Spin waves in amorphous media and thin films

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In the recent observations of spin-wave resonance in amorphous thin films, spin waves are found to propagate, and the resonance linewidth strongly depends on the spin-wave wavelength. The aim of this paper is the understanding of this dependence and its relation to the physical parameters, which are the exchange and anisotropy fluctuations. This is accomplished by means of a scattering theory that neglects conduction and takes advantage of the quasiselection rule of spin-wave resonance to become a one-dimensional theory. Long-wavelength behavior is dominated by anisotropy, while short-wavelength behavior is determined by exchange. The theoretical results qualitatively agree with the experimental results and define a measurement of the short-range fluctuations.

I. INTRODUCTION

In recent years several spin-wave-resonance (SWR) experiments have been performed on amorphous thin films[1-3] or on more or less strained thin films.[4] Because of the wave scattering by local inhomogeneities of various sizes and scattering intensities, one can expect poor propagation in such films. In SWR experiments as in a Fabry-Perot cell, the wave is reflected a large number of times on the external surfaces. Poor propagation can change the effective surface conditions, so one can expect a dispersion of the resonance condition, i.e., a frequency linewidth W for the modes of frequency ω and an enhancement of the even modes which are forbidden for perfectly symmetric surfaces.[5] The two features, linewidth W and enhancement of even modes, are observed and depend on the frequency. If one follows the analogy with light scattering by impurities one can observe a transition from a red sun in a blue sky to a gray sun in a gray sky according to the density of defects. In the samples considered by the previously quoted authors, the defects, which are typical of an amorphous structure, are probably short ranged because of the good uniformity. The typical curve of frequency linewidth W versus the perpendicular wave vector q, or its quantum number n defined from the film thickness d by $q=n\pi/d$, gives a first maximum value at $n=0$, then a decrease with increasing n, a minimum value for some values of n, then an increase with increasing n, followed by a saturation value. Even for the highest n or q observed in SWR, the wavelength is large in comparison to the atomic radii and the defect size, so the scattering theory one works with has no size resonance and is restricted to small defects. This defines the aim of this paper, the understanding of the curve $W(n)$ on a microscopic basis. The observed samples have different conduction properties, so one can omit the extra conduction contribution to the linewidth W in the first approximation. Thus the main parameters are the exchange and anisotropy fluctuations ΔJ and ΔI, respectively. Firstly, it seems useful to recall the basic results of Kittel's theory of SWR (Ref. 5) and the derived fluctuation treatment of W (Refs. 3 and 6) when one introduces the surface phase shifts related to the surface conditions. This gives a simple law W proportional to n^2, in poor agreement with experimental results. Secondly, we directly consider the scattering equations of spin waves, specializing for one small-sized impurity and a plane spin wave according to the quasiselection rule of SWR. This looks like the scattering by a barrier related to the impurity. Thus there appears a new phase shift $\Delta\phi$ which modulates the total one and a reflection term which determines a damping and so a coherence length. Both the fluctuations of the total phase shift and the finite coherence length contribute to the spin-wave linewidth W. For low n, i.e., long wavelength, the scattering is dominated by the anisotropy fluctuations. The notion of coherence length which allows us to interpret the saturation effect is similar to that of Anderson's localization of electrons in amorphous materials[7] which is due to a strong scattering by local barriers. The four parts of the $W(n)$ curve are considered in agreement with the experimental results. Moreover, the resulting phase-shift deviation explains the appearance of even modes in SWR when the surface conditions are the same, as is observed experimentally. This allows a check of the measurements of ΔJ and $\Delta(ZI)$.

Section I A is devoted to the first approach, while Sec. I B deals with the scattering theory, before the concluding remarks.

A. From Kittel's theory of the spin-wave frequency E, the linewidth W in amorphous thin films

Because of the inhomogeneities, spin waves propagate poorly in such films and can be considered as almost localized. So in different parts of the sample, spin waves experience different exchange and anisotropy couplings. This picture can be completed more quantitatively as it will be in Sec. I B, but here it is enough to introduce a simple model. The inhomogeneities lift the frequency degeneracy, and a resonance linewidth W appears. So the simple model consists in using the classical results of SWR (Ref. 5) and considers the causes of fluctuation. With a for interatomic distance and J for exchange integral, the resonant magnetic field H along film normal for the nth spin-wave mode occurs when

$$\omega_0-\lambda H=Ja^2d^{-2}(\phi+n\pi)^2 \tag{1}$$

is satisfied, where ω_0 is the radio frequency, $\phi=\phi_2-\phi_1$ is the total phase shift due to the two external surfaces, and λ is the gyromagnetic factor. Three effective causes of variation of the resonant field H can be outlined: the exchange constant Ja^2, the thickness d, and the phase shift ϕ. So one can derive the obvious "error" formula for the linewidth $W=\lambda\Delta H$:

$$W=\frac{Ja^2}{d^2}(\phi+n\pi)^2\left[\frac{\Delta(Ja^2)}{Ja^2}-2\frac{\Delta d}{d}+2\frac{\Delta\phi}{\phi+n\pi}\right]. \tag{2}$$

