revista mexicana de física 26 no. 2 (1979) 311-327

SURFACE SPIN FLUCTUATIONS AND LIGHT SCATTERING FROM FERROMAGNETIC SURFACES*

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RESUMEN

En este artículo discutimos el efecto de una superficie sobre el espectro de ondas de spin de un material ferromagnético. Se hace énfasis en la región espectral donde las interacciones de intercambio (exchange) y dipolar contribuyen de manera significante a la energía de excitación de las ondas. Experimentos recientes de dispersión de la luz exploran la respuesta superficial de ferromagnetos en esta región. Se revisa una teoría reciente de dispersión de la luz desde superficies magnéticas.

ABSTRACT

In this paper, we discuss the effect of a surface on the spin wave spectrum of a ferromagnetic material. The emphasis is on the wavelength regime where both exchange and dipolar interactions contribute signicantly to the excitation energy of the waves. Recent light scattering experiments probe the surface response of ferromagnets in this regime. A recent theory of light scattering from magnetic surfaces is reviewed.

I. INTRODUCTION

In recent years, a variety of experimental methods have been used to probe or excite surface waves on solids. There are numerous classes of surface waves, from acoustical modes(1) (Rayleigh waves) with frequency in the microwave region or below, the surface polaritons with frequency that may range from the infrared up to the ultraviolet(2).

The purpose of the present paper is to discuss the very striking and unusual properties of a surface wave of magnetic character which can propagate along the surface of a ferromagnet, when the magnetization

† Research supported by the U.S. Army Research Office, Durham, under ARO No. DAA 29-77-G-0101.
Technical Report No. 78-70.

lies parallel to the surface. While this wave has been known to theorists for nearly twenty years[3], as far as the present authors are aware it has been studied experimentally only very recently for a geometry that may be regarded as well approximating the semi-infinite geometry assumed in the simple theories. Early studies of ferromagnetic resonance in thin films deal with absorption lines associated with excitation of the surface wave, but the spectra are influenced strongly by the finite thickness of the film[4]. Within the last year, very beautiful light scattering studies of this mode have appeared[5]. In contrast to the microwave studies, the light scattering experiment probes the surface with photons of wavelength very short compared to the linear dimensions of the sample. Also, the experiments are performed with exciting radiation absorbed strongly by the substrate. The incident photon thus samples only a thin layer within an optical skin depth of the surface.

In Section II of the present paper, we review properties of spin wave excitations in ferromagnets, both in the bulk and on the surface. Here we summarize some recent work of the present authors which examines the influence of exchange interactions on the long wavelength response of the surface region of the ferromagnet. In Section III we discuss the light scattering experiments, and summarize a recent theory of this phenomenon formulated by us.

II. SURFACE AND BULK SPIN WAVES IN FERROMAGNETS

It is well known that in the study of the lattice vibrations of crystals, a convenient and rigorous description of the vibrations of harmonic lattices proceeds by introducing quantized waves of excitation called phonons. In a ferromagnet, at temperatures well below the Curie temperature, where the spins are aligned to produce a saturation magnetization $M_S(T)$ at finite temperature T close in value to the magnetization $M_S(0)$ at T = 0, one introduces quantized waves of excitation of the spin system similar in many respects to the phonons of lattice dynamics. These waves are called magnons, or spin waves. In Figure (1), we provide an illustration of the motion executed by the spin system, when a spin wave is excited. Each spin processes in an elliptical orbit as the wave passes through the spin system. If the wave vector of the spin -

wave is denoted by $\vec{k}$, then for the projections $\Delta S_x(\vec{\ell},t)$ and $\Delta S_y(\vec{\ell},t)$ of the spin vector $\vec{S}$ of lattice site $\vec{\ell}$ onto the xy plane we have

$$\Delta S_x(\vec{\ell},t) = \Delta S_x \cos(\vec{k}\cdot\vec{\ell} - \Omega(\vec{k})t) \qquad \text{(II.1a)}$$

and

$$\Delta S_y(\vec{\ell},t) = \Delta S_y \sin(\vec{k}\cdot\vec{\ell} - \Omega(\vec{k})t) \qquad \text{(II.1b)}$$

where for elliptical motion, $\Delta S_x \neq \Delta S_y$.

