PHYSICAL REVIEW B VOLUME 26, NUMBER 5 1 SEPTEMBER 1982

Stokes—anti-Stokes asymmetry in Brillouin scattering from magnons in thin ferromagnetic films

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(Received 9 March 1982)

A large asymmetry is found in the intensity between Stokes peaks and the equivalent anti-Stokes peaks in scattering from magnons in very thin ferromagnetic films. It is shown that this asymmetry is due to the contribution of the off-diagonal spin-spin correlation function $\langle S_x S_y \rangle$ to the light scattering intensity. This contribution is only seen when the wave vector of the light in the ferromagnet is complex, the case for absorptive materials. The asymmetry is studied both theoretically and experimentally, and good agreement is found.

INTRODUCTION

For many years it has been known that in light scattering from magnetic materials there were at least two sources for an asymmetry in the intensity of Stokes peaks compared to anti-Stokes peaks. The first of these is due to thermal population factors.[1,2] Essentially it is more probable to create a spin wave (Stokes process) than to destroy one (anti-Stokes process), because in order to destroy a spin wave it must first have been thermally excited. This difference is eventually reflected in a Stokes—anti-Stokes (S-aS) ratio of $\exp(\hbar\Omega/k_B T)$, where Ω is the frequency of the spin wave, k_B is Boltzmann's constant, and T is the temperature.

The second cause may be understood as follows.[3,4] The change in the dielectric constant due to spin fluctuations may be expanded to second order in the spin densities. Thus

$$\delta\epsilon_{\alpha\beta}(\vec{x},t)=\sum_{\gamma}K_{\alpha\beta\gamma}S_{\gamma}(\vec{x},t)+\sum_{\gamma\delta}G_{\alpha\beta\gamma\delta}S_{\gamma}(\vec{x},t)S_{\delta}(\vec{x},t)\ . \qquad (1)$$

In this equation $S_\gamma(\vec{x},t)$ is the γth component of spin density at position $\vec{x}$ and time t, and $K_{\alpha\beta\gamma}$ and $G_{\alpha\beta\gamma\delta}$ are the elements of the linear and quadratic coupling tensors, respectively. The quadratic terms may contribute to one-magnon scattering if γ or α is in the direction of the saturation magnetization M_s. In this case, $S_\gamma(\vec{x},t)$ or $S_\delta(\vec{x},t)$ may be replaced by nS, where n is the number of spins per unit volume. The scattering from the quadratic terms may then interfere with that from the linear terms. For a cubic material with the field along the [100] direction, there is only one independent element of the linear and quadratic coupling tensors, K and G, respectively. Then one can show that on the Stokes side the linear and quadratic terms interfere destructively, and the scattering intensity is proportional to $|K-nSG|^2$. On the anti-Stokes side the two terms interfere constructively and the scattering intensity is proportional to $|K+nSG|^2$.

The two mechanisms above, the thermal population effect and the linear-quadratic effect, have one feature in common. A reversal of the magnetic field does not alter the S-aS ratio.

Recently in a series of experimental[5-9] and theoretical[10-14] papers involving light scattering from surface magnons, a large S-aS intensity difference has been seen which could not be explained by the above two mechanisms. The origin of this symmetry is that the eigenvectors of the created and destroyed waves are not equivalent. This inequivalence has been viewed primarily as a *localization effect* due to the surface. This is illustrated in Fig. 1. The surface spin wave propagating as shown in Fig. 1(a) necessarily has its largest amplitude near the top surface. A surface spin wave propagating in the opposite direction will be localized at the bottom surface. If the geometry of the light scattering experiment is designed so as to create the spin wave in Fig. 1(a), then the spin wave destroyed will be the one shown in Fig. 1(b).

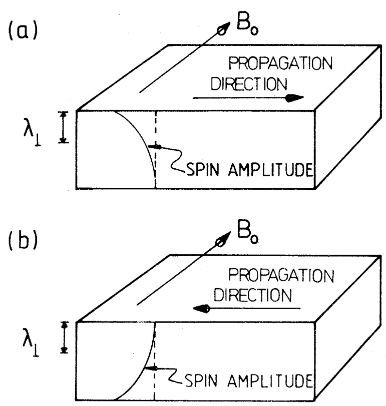

FIG. 1. Schematic of surface spin-wave localization as a function of propagation direction. In (a) the spin wave is localized near the top, in (b) near the bottom. λ_l is the penetration depth of the incident laser light. Light scattering from (a) will be greater than from (b).