The surface phase shift ϕ is usually proportional to n and to the effective surface anisotropy I (Ref. 8). Therefore at this level of approximation $\Delta\phi$ is proportional to n too and to ΔI. Thus the total linewidth W_n looks like

$$W_n=W_1n^2\ , \tag{3a}$$

with

$$W_1=\frac{Ja^2}{d^2}(\phi_1+\pi)\left[\frac{\Delta(Ja^2)}{Ja^2}-\frac{2\Delta d}{d}+2\Delta\phi_1\right]. \tag{3b}$$

Unfortunately, this simple law is not followed. One can argue that the sizes of the decoupled domains depend on the wave mode n, i.e., on the wave vector $q=n\pi/a$, but this is already beyond the scope of this simple model, and it is better to consider a more detailed one in order to derive W directly.

B. One-dimensional model

Here, as in the preceding part, the size and scattering intensity of the scatterers are assumed to be small enough so that the two-dimensional extension of the spin wave in the planes parallel to the surfaces is larger than d^2. In that case it is well known[9] that the only spin waves observed by SWR have zero wave-vector components in the plane q_xq_y reciprocal to the surface plane xy. In other words the involved spin waves propagate along the z axis which is normal to the specimen and parallel to the magnetization and the external field. This is the reason for the restriction to a one- dimensional problem in the z direction. Before going on to a continuum picture more suitable for amorphous structures, one has to recall the magnetic Hamiltonian $\mathscr{H}$ with exchange integral J, uniaxial anisotropy constant I, and Zeeman effect measured by h':

$$\mathscr{H}=-\tfrac{1}{2}\sum J_{ij}S_iS_j-\tfrac{1}{2}\sum_{ij} I_{ij}S_i^zS_j^z-\sum h'S_i^z\ . \tag{4}$$

With the usual definition of Pauli operators b and $b^\dagger$:

$$b_f=S_f^x+iS_f^y\ ,\quad b_f^\dagger=S_f^x-iS_f^y; \tag{5}$$

when one takes into account their commutation relations, the equation of motion of b_g reads

$$i\frac{db}{dt}f=\sum_g J_{fg}\frac{b_g-b_f}{4}+\left[h'+\sum_g\frac{I_{fg}}{2}\right]b_f\ . \tag{6}$$

It is useful to introduce a simple local definition of J_M and I_M when the interaction is restricted to nearest neighbors as here:

$$J_{M=(f+g)/2}=J_{f,g},\quad I_{M=(f+g)/2}=I_{f,g}\ . \tag{7}$$

When one defines the vector $\vec{\delta}$ linking nearest neighbors by

$$\vec{\delta}=\vec{g}-\vec{f} \tag{8}$$

and δ_α its coordinate along the α axis and labels the spatial derivations of b, J, and I by the respective lower indices, the equation of motion of b at frequency ω reads in this local picture

$$\left[h'+\frac{ZI}{4}+\sum_{g,\alpha}\frac{\delta_\alpha I_\alpha}{8}-\omega\right]b - \frac{J}{4}\sum_{g,\alpha}\delta_\alpha b_\alpha - \sum_{g,\alpha,\beta}\frac{\delta_\alpha\delta_\beta}{8}(J_\alpha b_\beta + J b_{\alpha\beta}) = 0 , \tag{9}$$

where the developments of J, I, and b_f have been restricted to the lower powers of the components of δ, and Z is the local number of nearest neighbors while the sum of these nearest neighbors is specified in $\sum_g$. A similar treatment leads to Portis's equation[10] by considering $J_{\alpha\alpha}$. If we assume a local center of symmetry in the amorphous structure, Eq. (9) reduces to

$$\left[h'+\frac{ZI}{4}-\omega\right]b-\sum_{g\alpha}\frac{\delta_\alpha^2}{8}(J_\alpha b_\alpha + J b_{\alpha^2})=0 . \tag{10}$$

One easily recognizes a wave equation with a damping term $\vec{\nabla} J\cdot\vec{\nabla} b$ written for a full rotational symmetry of the local sites. J_α is of order $\Delta J/a$, while b_{α^2}/b_α is of order $n\pi/d$, so the condition for undamped spin waves reads

$$\frac{\Delta J}{J}\frac{d}{a}\frac{1}{n}<<1 . \tag{11}$$

The higher n is, the more easily this condition is fulfilled.