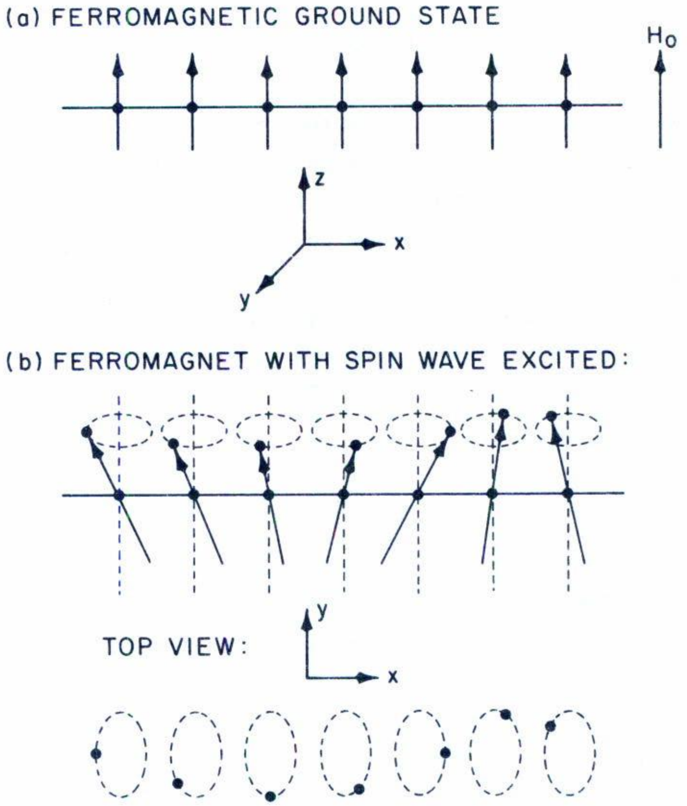

Fig. 1. (a) The spins fully aligned in the ferromagnetic ground state. The external magnetic field H_0 orients the magnetization.
(b) A sketch of the spin motion when a spin wave is -- excited.

To calculate the frequency $\Omega(\vec{k})$ of the wave, we need to know the nature of the interactions between spins in the system. For our purposes, we are concerned with two interactions of distinctly different character. These are

(i) The dipole-dipole interaction. If we have two spins $\vec{S}_1$ and $\vec{S}_2$ separated by the distance $\vec{r}_{12}$ with magnetic moment μ

$$V_{12}^{(D)} = \mu^2 \left\{ \frac{3(\vec{r}_{12}\cdot\vec{S}_1)(\vec{r}_{12}\cdot\vec{S}_2) - r_{12}^2\vec{S}_1\cdot\vec{S}_2}{r_{12}^5} \right\} \tag{II.2a}$$

and

(ii) The exchange interaction

$$V_{12}^{(x)} = - J\, \vec{S}_1\cdot\vec{S}_2 \quad . \tag{II.2b}$$

The exchange interaction, of quantum mechanical origin, is strong and of short ranged. It acts only between nearest and next nearest neighbors, in most magnetic materials. The dipole-dipole interaction is very much weaker than the exchange in general, but plays and important role because of its long range. As one sees from Eq.(II.2a), $V_{12}^{(D)}$ falls off slowly with increasing distance, like r_{12}^{-3} .

From these two interactions, one may compute the frequency $\Omega(\vec{k})$ of the spin wave of wave vector $\vec{k}^{(6)}$. If γ is the gyromagnetic ratio, then

$$\Omega(\vec{k}) = \gamma \left[(H + Dk^2)\ (H + 4\pi M_s \sin^2\theta + Dk^2)\right]^{1/2} \tag{II.3}$$

where H is the strength of an external magnetic field applied parallel to the magnetization, M_s is the saturation magnetization, D is a measure of the strength of the exchange, and θ the angle between $\vec{k}$ and the magnetization. Thus, the frequency $\Omega(\vec{k})$ depends on the *direction* as well as the magnitude of the wavevector.

There are two distinct limiting regimes that follow from Eq. (II.3). Quite generally, H and $4\pi M_s$ are comparable in magnitude and equal to a few hundred or a few thousand gauss. If the wavelength is so long that $Dk^2 \ll H$ or $4\pi M_s$, then the frequency $\Omega(\vec{k})$ is dependent on the *direction* of $\vec{k}$, but not its magnitude. The excitation energy of the spin wave has its origin in precession about the Zeeman field, and in the macroscopic fields set up by virtue of the dipole-dipole interaction. We refer to this as the *dipole dominated* regime. In the opposite limit $Dk^2 \gg H$, $4\pi M_s$, we have $\Omega(\vec{k}) \simeq \gamma Dk^2$. Here, in the *exchange dominated regime*, the frequency increases quadratically with wave vector $k^{(7)}$, and is independent of direction.

In the next portion of the discussion, we shall focus attention on the dipole dominated regime, where the role of exchange interactions may be ignored. Then as we have seen, $\Omega(\vec{k})$ depends on the direction of $\vec{k}$. If $\theta = 0$, so the wave propagates parallel to the magnetization, then from Eq. (II.3) $\Omega(\vec{k}) = \gamma H$. This is the Larmor frequency of precession of a free spin. As θ is increased, $\Omega(\vec{k})$ increases to the value $\Omega(k) = \gamma(HB)^{1/2}$ at $\theta = \pi/2$, where $B = H + 4\pi M_s$. Thus, in the absence of exchange, all the bulk waves lie in the frequency regime $\gamma H \leq \Omega(\vec{k}) \leq \gamma(HB)^{1/2}$. We shall refer to the band of frequencies between γH and $\gamma(HB)^{1/2}$ as the *bulk spin wave manifold*.