Because of the different localization of the two spin waves and the finite penetration depth of the incident light, then scattering with creation of a surface magnon at the top will be greater than that with destruction of a suface magnon at the bottom. Since a reversal of the applied magnetic field and sample magnetization will cause a reversal of the localization of the surface spin waves there will also be a reversal of the S-aS asymmetry.[5] Owing to the presence of the surface, bulk waves will also be modified, and scattering from bulk waves may also show asymmetry due to localization.

If one investigates very thin samples ($\lesssim 15$ nm), the localization of the surface spin waves disappears because the penetration depth λ_s ($\lambda_s = 1/Q_{||}$, where $Q_{||}$ is the wave vector of the surface wave parallel to the surface) of the surface spin wave becomes much larger than the thickness of the film. One might expect the S-aS ratio to approach unity since the localization mechanism no longer holds. Surprisingly, however, this is not the case. A large S-aS intensity difference remains. This difference is not due to thermal factors because the spin wave energies are small compared to $k_B T$. The difference is also not due to the linear-quadratic effect because reversal of the applied field essentially reverses the S-aS ratio.

In this paper, we therefore investigate experimentally and theoretically the origin and behavior of the S-aS intensity ratio in very thin ferromagnetic films. We find that the asymmetry is due to the contribution of the off-diagonal spin-spin correlation function $\langle S_x S_y \rangle$ to the light scattering intensity. Thus it is a function of the *rotation* and *ellipticity* of the spin precession. We will show on the basis of a symmetry argument that the Fourier transform of this correlation function changes sign when contributing to the intensity on the Stokes and anti-Stokes side. The theoretical predictions for the S-aS intensity ratio made on this basis are shown to be in good agreement with experimental results obtained from thin films of Fe, Ni, and $Ni_{0.8}Fe_{0.2}$ (Permalloy).

THEORY

The basic formalism used here for light scattering from a semi-infinite ferromagnet and a ferromagnetic thin film was developed in Refs. 10–12. Here we consider the case of very thin

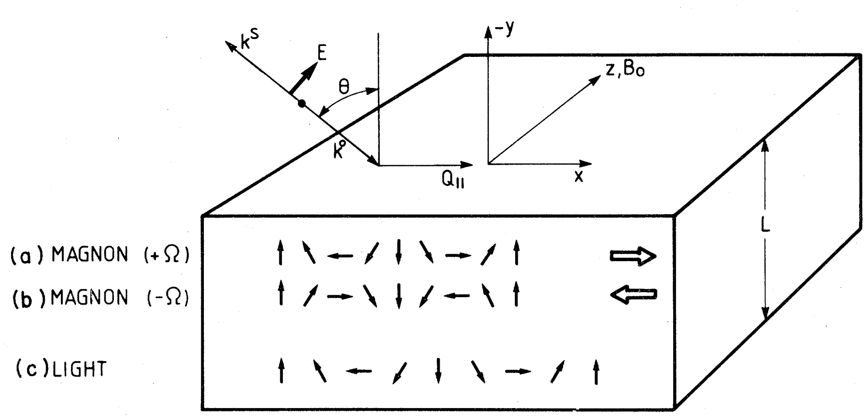

FIG. 2. Geometry considered in this paper. The incident light, with wave vector k^0, strikes the surface at an angle θ with respect to the surface normal. The scattered light, with wave vector k^s, is backscattered in the direction of the incident beam. Also shown is the sense of rotation for magnons propagating from (a) left to right and (b) right to left and (c) the rotation of the electric field vector in the medium.

films and reduce the previously derived equations to a simpler form so that the origin of the S-aS asymmetry is easily understood.