As a matter of fact, three cases occur in the analysis of Eq. (10): overdamped spin waves, damped spin waves, and undamped spin waves. As we assume quite homogeneous samples and one-dimensional effects, i.e., an average over the planes parallel to the surfaces, the effective ΔJ are weak and the condition (11) is fulfilled, so we can neglect the damping term $(\partial J/\partial z)(\partial b/\partial z)$. On the other hand, as it has already been said in the Introduction, even in the case of these undamped equations we shall take into account an effective damping of the spin waves. So the following theory restricted to the undamped equation does not lose the generality of the damping phenomena.

The equation without damping and with a full local rotational symmetry reads

$$\left[h'+\frac{ZI}{4}-\omega\right]b-Je^2\nabla^2 b=0 , \tag{12a}$$

where

$$e^2=\sum_g\frac{\delta_g^2}{8} . \tag{12b}$$

One recognizes a Schrödinger equation for a particle of mass $(Je^2)^{-1}$ moving in a potential $h'+ZI/4$. The potential is rather weak, so the particle is nearly free, and the deviation from Kittel's law $\omega\sim q^2$ is weak. One can notice here that both inertial terms and potential terms are nonuniform in space. Obviously, the wave scattering will be dominated either by inertial (exchange) terms or by potential (anisotropy) terms.

Out of the film there is no spin, so the wave function is restricted to it, i.e., from $z=0$ to $z=d$. The spin boundary conditions on the surfaces can be interpreted in terms of spin pinning (9) which links $b(0)$ and $b_z(0)$ for the first surface. This spin pinning which is due to surface anisotropy and to surface exchange (8) introduces the surface phase shifts ϕ_1 and ϕ_2 (Ref. 8) in a quite obvious way:

$$\tan\phi=b(0)/b_z(0) .$$

In the film, spin waves propagate, as indicated by the SWR experiments. So in a zeroth order of perturbation, one has to neglect the scattering, and one deals with a Fabry-Perot-type cell, with the wave behavior characterized by the integer n called the mode number:

$$b\sim\sin\left[\frac{n\pi+\phi}{d}z+\phi_1\right] .$$

Then the local equation (12a) reads

$$\omega_n=h'+\frac{ZI}{4}+(n\pi+\phi)^2\frac{Je^2}{d^2} = h'+\frac{ZI}{4}+n'^2\pi^2\frac{Je^2}{d^2} , \tag{13}$$

where the noninteger definition of n' is quite obvious. One can notice that if one takes the mean value, Eq. (13) is nothing else than Kittel's equation (1). Moreover, Eq. (13) has a local meaning if Je^2 does not vary too quickly as has already been assumed in Eq. (11), and is the basis for the treatment of inhomogeneities as square barriers.

1. Inhomogeneities as square barriers

There are two basic assumptions in this treatment: Firstly, inhomogeneities are rather dilute and do not interact strongly between themselves; secondly, the result is independent of the shape of the barriers. The first assumption is justified for quite homogeneous samples, and the second one for the sake of simplicity, i.e., it avoids introducing extra parameters.

So we consider first a one-dimensional one-impurity problem defined by a square barrier or well between two bulk parts and later the multi-impurity problem. The barrier is defined by $\Delta(ZI)$ and $\Delta(Je^2)$, and so for the spin wave of mode number n, it defines a variation Δn. Now one is dealing with a standard problem of wave mechanics or of quantum mechanics, i.e., scattering by a square barrier or well.[11] The ingoing wave is partially reflected, partially transmitted through the barrier with an extra phase shift $\Delta\phi$. If $\Delta n/n$ is weak the reflection can be neglected; if not, the one-dimensional reflection which occurs leads to the loss of one part of the wave for the coherent interference condition. Of course there are different cases

a. $\Delta n/n$ weak. This condition means [from Eq. (13)]

$$\frac{\Delta n}{n}=-\frac{1}{n}\frac{\Delta(ZI)d^2}{8\pi^2 Je^2}-\frac{n}{2}\Delta\frac{(Je^2)}{Je^2}<<1 . \qquad (14)$$

