pass the high-field branch ($H > H_c = 4.0$ kOe) through the high-field data points. The value of $K_u^{(2)}$ was then chosen to pass the low-field branch

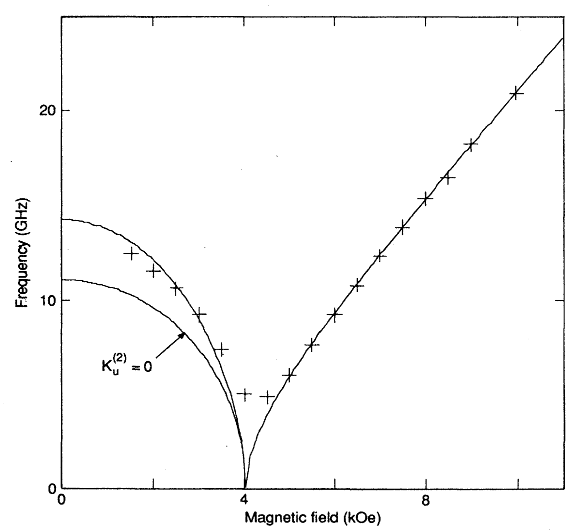

FIG. 1. Magnetic field dependence of the surface mode frequency for the 60 ML Cu, 3.0 ML Fe, Cu(001) sample. The crosses are measured data points. The solid curve which passes through the data points has been calculated using the following parameters: effective magnetization $(4\pi M_s)_{\text{eff}} = -4.0$ kG, g factor $g = 1.95$, second-order uniaxial anisotropy field $4K_u^{(2)}/M_s = 1.17$ kOe, Gilbert damping parameter $G = 7.0 \times 10^7$ Hz, and resistivity $\rho = 1.0 \times 10^{-5}$ Ω cm. The curve calculated using the same values of $(4\pi M_s)_{\text{eff}}$, g, G and ρ, and $K_u^{(2)} = 0$, is also indicated.

$(0 < H < H_c)$ through the low-field data points. The value of $K_u^{(2)}$ does not affect the high-field branch.[6] The low-field branch corresponding to $K_u^{(2)} = 0$ is also shown in Fig. 1. Since the value of $(4\pi M_s)_{\rm eff}$ required to fit the data is negative, the magnetization is oriented perpendicular to the film plane in zero applied field. Figure 1 is very similar to the results obtained previously for single films and superlattices of Fe on Cu(001).[5,6,11]

The magnetic field dependence of the surface mode scattering intensity is shown in Fig. 2. The crosses are experimental data and the curve has been calculated using the magnetic parameters determined from the surface mode frequency measurements in Fig. 1. The background level in the BLS spectra corresponds to a scattering intensity of approximately 0.002 in the units of Fig. 2. The vertical scale of the calculated curve has been adjusted to pass the high-field branch of the curve through the high-field data points. The agreement between the data and the calculation is very good. For both the data and the calculation, there is a dramatic increase in the intensity of the surface mode peaks near $H = H_c = -(4\pi M_s)_{\rm eff} = 4.0$ kOe. Also, for both the data and the calculation, the scattering intensities for the low-field branch are considerably lower than those for the high-field branch for equal positive and negative field increments away from $H = H_c$. The peak in the scattering intensity can be understood as follows. When the applied field $H = H_c$, the effective field acting on the magnetization is equal to zero and the precessing magnetization deviates substantially from its equilibrium direction. The large transverse magnetization components give rise to a large scattered intensity. The peak in the intensity for a nonzero value of H shows that $(4\pi M_s)_{\rm eff} < 0$. In the calculation, it was assumed that the transverse magnetization values remained small compared with the saturation magnetization. For the curve shown in Fig. 2, the transverse magnetization components are always less than 0.2% of the saturation magnetization. This justifies the use of the linearized form of the Landau-Lifshitz equation to calculate the solid curve of Fig. 2.

Dutcher *et al.*[15] have estimated the local heating due to the focused laser beam from precision BLS measurements on Fe whiskers. For the Fe whisker samples, an incident laser power of 140 mW produced a temperature increase of 26 K. Accounting for the differences in the thermal conductivities and reflectivities of Fe and Cu, we estimate the local heating of the present sample to be less than 10 K. This low value of the local heating means that the focusing of the incident laser beam should not have a significant effect on the magnetic properties of the sample.

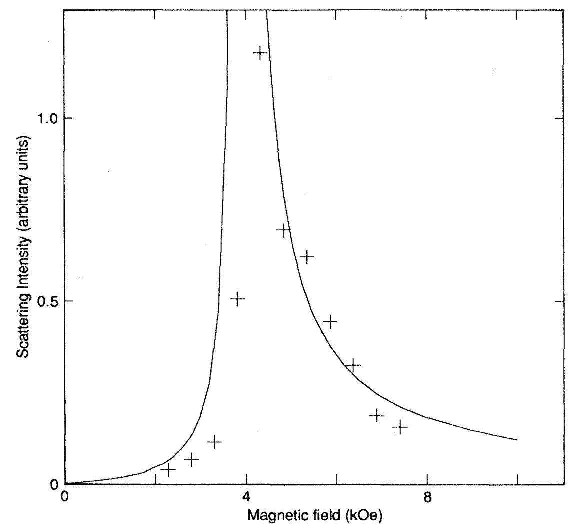

FIG. 2. Magnetic field dependence of the surface mode scattering intensity for the 60 ML Cu, 3.0 ML Fe, Cu(001) sample. The crosses are measured data points. The solid curve has been calculated using the magnetic parameters determined from Fig. 1: $(4\pi M_s)_{\rm eff} = -4.0$ kG, $g = 1.95$, and $4K_u^{(2)}/M_s = 1.17$ kOe. Also, $G = 7.0 \times 10^7$ Hz and $\rho = 1.0 \times 10^{-5}$ Ω cm. Only the first-order magneto-optic constant (Ref. 14) was used in the calculation of the scattering intensity.

We have shown that BLS measurements of the intensity of light scattered from the magnetic surface mode can be used to observe perpendicular anisotropy in ultrathin magnetic films.

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