Oscillations in the Stokes–anti-Stokes ratio in Brillouin scattering from magnons in thin permalloy films

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A Brillouin light-scattering study of surface magnons in permalloy thin films shows a distinct oscillation in the Stokes–anti-Stokes intensity ratio as a function of the in-plane wave number in the 0– 2.5×10^5 cm^{-1} range. This result differs from the previously reported monotonic dependence for thin iron thin films.

I. INTRODUCTION

Brillouin light scattering (BLS) has become a well-established technique for the detection and characterization of magnons in magnetic materials.[1] While the main focus has been on magnon frequencies, the relative intensities for the magnon lines also contain important information about magnon properties. The measurement of the Stokes–anti-Stokes (S/AS) intensity ratio, while somewhat difficult, has yielded interesting results.[2,3] The objective of this work is to examine this intensity ratio versus wave number for surface magnons in permalloy films.

There are several physical effects which cause Stokes–anti-Stokes intensity asymmetry and yield a Stokes–anti-Stokes intensity ratio (S/AS ratio) different from one. The main effect is related to the nonreciprocal propagation behavior of surface magnons.[4] This property is shown schematically in Fig. 1. For the magnetic field **H** into the page, the surface wave vector labeled $+\mathbf{k}_m$ can only be supported for a top surface magnon. The bottom surface can support only $-\mathbf{k}_m$. The propagation directions reverse for a reversal of the magnetic field. The amplitude of the surface magnon associated with one surface decays exponentially as one moves into the film with a decay length on the order of the in-plane propagation wavelength.

The BLS geometry for this work is also shown in Fig. 1. For surface magnons in metal films, it is the net parallel component of the wave vectors that is conserved in the scattering process.[5] For the scattering geometry and the allowed surface magnon propagation directions discussed above, the scattering can only result in the creation of a surface magnon associated with the top surface or the destruction of a magnon associated with the bottom surface. The creation of a magnon results in a decrease in the frequency of the scattered light or a Stokes peak; magnon destruction yields an increase in the frequency of the scattered light or an anti-Stokes peak.

The S/AS intensity ratio for the above situation will simply scale with the relative intensity of the top and bottom surface magnons at the top surface. To a good approximation, this ratio is given by

$$\mathrm{S/AS}^+ = \exp(2k_m d), \tag{1}$$

where d is the film thickness and k_m is the in-plane propagation wave number. $\mathrm{S/AS}^+$ refers to the situation in Fig. 1 for **H** into the page. If the field is reversed, the above expression then applies to the anti-Stokes–Stokes ratio, $\mathrm{AS/S}^-$. For very thin film, such that the light reaches both surfaces, the S/AS ($\mathrm{S/AS}^+$ or $\mathrm{AS/S}^-$) ratio approaches unity. Qualitative support for this simple model has come from the observed inversion of the S/AS ratio for a reversal in the direction of the magnetic field[5,6] or the in-plane component of the incident light.[7] In fact, this inversion has become the main test for surface magnons.

There are two other effects which may effect the S/AS ratio. The first is due to the Boltzmann distribution governing the population of thermal magnons. For BLS experiments at room temperature and magnon frequencies below 2 cm^{-1} (60 GHz), this effect yields a deviation of the S/AS ratio from one of less than 1%. The second effect involves the interference of linear and quadratic terms in the magneto-optic dielectric tensor.[3,8] For these two effects, the S/AS ratio is not influenced by a reversal of the magnetic field.

Quantitative data on the S/AS ratio as a function of wave number are sparse. Experimental results by Metawe and Grünberg[9] for a 1000-Å EuO film and $k_m = 0.68\times10^5$ cm^{-1} agree with the simple theory within experimental error. Camley and co-workers[2] report measured S/AS ratios for 100-Å Ni and $Ni_{0.8}Fe_{0.2}$ (permalloy) films for $k_m = 1.83\times10^5$ cm^{-1} and for a 100-Å Fe film for four different values of k_m between 0.89×10^5 and 2.24×10^5 cm^{-1}. These data show a much larger deviation of the S/AS ratio from unity than expected from Eq. (1). Kabos *et al.*[10] have measured the S/AS ratio for a 232-Å Fe film for ten different values of k_m between 0.5×10^5 and 2.2×10^5 cm^{-1}. They find reasonably good agreement with Eq. (1) for $k_m > 1.0\times10^5$ cm^{-1}. For lower k_m values, however, their S/AS ratio appears to level off at a value well above unity. This result is attributed to either the problem of doing low-angle backscattering or to a failure of the simple Damon–Esbach (DE) theory for $2\pi/d \gg k_m$. Srinivasan and Patton[11] report a measurement of the S/AS ratio for a 50-Å $Ni_{80}Fe_{20}$ film with $k_m = 0.42\times10^5$ cm^{-1}. Contrary to the statement in text, their experimental result of about 1.2 for the S/AS ratio is somewhat bigger than the value of 1.04 from Eq. (1).