The geometry is illustrated in Fig. 2. The xz plane defines the top surface. The applied field is parallel to the z axis. The incident light has wave vector $\vec{k}^0$ and frequency ω_0, while the scattered light has wave vector $\vec{k}^s$ and frequency ω_s. We briefly review here some results and notations from the earlier work. Eventually the light scattering cross section is related to the spin-spin correlation function. We write

$$S_{ij}(\vec{x};\vec{x}\,';t-t')=\langle S_i(\vec{x},t)S_j(\vec{x}\,',t')\rangle \ . \tag{2}$$

Since the system has translational invariance parallel to the surface, we may write $S_{ij}(\vec{x},\vec{x}\,';t-t')$ in terms of its Fourier transform

$$S_{ij}(\vec{x},\vec{x}\,';t-t')=\int \frac{d\vec{q}_{\parallel}d\omega}{(2\pi)^3}S_{ij}(\vec{q}_{\parallel},\omega,y,y)\exp[i\vec{q}_{\parallel}(\vec{x}_{\parallel}-\vec{x}\,'_{\parallel})-i\omega(t-t')] \ , \tag{3}$$

where $\vec{x}_{\parallel}$ is the projection of $\vec{x}$ on the plane parallel to the surface. The diagonal elements $S_{ii}(\vec{q}_{\parallel},\omega,y,y)$ have the following physical interpretation. Imagine a thin slab of thickness dy, with surfaces parallel to the surface of the film. Then $S_{xx}(q_{\parallel},\omega,y,y)$ is a measure of the square of the amplitude to find a spin fluctuation, in the $\hat{x}$ direction, located at distance y from the surface, with a wave vector $\vec{q}_{\parallel}$ and frequency ω. These functions were studied in detail in Refs. 10 and 12.

From Ref. 10 one may relate the intensity of the scattered light to the Fourier transform of the spin-spin correlation function. We find

$$\begin{aligned} I(\vec{k}^0,\vec{k}^s,\Omega)= \int_0^L & dy\,dy'\exp(i\Delta k_{\perp}y-i\Delta k_{\perp}^{*}y') \\ & \times[\,r_{yy}S_{yy}(\vec{Q}_{\parallel},\Omega,y,y')+r_{xy}S_{xy}(\vec{Q}_{\parallel},\Omega,y,y')+r_{yx}S_{yx}(\vec{Q}_{\parallel},\Omega,y,y')+r_{xx}S_{xx}(\vec{Q}_{\parallel},\Omega,y,y')] \ . \end{aligned} \tag{4}$$

In the above expression $\vec{Q}_{\parallel}$ is the wave vector of the spin wave created or destroyed in the light scattering experiment. Thus

$$\vec{Q}_{\parallel}=\vec{k}^0_{\parallel}-\vec{k}^s_{\parallel} \ . \tag{5}$$

Similarly

$$\Omega=\omega_0-\omega_s \ . \tag{6}$$

Thus for a Stokes process Ω is positive; for an anti-Stokes process Ω is negative. The terms $r_{yy},r_{yx},r_{xx},r_{xy}$ are related to factors of the light scattering geometry, such as the angles that the incident and scattered light make with the surface normal, the polarizations, and the penetration depth of the incident laser light. Expressions for these terms may be found in Refs. 10 and 11. Finally,

$$\Delta k_{\perp}=k^0_{\perp}+k^s_{\perp} \ , \tag{7}$$

where $k^0_{\perp}$ and $k^s_{\perp}$ are the components of $\vec{k}^0$ and $\vec{k}^s$ perpendicular to the surface *in the material.*

From Eq. (4) we see that the intensity difference between the Stokes and anti-Stokes sides results from the change of $+\Omega$ to $-\Omega$ in the functions $S_{ij}(\vec{Q}_{\parallel},\Omega,y,y')$. We can relate $S_{ij}(\vec{Q}_{\parallel},+\Omega,y,y')$ to $S_{ij}(\vec{Q}_{\parallel},-\Omega,y,y')$ through the following symmetry argument. Time reversal is not a good symmetry operator for our system. However, time reversal followed by reflection through the $y=L/2$ plane is a symmetry operation. As an example we consider the correlation function