Under a variety of conditions, surface spin waves may propagate along the surface of the ferromagnet. These are waves of magnetic excitation quite similar to ocean waves. All spins which lie in a given plane parallel to the surface precess on an ellipse of constant size, while the size of the ellipse decreases exponentially with distance in from the surface. The situation is illustrated in Fig. (2). When the surface spin wave is excited, the pattern of spin deviations (in certain simple cases) may be described by the expressions

$$\Delta S_x(\vec{\ell},t) = \Delta S_x \cos(\vec{k}_\parallel\cdot\vec{\ell}_\parallel - \Omega_s(\vec{k}_\parallel)t)\exp(-\alpha(\vec{k}_\parallel)\ell_z) \qquad \text{(II.4a)}$$

and

$$\Delta S_y(\vec{\ell},t) = \Delta S_y \sin(\vec{k}_\parallel\cdot\vec{\ell}_\parallel - \Omega_s(\vec{k}_\parallel)t)\exp(-\alpha(\vec{k}_\parallel)\ell_z) \qquad \text{(II.4b)}$$

Here $\vec{\ell}$ is the projection of $\vec{\ell}$ onto a plane parallel to the surface and ℓ_z is the distance of site $\vec{\ell}$ in from the surface. Also, $\vec{k}_\parallel$ lies in the plane of the surface, and $\Omega_s(\vec{k}_\parallel)$ is the frequency of the wave of wave vector $\vec{k}_\parallel$.

The properties of surface spin waves are most striking in the dipole-dominated regime, where the modes are called the Damon-Eshbach waves.

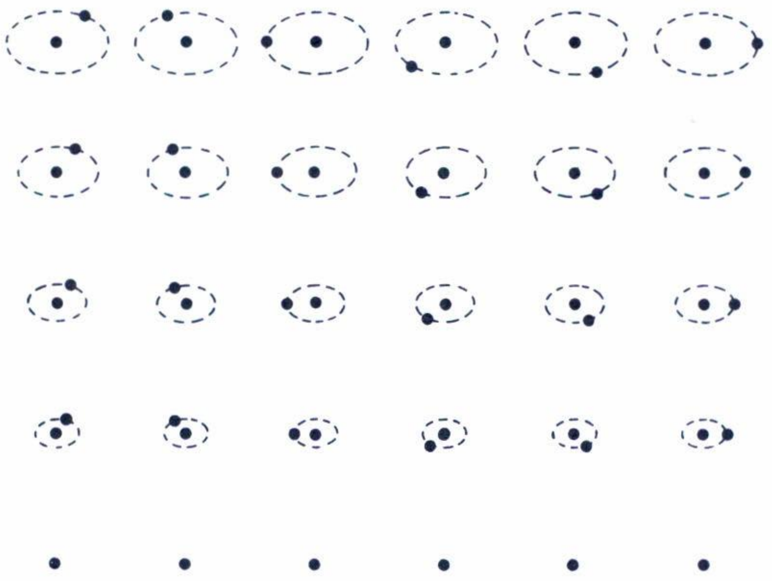

Fig. 2. A sketch of the pattern of spin deviations present when a surface spin wave is excited.

A most unusual feature is that the waves can propagate only in one direction on the surface; if one faces the crystal with the magnetization directed upward, the surface waves may propagate from left to right, but not from right to left. More precisely, if θ is the angle made by $\vec{k}_{\parallel}$ with the x axis, and the magnetization is along $\hat{y}$, the waves may propagate for $-\theta_c \leq \theta \leq +\theta_c$, where $\cos\theta_c = (H/B)^{1/2}$. The situation is illustrated in Fig. (3). We thus have unidirectional propagation, where energy may flow in the surface wave in only the one sense described above.

In the dipole dominated regime, the frequency of the surface wave $\Omega_s(\vec{k}_{\parallel})$ is

$$\Omega_s(\vec{k}_{\parallel}) = \frac{\gamma}{2}\left[\frac{H}{\cos\theta} + B\cos\theta\right] , \qquad \text{(II.5)}$$

where the angle θ is given in Fig. (3). For $\theta = 0$ (propagation perpendicular to the magnetization), we have $\Omega_s = \gamma(H + B)/2$, and as θ increases toward θ_c, the frequency $\Omega_s(\vec{k}_{\parallel})$ decreases to assume the value $\gamma(HB)^{1/2}$ when $\theta = \theta_c$.

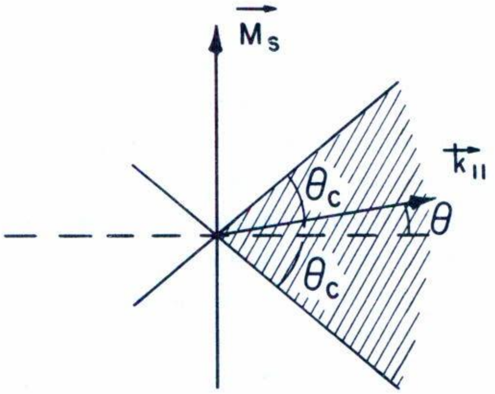

Fig. 3. The shaded area of the figure indicates the regime of angles where the Damon-Eshbach surface spin wave may propagate.