The theoretical situation is also unclear. A theory by Camley and co-workers[2] for absorptive materials takes into account the contribution of the off-diagonal spin-spin correlation function and shows qualitative agreement with the Ref. 2 data. In connection with the present measurements, calculations based on the theory of Cochran and Dutcher[12] have been made. These calculations are in agreement with

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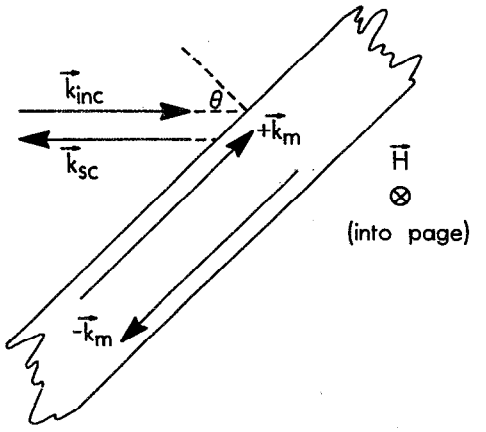

FIG. 1. Diagram of permalloy film in cross section and the experimental light-scattering geometry. The top-surface and bottom-surface magnons have wave vectors of $+\ \mathbf{k}_m$ and $-\ \mathbf{k}_m$, respectively. The incident and scattered light beams at an angle of incidence Θ have wave vectors $\mathbf{k}_{inc}$ and $\mathbf{k}_s$, respectively. The static field $\mathbf{H}$ is directed into the page.

the theory of Ref. 2 for thicker films, but not for very thin films.

This brief review of the few published results on the Stokes–anti-Stokes intensity ratios for surface magnons in thin films indicates the rather confusing state of the subject and the need for a systematic experimental investigation of the matter.

II. EXPERIMENT

The light-scattering measurements were made on a series of $Ni_{80}Fe_{20}$ permalloy films sputtered on silicon. Results described here are for three films with thicknesses of 47, 107, and 242 Å, as determined from calibrated hysteresis loop measurements. The 242-Å sample was also characterized by ferromagnetic resonance (FMR); the FMR data yielded a gyromagnetic ratio of $\gamma = 2.90$ GHz/kOe and a saturation magnetization of $4\pi M = 9.34$ kOe.

The BLS measurements were made following the procedure described in Refs. 10 and 11. A p-polarized, single-mode, 488-nm Ar^+ - ion laser beam is focused on the sample at an angle of incidence θ as shown in Fig. 1. The backscattered light is analyzed with a scanning multipass tandem interferometer.[1] The surface magnon wave number k_m is selected by changing Θ and is given by $k_m = (4\pi/\lambda)\sin\Theta$.

Figure 2 shows typical spectra for the 107-Å film with $k_m = 1.43\times10^5$ cm^{-1}. The configuration for Fig. 2(a) is as shown in Fig. 1, with $H = +1$ kOe. The stronger surface magnon line, the Stokes peak, is to the right of the central peak. For Fig. 2(b), the field is reversed ($H = -1$ kOe). Here, the anti-Stokes peak is stronger. This inversion indicates that the BLS peaks are associated with DE surface magnons.

For each film, four spectra were obtained at each of 14 Θ values ranging from 0° to 79° corresponding to a range of k_m values between 0 and 2.5×10^5 cm^{-1}, for a total of 162 spectra. The in-plane magnetic field was set at 1 kOe. Two of the four spectra were taken with the magnetic-field direction as indicated in Fig. 1 ($H = +1.0$ kOe); the S/AS

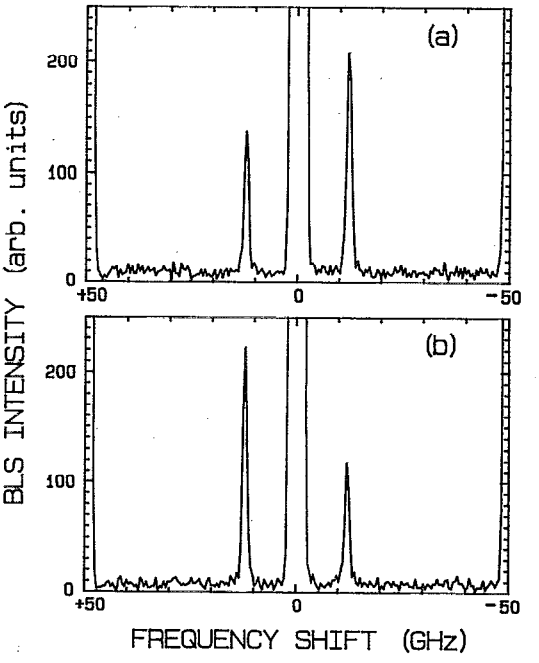

FIG. 2. Brillouin light-scattering spectrum for the 107-Å permalloy film. The scattering angle corresponds to a surface magnon wave vector of $k_m = 1.43\times10^5$ cm^{-1}. The applied field H is in the plane of the film: (a) $H = +1.0$ kOe; (b) $H = -1.0$ kOe. The surface magnon peaks are located on either side of the central Rayleigh scattered peak.

ratios for these spectra are designated by S/AS^+. The other two spectra were taken with the magnetic field in the opposite direction ($H = -1.0$ kOe) and the S/AS ratios are specified by S/AS^-. S/AS intensity ratios were determined by integration over the area of each peak.