$$\langle S_x(\vec{x},t)S_y(\vec{x}\,',0)\rangle \ . \tag{8}$$

Application of time reversal changes the sign of t to $-t$, and because S is an axial vector S_x and S_y also change sign. We then have

$$\langle S_x(\vec{x},-t)S_y(\vec{x}\,',0)\rangle \ . \tag{9}$$

We then reflect through the $y=L/2$ plane. This changes y to $L-y$, and because S is an axial vector S_x changes to $-S_x$ but S_y is unchanged. We obtain

$$-\langle S_x(\vec{x}_{\parallel},L-y,-t)S_y(\vec{x}\,'_{\parallel},L-y',0)\rangle \ . \tag{10}$$

Since we have now performed a symmetry transformation the correlation function in (8) must be equivalent to the correlation function in (10). When we write (8) and (10) in their Fourier expansions as done in Eq. (3) and equate Fourier coefficients, we obtain

$$S_{xy}(\vec{Q}_{\parallel},+\Omega,y,y')=-S_{xy}(\vec{Q}_{\parallel},-\Omega,L-y,L-y') . \tag{11}$$

By a similar argument one can show

$$S_{yx}(\vec{Q}_{\parallel},+\Omega,y,y')=-S_{yx}(\vec{Q}_{\parallel},-\Omega,L-y,L-y') , \tag{12}$$

$$S_{xx}(\vec{Q}_{\parallel},+\Omega,y,y')=+S_{xx}(\vec{Q}_{\parallel},-\Omega,L-y,L-y') , \tag{13}$$

$$S_{yy}(\vec{Q}_{\parallel},+\Omega,y,y')=+S_{yy}(\vec{Q}_{\parallel},-\Omega,L-y,L-y') . \tag{14}$$

We see in the diagonal functions S_{xx} and S_{yy} a common feature: Reversing the frequency is equivalent to turning the film upside down. For example, if we examine the spin fluctuations with positive frequency near the top surface ($y=y'=0$), this is equivalent to examining spin fluctuations with negative frequency but at the bottom surface ($y=y'=L$). In thicker samples where the surface spin wave is localized near the top or bottom surface, this provides the asymmetry between the S-aS intensities as discussed in the Introduction.

Here we are interested in very thin samples. In this case the function $S_{ij}(\vec{Q}_{\parallel},\Omega,y,y')$, evaluated in the region of the surface wave, is nearly constant across the thickness of the film, and we may neglect variations in y and y'. Thus we set

$$S_{ij}(\vec{Q}_{\parallel},\Omega)=S_{ij}(\vec{Q}_{\parallel},\Omega,y=y'=0) . \tag{15}$$

The equation for the intensity may then be written

$$I=|B|^2 r_{yy}S_{yy}(\vec{Q}_{\parallel},\Omega)+r_{xx}S_{xx}(\vec{Q}_{\parallel},\Omega) +2\,\mathrm{Re}[r_{yx}S_{yx}(\vec{Q}_{\parallel},\Omega)] , \tag{16}$$

where

$$B=\int_0^L dy\,\exp(i\Delta k_{\perp}y) , \tag{17}$$

and we have used

$$S_{xy}(\vec{Q}_{\parallel},\Omega)=S^{*}_{yx}(\vec{Q}_{\parallel},\Omega) , \tag{18}$$

which may be derived from the definitions in Ref. 10.

We see from Eqs. (11)–(14) that when Ω changes from positive to negative the diagonal elements S_{xx} and S_{yy} remain unchanged, but the off-diagonal elements S_{yx} change sign. Thus if S_{yx} has a positive contribution to the intensity on the Stokes ($+\Omega$) side, then S_{yx} will have a negative contribution on the anti-Stokes ($-\Omega$) side. This is the origin of the asymmetry.

The asymmetry in the S-aS ratio that one sees in scattering from surface spin waves in thicker samples occurs because of the unusual properties of the long-wavelength surface spin wave, i.e., that the spin wave is localized at the top or bottom surface depending on the direction of propagation. This feature is due explicitly to the dipole-dipole interaction and the existence of the surface. The mechanism considered here, in contrast, is a general feature of magnetic systems. From the symmetry argument we see that this result does not depend on the nature of the coupling between spins. Also, the existence of the surface is not necessary. Scattering from bulk waves will also show this asymmetry if the spin deviations are symmetric about the $y=L/2$ plane.