For all values of θ allowed for surface wave propagation, a comparison between Eq. (II.5) and Eq. (II.3) shows that in the dipole dominated regime the frequency of the surface wave lies *above* the bulk spin wave manifold. If we then examine the influence of exchange, one sees that bulk waves become degenerate in frequency with the surface wave. This occurs through action of the Dk^2 terms in Eq. (II.3) which can upshift the frequency of the surface wave, to make it coincide with that of the Damon-Eshbach wave. In this circumstance, it is not possible to sustain a true surface mode. If we propagate a surface wave down the system, the energy in the surface wave can "leak" into the bulk of the material; in essence the surface wave radiates its energy away in the form of bulk surface waves. One calls such a mode a "leaky surface wave" elsewhere in the literature.

Mathematically, when exchange is added, the spin deviation associated with the surface wave no longer has the simple form given in Eqs. (II.4). A full description of the wave requires three exponential functions $\exp|-\alpha_i(\vec{k}_\parallel)\ell_z|$ to be superposed. One of the $\alpha_i(\vec{k}_\parallel)$'s is pure imaginary, and this portion of the wave describes the "radiation" carried off to the crystal interior.

We have recently completed a detailed and quantitative study of the influence of exchange on the Damon-Eshbach wave. The full details can be found elsewhere[8]. Here we confine our attention to a summary of the principal results.

One must first inquire *how* one is to study the radiative life-time of the surface wave in the presence of exchange. We chose a method different than that commonly employed in the literature, but which we find quite unambiguous and clear. By means of a method developed in Reference (8), we construct a spectral density function $A(\vec{Q}_\parallel, \Omega; z)$ with the following physical interpretation. Suppose one penetrates a distance z into the material from the surface, and examines a slab of thickness d_z parallel to the surface. If a Fourier decomposition of the spin fluctuations within the slab is made, then $A(\vec{Q}_\parallel \Omega; z)$ measures the amplitude of spin fluctuations with wave vector $\vec{Q}_\parallel$ and frequency Ω.

We may now study the surface wave as follows. Pick a value of z, say $z = 0$ which corresponds to sitting right at the surface. Pick a value of $\vec{Q}_\parallel$ and scan the frequency Ω. The Damon-Eshbach wave appears as a peak in $A(\vec{Q}_\parallel \Omega; z)$, as we shall see. The position of the peak as a function of $\vec{Q}_\parallel$ gives us the dispersion relation, and a study of its width as a function of the exchange constant D gives us the radiative contribution to the lifetime of the wave. We have carried out an extensive set of such calculations on a computer; one uses the computer to probe the response of the system very much as the experimentalist does in the laboratory.

In Fig. (4), we present a summary of some of our studies of the spectral density function. The calculations presume the ratio B/H to be 7, and we have a damping term in the equations of motion also. The relaxation time τ that enters the equations of motion has been chosen so that $(\gamma H \tau)^{-1} = 0.01$. Thus, even in the absence of the radiative damping described above, each mode will have a finite lifetime by virtue of this damping term. The above value of τ is typical of that found in ferromagnetic resonance experiments; we may then examine the conditions under which the radiation damping dominates the dissipation in the spin system.

In Fig. (4), a measure of the influence of exchange is provided by the parameter $\omega_x = D\, k_\parallel^2/\gamma H$, where $k_\parallel$ is the wave vector of the surface wave parallel to the surface. In Fig. (4a), we plot the spectral density for $\omega_x = 0.01$ and various values of the propagation angle θ in Fig. (3). For θ in the range $0 < \theta < 0.3\pi$, the radiation damping plays little

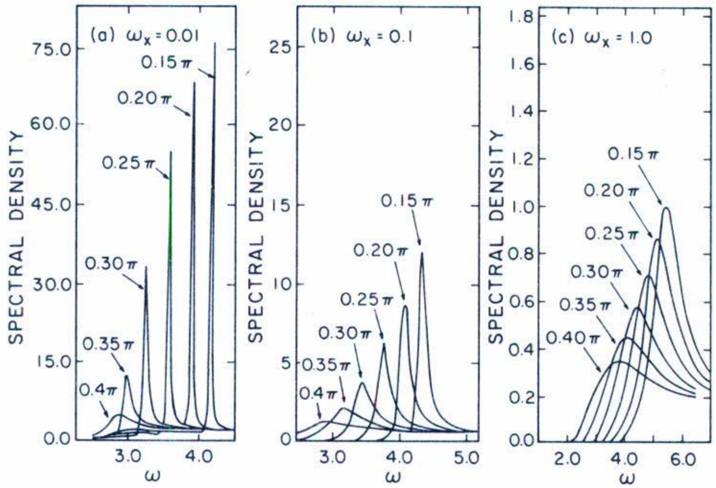

Fig. 4. We show here the spectral density for surface spin fluctuations for various propagation angles and (a) $\omega_x = 0.01$ (b) $\omega_x = 0.1$ and (c) $\omega_x = 1.0$. The parameter $\omega_x = D k_\parallel^2/H$, and frequency is measured in units of γH, i.e. ω is the spin wave frequency in units of γH. The calculations are performed for the ratio $B/H = 7$.