III. RESULTS

S/AS^+ and AS/S^- ratio determinations versus magnon wave number k_m for the 107-Å film are shown in Fig. 3. Each data point represents the average of two measurements. The data are typical of data for the other films. The data in Fig. 3 show the key result of this paper, the clear

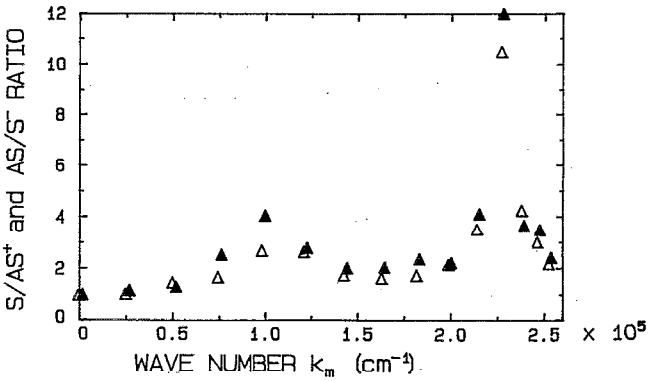

FIG. 3. Stokes–anti-Stokes intensity ratio vs wave number k_m for the 107-Å permalloy film. The open triangles are for S/AS^+ ratios and the solid triangles for AS/S^- as discussed in the text.

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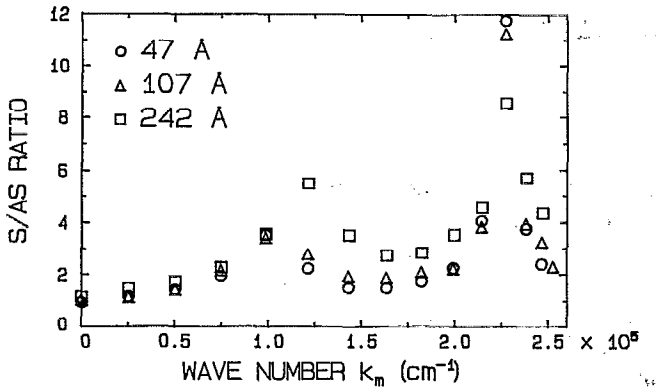

FIG. 4. Stokes–anti-Stokes intensity ratio vs wave number k_m for the 47-, 107-, and 242-Å permalloy films.

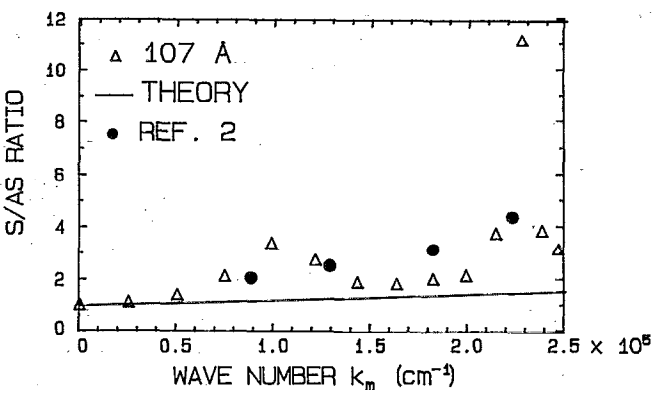

FIG. 5. Stokes–anti-Stokes ratio vs wave number k_m. The open triangles are for the 107-Å permalloy film. The solid circles represent data from Ref. 2 for a 100-Å iron film. The solid line is calculated from the simple exponential model for a 100-Å film.

and distinct oscillation of the Stokes–anti-Stokes ratio as a function of surface magnon wave nunber k_m. All available theories predict a monotonic dependence of the S/AS ratio on wave number k_m.

There are two other noteworthy features of the results in Fig. 3. First, the S/AS^+ and AS/S^- ratios converge to unity in the limit $k_m d \to 0$, as expected. Second, there is a tendency for the AS/S^- ratio to be somewhat larger than the S/AS^+ ratio. This is probably related to the asymmetry of the two surfaces in the experiment due to the silicon substrate on the rear surface.

Figure 4 shows experimental results for all three films. Each data point represents the average of two measurements of the S/AS^+ ratio and two measurements of the AS/S^- ratio. The oscillations for the three films are very similar. While the oscillations are not accounted for by any known theory, the simple surface mode exponential decay model of Eq. (1) does account qualitatively for the change in the S/AS ratio with thickness.

Although the previous data of Camley and co-workers[2] show no distinct oscillation of the kind reported here, those data do not preclude such oscillations. Figure 5 shows the data of Ref. 2 for a 100-Å film along with the present 107-Å permalloy film results. These two sets of data, even if for different materials, are not inconsistent. The Ref. 2 results do not preclude the oscillatory character documented herein. Both sets of data show S/AS ratios well above the prediction of the simple exponential model of Eq. (1).

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