We note the following properties. The factors r_{xx} and r_{yy} are real and positive; r_{yx} is complex. The correlation functions S_{xx} and S_{yy} are also real and positive, but the off-diagonal correlation function S_{yx} is purely imaginary and, as we have seen, may be positive or negative. That S_{yx} is imaginary simply reflects the fact that S_x and S_y are 90° out of phase. Since the final intensity must be purely real, the off-diagonal correlation function S_{yx} only contributes to the intensity when r_{yx} has an imaginary part. From the definitions of r_{yx} in Refs. 10 and 11 one can see that r_{yx} is complex only when the wave vector of the incident light in the ferromagnet is complex. This is the case when there is strong absorption of the light. This explains why S-aS asymmetries that cannot be explained by some other mechanism have not been seen earlier. Most previous work has been on light scattering from nearly transparent materials. Only recently, with the investigation of light scattering from ferromagnetic metals, have the measured absorption values been large. And in many of these investigations a large S-aS asymmetry has been seen.

We illustrate the physical difference between the spin waves with $+\Omega$ and $-\Omega$ in Fig. 2. $Q_{\parallel}$ is directed along the $+\hat{x}$ axis. An individual spin always, regardless of direction of propagation precesses clockwise about the magnetic field. One can then see that for propagation from left to right ($+\Omega$) the spatial arrangement of a spin wave must be as illustrated in Fig. 2(a). For propagation from right to left ($-\Omega$) the spatial arrangement is illustrated in Fig. 2(b). These two arrangements are not equivalent in that the sense of precession as one moves from left to right is opposite. This difference between the wave with $+\Omega$ and $-\Omega$ can result in an asymmetric S-aS ratio through the interaction with the light. Consider an incident light wave polarized parallel to the plane of incidence. If the medium is absorptive so the wave vector of the light beam in the medium is complex, the polarization of the light will precess in the material. Such a precession is schematically illustrated in Fig. 2(c). When the sense of precession of the incident beam matches that of the spin wave, the scattering is greater.

RESULTS AND DISCUSSION

Here we present both experimental and theoretical results in studying the S-aS asymmetry in thin ferromagnetic films. Three materials were studied—Fe, Ni, and Permalloy. All three materials absorb light strongly (hence the wave vector for the light in the solid is complex), and all three materials show a large S-aS intensity asymmetry which cannot be explained by thermal effects, linear-quadratic interference, or localization of the surface wave.

In all the theoretical calculations presented here we have assumed that the surface spins are unpinned and have used only the linear terms in the spin density in the expansion of the dielectric constant. In Figs. 3(a) and 3(b), we present the experimental scattering spectra of the three materials under consideration. The plane of incidence is perpendicular to the surface and to the applied field. The incident light is polarized in the plane of incidence. $Q_{\parallel}$ is directed along the $+x$ axis. In Fig. 3(b) the magnetic field has been reversed with respect to Fig. 3(a). Figure 3(c) shows the theoretical calculation for the same geometry as Fig. 3(a).

In all three cases we find a large S-aS asymmetry which essentially reverses upon reversal of the magnetic field. This is true in particular for Fe, where the linear-quadratic mechanism can thus be neglected within experimental error. For Permalloy, and even more so for Ni, reversing the applied field does not exactly reverse the S-aS ratio. This is an indication that some linear-quadratic effect also plays a role here.

In Fig. 4(a) we plot the S-aS ratio as a function of applied field for Fe. We see here that as the field increases, the S-aS ratio also increases. The reason for this is that as the field increases, the spin deviations become more nearly circularly polarized and the contributions from the off-diagonal correlation functions become less important. In Fig. 4(b) we plot the S-aS intensity ratio of Fe as a function of the angle θ that the scattered light makes with the surface normal. The applied field is 0.1 T. We see here a strong dependence of the S-aS intensity ratio as a function of incident angle.