role. As θ increases to $\theta_c = 0.37\pi$, and the surface mode frequency sinks toward the top of the bulk spin wave manifold, it is evident that the surface wave peak broadens substantially, as one sees from the curve labeled $\theta = 0.35\pi$. This is the effect of the radiation damping. For $\theta = 0.40\pi$, and angle beyond θ_c, one sees a remnant of the surface mode in the form of a resonance within the bulk spin wave manifold. We believe these spectral density studies provide a vivid picture of the manner in which the Damon-Eshbach wave merges with the bulk continuum.

In Fig. (4b) and Fig. (4c), we show the effect on the spectral density of increasing the exchange. For $\omega_x = 0.1$, where the exchange interactions contribute very little to the excitation energy of the mode, the radiation damping is quite severe for all propagation angles. When

$\omega_x = 1.0$, so exchange, dipolar and Zeeman contributions to the excitation energy are comparable in magnitude, in place of the well defined surface wave peak of Fig. (4a), we have a broad heavily damped resonance level.

In recent light scattering studies of ferromagnetic surfaces, as remarked earlier, the Damon-Eshbach wave has been studied under conditions (in one case) which approximate those envisioned in the above calculations. We turn to a discussion of these experiments in the next section.

III. LIGHT SCATTERING FROM SPIN WAVES AT MAGNETIC SURFACES

After examining a number of properties of the Damon-Eshabach spin wave in the preceding section, it is natural to inquire into how one may probe or excite this wave. It is of particular interest to excite waves with wavelength sufficiently short for the radiative leak to the crystal interior to be appreciable.

A traditional way to excite spin waves is to use a thin film of ferromagnetic material placed in a microwave cavity. If the film has thickness L and we think of only bulk spin waves for the moment, these waves have quantized wavevector in the direction perpendicular to the film given by $k_\perp \approx n\pi/L$, where $n = 0, 1, 2....$ The experiments proceeds by operating the cavity at fixed frequency, then bring the various spin waves into resonance with the microwave field through changing an external magnetic field.

Modes excited by this technique have wave vector $\vec{k}_\parallel$ parallel to the surface very close to zero, simply because the microwave field will be very nearly constant everywhere within a small sample. The Damon-Eshbach mode has been studied in such experiments[4], but the attenuation constant $\alpha(\vec{k}_\parallel)$ of Eqs.(II.4) is of order L^{-1}, i.e. the wave is really a geometrical resonance of the sample as a whole, and not a mode tightly bound to the surface of a sample that is effectively semi-infinite in extent.

In recent months, a new experimental method has been employed to excite bulk spin waves and also the Damon-Eshbach wave at the surface of a ferromagnet. This is the inelastic scattering of light (Brillouin scattering) from spin waves at the surface. The experiment is illustrated in Fig. (4). An incident photon with frequency Ω_o and wave vector $\vec{k}^{(o)}$ is incident on the sample surface. By means of a mechanism described below, the incident photon "sees" thermally excited spin waves, and can scatter off them inelastically. The photon scatters from a surface spin wave (or bulk spin wave) with wave vector in the plane of the surface equal to $\vec{k}_\parallel$. The scattered photon emerges with either the frequency --- $\Omega_o + \Omega_s(\vec{k}_\parallel)$ and wave vector $\vec{k}_\parallel^{(o)} + \vec{k}_\parallel$ parallel to the surface (anti-Stokes scattering) or with frequency $\Omega_o - \Omega_s(\vec{k}_\parallel)$ and wave vector $\vec{k}_\parallel^{(o)} - \vec{k}_\parallel$ (Stokes scattering). Detection of the frequency and direction of the scattered photon thus provides both $\Omega_s(\vec{k}_\parallel)$ and $\vec{k}_\parallel$.

In the experiments reported to date scattered photons which emerge antiparallel to the incident photon are detected, as illustrated in Fig. (5). If a Damon-Eshbach wave is created, it has wave vector $\vec{k}_\parallel$ given by $2\Omega_o \sin\theta_o/c$, where θ_o is the angle of incidence. For visible light

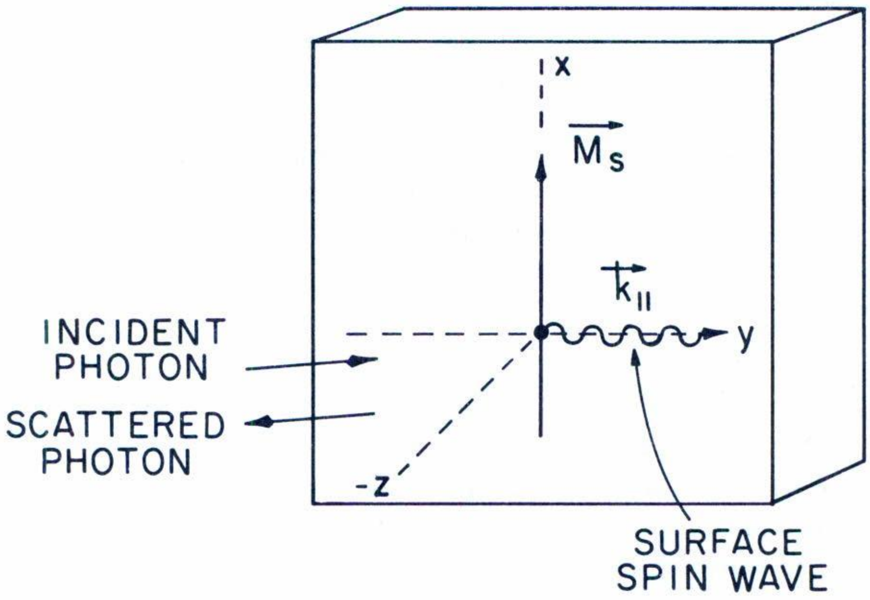

Fig. 5. The geometry employed in the light scattering studies of EuS by Grunberg and Metawe, and of Fe and Ni by Sandercock and Wettling.

incident on the surface, one has $k_\parallel \approx 10^5 cm^{-1}$, several orders of magnitude larger than in the microwave studies. One is safely in the regime where the sample may be presumed semi-infinite. For the ferromagnetic metals, which have very large values of the spin wave exchange stiffness constant D, we estimate the ratio $\omega_x = D\,k_\parallel^2/H$ of Fig. (4) to be large enough for the radiation damping to be observed in the linewidth. We are thus in a most interesting parameter regime.

The experiments are also performed under conditions where the substrate is opaque to the incident radiation. In the Eu chalcogenides, radiation beyond the absorption edge is used, to give a skin depth $\approx 1500\text{Å}$. In Fe and Ni, the skin depth is $\approx 150\text{Å}$ for visible radiation. For these small skin depths, the Damon-Eshbach surface spin wave is a strong feature in the spectrum, not swamped by the signal from the bulk spin waves.

We have recently formulated a detailed theory of light scattering from spin waves at ferromagnetic surfaces, to obtain a quantitative description of the experiments outlined above. The theory is described in detail elsewhere. Here we outline its principal features.

The first question to address is how the incident light "sees" the thermal fluctuations in the spin system. As the spins fluctuate, the dielectric tensor of the material also fluctuates. We are concerned here with spin fluctuations of wavelength long compared to the lattice constant, so these effects may be described through use of a continuum theory. Let $\delta\varepsilon_{\mu\upsilon}(\vec{x},t)$ be the fluctuating part of the dielectric tensor, and $S_\lambda(\vec{x},t)$ the spin density at point $\vec{x}$ and time t. In continumm theory we write

$$\delta\varepsilon_{\mu\upsilon}(\vec{x},t) = \sum_\lambda K_{\mu\upsilon\lambda} S_\lambda(\vec{x},t) + \sum_{\lambda\delta} G_{\mu\upsilon\lambda\delta}\, S_\lambda(\vec{x},t) S_\delta(\vec{x},t) \qquad \text{(III.1)}$$

where a particularly complete discussion of the coefficientes $K_{\mu\nu\lambda}$ and $G_{\mu\upsilon\lambda\delta}$ was presented a number of years ago by Landau and Lifshitz [(10)]. While $G_{\mu\upsilon\lambda\delta}$ is symmetric under interchange of μ and υ, $K_{\mu\upsilon\lambda}$ is antisymmetric under interchange of these indices.

The contributions to the first term ($K_{\mu\upsilon\lambda}$) on the right hand side of Eq. (III.1) that come from $S_x(\vec{x},t)$ and $S_y(\vec{x},t)$ describe scattering events in which a single bulk or surface spin wave is created, as do the terms in the second term ($G_{\mu\upsilon\lambda\delta}$) in S_zS_x and S_zS_y. We thus have two physically distinct terms which couple the incident photon to spin waves. These terms interfere coherently, to produce some striking features in the spectrum noted below.