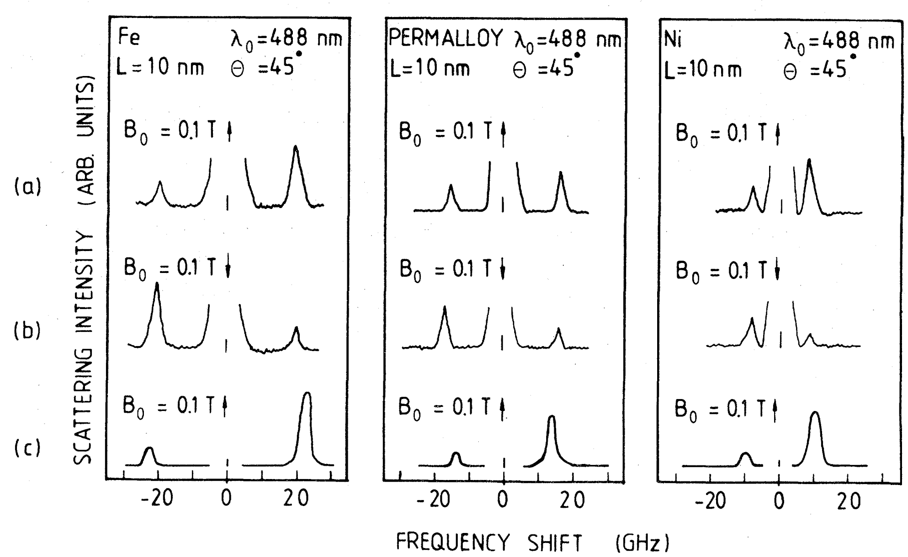

FIG. 3. Comparison of experimental, (a) and (b), and theoretical (c) results for light scattering from thin films of different materials. Thickness L is 10 nm, $\theta=45°$, and the incident light is polarized parallel to the plane of incidence.

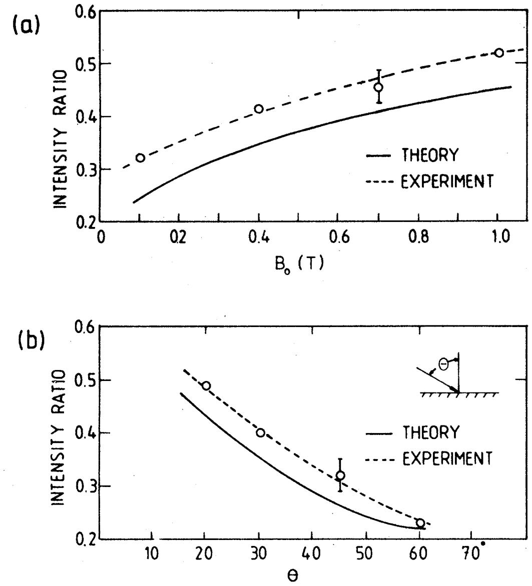

FIG. 4. (a) S-aS ratio as a function of the applied field for light scattering from 10-nm Fe film. $\theta=45°$. (b) S-aS ratio as a function of angle θ for light scattering from 10-nm Fe film. Applied field is 0.1 T.

As θ increases, the asymmetry between the intensities also increases.

We note that in the comparison of theory to experiment in Figs. 4(a) and 4(b) the qualitative behavior of the S-aS intensity ratio as a function of field and angle θ has very good agreement. The small quantitative differences are probably caused by the large collection angle in the experiment, and that quadratic coupling has been neglected in the theoretical calculations.

In the above study we have seen that the S-aS asymmetry in the light scattering from surface waves in thin ferromagnetic films can be explained by the contribution of the off-diagonal correlation function $\langle S_x S_y \rangle$. As we noted previously, the same mechanism will also result in an asymmetry in scattering from bulk waves. This in fact has been seen before[8,9,14] but was not explicitly recognized. As we have seen, the S-aS asymmetry found here is strongly angle dependent, and the angle-dependent asymmetry seen previously in the earlier studies is also due to the contribution of the correlation function $\langle S_x S_y \rangle$.

ACKNOWLEDGMENT

One of us (R.E.C.) wishes to acknowledge the Max Planck Institut für Festkörperforschung where the work was performed.

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