The calculation of the spectrum of scattered light is presented in detail in Reference (9). The procedure is as follows. One writes down Maxwell's equations for a material with dielectric constant $\varepsilon_{\mu\upsilon}(\vec{x}t) = \varepsilon\,\delta_{\mu\upsilon} + \delta\varepsilon_{\mu\upsilon}(\vec{x},t)$. Here ε is the complex dielectric constant of the substrate in the absence of spin fluctuations, and $\delta\varepsilon_{\mu\upsilon}(\vec{x},t)$ is the fluctuating part described in Eq. (III.1). In our work, in the interests of simplicity, we take the time independent term to be isotropic, even below the Curie temperature where the spontaneous magnetization imparts a gyrotropic character to the dielectric constant. This approximation is readily lifted, if desired.

We then proceed by treating the scattering produced by the fluctuating part $\delta\varepsilon_{\mu\upsilon}(\vec{x},t)$ of the dielectric tensor by methods of scattering theory, to find the component of scattered light first order in $\delta\varepsilon_{\mu\upsilon}(\vec{x},t)$. The light scattering efficiency can be found, and related to the same correlation functions $\langle S_\mu(\vec{x}t)S_\upsilon(\vec{x}'t')\rangle$ examined in the studies of the spectral density function summarized in Section II of the present paper. The shape of the spectra are in excellent accord with the data, and here we present a summary of some of our theoretical spectra.

In Fig. (6), we show a light scattering spectrum for scattering from the surface of Fe. A magnetic field of 2 Koe has been presumed present, while the incident light makes an angle of 18° with the normal to the surface. The wave vector $\vec{k}_\parallel$ is perpendicular to the magnetization. Frequencies are measured in units of γH in the figure, so the frequency shifts of the light are in the GHz range.

There are two striking features in the spectrum. The first is the dramatic asymmetry between the Stokes and the anti-Stokes side of the spectrum. In most circumstances encountered in Brillouin spectroscopy, when the frequency shift Ω of the scattered light is very small compared to $k_BT/\hbar$, a condition very well fulfilled here, the Stokes and anti-Stokes spectra form mirror images of each other. In the present case, the intense line on the anti-Stokes side of the spectrum centered about the dimensionless frequency -7 is the scattering from the Damon-Eshbach surface spin wave mode. This line is missing from the Stokes side of the spectrum.

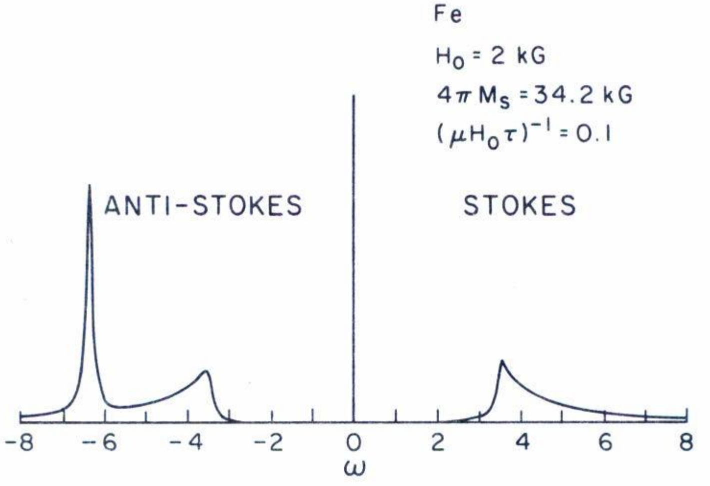

Fig. 6. A theoretical calculation of the Brillouin spectrum for scattering from spin waves at the surface of Fe. The angle of incidence is 18°, and the wave vector transfer is perpendicular to the magnetization. Frequency is measured in units of γH. A more detailed description of this material can be found in Reference (8).

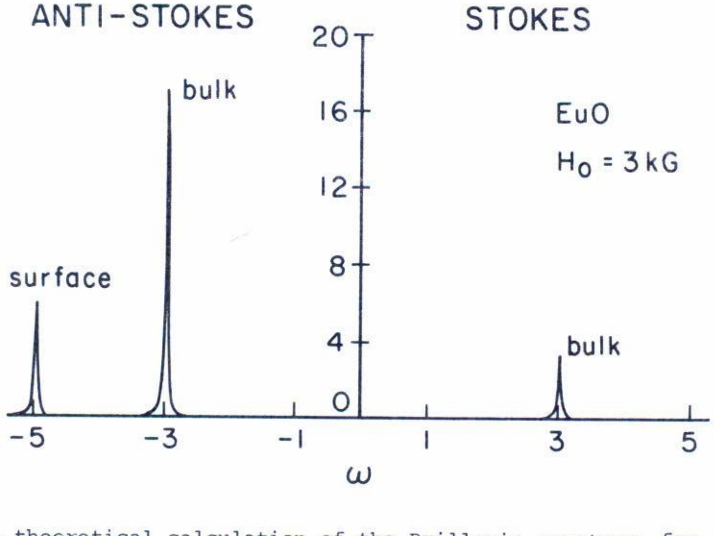

Fig. 7. A theoretical calculation of the Brillouin spectrum for scattering from spin waves at the surface of EuO. The wave vector transfer is perpendicular to the magnetization. The angle of incidence is 45°, and frequency is measured in units of γH. A more detailed description of this material can be found in Reference (9).

One can see that this is a direct consequence of the "one-sided" nature of the Damon-Eshbach surface wave dispersion relation. If we consider a fixed scattering geometry, with the *direction* of the incident and scattered photon both fixed, then a surface mode which participates in a Stokes process (spin wave created in the scattering event) necessarily has wave vector opposite in sign to one which participates in an anti-Stokes event (spin wave destroyed in the scattering event). If $\vec{k}_\parallel$ lies within the shaded regions allowed for propagation of the Damon-Eshbach wave (see Fig. (3)), then $-\vec{k}_\parallel$ lies outside. Thus, if the surface spin wave appears on the anti-Stokes side of the spectrum, as in Fig. (6), it is necessarily absent from the Stokes side. If the magnetic field is reversed in sign, with a reversal of the magnetization direction as a consequence, the surface spin wave pops over from the anti-Stokes to the Stokes side of the spectrum. That this is so is verified in the experiments[5].

A second striking feature in the spectrum displayed in Fig. (6) is the broad, asymmetric band that sets on at a frequency of ≈ 3.5 on both sides of the spectrum,and falls off slowly as the frequency increases. The origin of this part of the spectrum is scattering from bulk spin waves. One sees a broad spectrum from these modes rather than a well defined line, because in the presence of a finite skin depth, wave vector components normal to the surface are no longer conserved in the scattering event. Bulk spin waves with wave vectors as large as the inverse of the skin depth δ may then participate in the scattering[11]. Spin waves with wave vectors as large as this have excitation energies influenced importantly by the exchange contributions $D\,k^2$ in Eq. (II.3). It is this high frequency "exchange tail" that produces the radiative damping discussed in Section II. The light scattering spectra thus reveal the overlap in frequency between the bulk spin wave band and the Damon-Eshbach wave that is responsible for this effect. Unfortunately, the experimental spectra reported to date do not have sufficient resolution to measure the intrinsic width of the Damon-Eshbach mode. We hope that further experimental work will allow direct study of the leaky - nature of the Damon-Eshbach surface mode.

In Figure (7), we show our calculated spectra for EuO, which has been studied by Grunberg and Metawe. Again the surface spin wave feature in the spectrum appears on one side of the laser line only. The bulk spin wave features now appear as line structures, and the prominent high frequency tail present in Figure (6) is missing. There are two reasons why this is so in EuO. First of all, the skin depth in EuO is around 1500Å, a value an order of magnitude larger than that in Fe. Thus, the breakdown of wave vector conservation normal to the surface is much less severe in EuO, compared to Fe. Secondly, the exchange stiffness constant D in Fe is very much larger than in EuO; the fact that the Curie temperature of Fe is over 1000K, while that in EuO is only around 80K provides a rough calibration of the relative strength of the exchange interactions.

It is also noteworthy that there is a large asymmetry in the Stokes/anti-Stokes intensity in the bulk spin wave lines in the spectra. As pointed out sometime ago by Wettling, Cottam and Sandercock[12], the physical origin of this asymmetry is in the interference between the first and second terms on the right hand side of Eq. (III.1). Both terms contribute to the scattering cross section, as remarked earlier. One must add together the contribution from each to the *matrix element* for the scattering process before squaring the matrix element to form an expression for the final scattering cross section. The two terms interfere constructively on one side of the laser line, and destructively on the other.

We believe that the light scattering method described here will be a powerful means of probing spin fluctuations at magnetic surfaces. Future experiments should explore scattering from surface spin waves with wave vector titled away from the perpendicular to the magnetization. Such a geometry with improved resolution should allow direct examination of the leaky character of the Damon-Eshbach surface spin wave. In addition, any perturbation of the magnetic response in the near vicinity of the surface should be revealed through study of the frequency of the wave, and the temperature variation of its frequency.

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4. See T. Wolfram and R.E. De Wames, Progress in Surface Science 2 (1972) 233.
5. P. Grumberg and F. Metawe, Phys. Rev. Letters 39 (1977) 1561 and W. Wettling and J. Sandercock (to be published).
6. The general methods for doing this are discussed in Chapter 4 of C. Kittel, Quantum Theory of Solids (John Wiley and Sons, Inc., New York, 1963).
7. We suppose only wavelengths long compared to the lattice constant are of interest.
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9. R.E. Camley and D.L. Mills, Solid State Communications. 28 (1978) 321.
10. See p. 331 of Electrodynamics of Continuous Media, L.D. Landau and E.M. Lifshitz (Pergamon Press, Oxford, 1960).
11. See D.L. Mills, A.A. Maradudin and E. Burstein, Annals of Physics 56 (1970) 504 for a general description of the consequences of the break-down of wave vector conservation normal to the surface.
12. W. Wettling, M.G. Cottam and J.R. Sandercock, J. Phys. C8 (1975) 211.