For a given sample with given fluctuations that means a range of n with

$$n_1=\left[\frac{Je^2}{\Delta(ZI)d^2}8\pi^2\right]^{-1}<<n<<\frac{2Je^2}{\Delta(Je^2)}=n_2 . \qquad (15)$$

For these waves there is no practical reflection at the barrier, just a transmission factor $t=\exp(i\Delta\phi)$ which involves the phase shift derived after a straightforward calculation:

$$\Delta\phi_n=\frac{ad}{n\pi}\frac{\Delta(ZI)}{8Je^2}+n\frac{\pi}{2}\frac{a}{d}\frac{\Delta(Je^2)}{Je^2} . \qquad (16)$$

The anisotropy fluctuations determine these phase shifts for the first few modes, while exchange fluctuations are the leading ones for the modes of high n. Of course there is a balance between the two contributions for a value n_0, with

$$n_0=\frac{d}{2\pi}\left[\frac{\Delta(ZI)}{\Delta(JE^2)}\right]^{1/2} , \qquad (17a)$$

and that allows us to write Eq. (16) as

$$\Delta\phi_n=\frac{ad}{n\pi}\frac{\Delta(ZI)}{8Je^2} , \quad n_1<<n<n_0 \qquad (17b)$$

$$\Delta\phi_n=\frac{n\pi}{2}\frac{a}{d}\frac{\Delta(Je^2)}{Je^2} , \quad n_2>>n>n_0 . \qquad (17c)$$

And one notices that $\Delta\phi$ is the minimum for this value n_0, which is in the middle of the range defined in Eqs. (15).

After this one-impurity problem, where because of the one-dimensional character, a two-dimensional average has been taken into account for $\Delta(ZI)$ and $\Delta(Je^2)$, one must consider the many-impurity problem. The impurity p introduces a phase shift $\Delta_p\phi_n$ which determines the transmission factor $t_{n,p}$ of the wave n through this barrier p:

$$t_{n,p}=\exp(i\Delta_p\phi_n)$$
$$\approx 1+i\Delta_p\phi_n-\frac{(\Delta_p\phi_n)^2}{2} . \qquad (18)$$

Neglecting the reflection at the barrier, one obtains the transmission factor T_n through the sample

$$T_n=\prod_p t_{n,p}\approx 1-\sum_p\frac{(\Delta_p\phi_n)^2}{2} , \qquad (19)$$

where, of course, $\sum(\Delta_p\phi_n)$ has been assumed equal to zero because of randomness. So we define the rms effective phase shift $\delta\phi_n$ for one run through the sample

$$\delta\phi_n=\left[\sum_p(\Delta_p\phi_n)^2\right]^{1/2} , \qquad (20a)$$

with

$$T_n\approx\exp(i\delta\phi_n) . \qquad (20b)$$

This effective phase shift $\delta\phi_n$ modulates the tuning equation (13) of the Fabry-Perot-type cell. A straightforward differentiation of Eq. (13) gives the frequency width W_n:

$$W_n=2n\pi\frac{Je^2}{d^2}\delta\phi_n . \qquad (21a)$$

As a matter of fact the physical transmission factor T_n of Eq. (19) corresponds to two rms total phase shifts, namely, $\pm\delta\phi_n$, and as a consequence to the two frequency widths $\pm W_n$. Different parts of the sample experience different local barriers, and therefore different $\delta\phi_n$. So W_n defined from Eq. (21a) with the mean value of $\delta\phi_n$ is a true linewidth measurement. And the results of Eqs. (17) read

$$W_n\sim n^2 , \quad n_2>>n>n_o \qquad (21b)$$

$$W_n\,\text{const}, \quad n_1<<n<n_0 \qquad (21c)$$

where the limiting values of W_n at the boundaries n_1 and n_2 can be estimated in the case of p identical impurities in the sample:

$$W_{n_1}=\frac{a}{8d}\Delta(ZI)p^{1/2}\ ,\quad n_1=\left[\frac{8\pi^2Je^2}{\Delta(ZI)d^2}\right]^{-1}$$
$$W_{n_2}=\frac{a}{d}\frac{\pi^2}{d^2}\frac{(Je^2)^2}{(\Delta Je^2)}p^{1/2}\ ,\quad n_2=\frac{Je^2}{\Delta(Je^2)}\ . \tag{22}$$

These results, valid for the central part of the spin-wave spectrum, are consistent with the experimental results.[1-4] Now one has to consider the two other parts.

b. $\Delta n/n$ non-negligible and $n<n_0$: $n\lesssim n_1$. In this case the condition (11) for a wave equation without damping requires a very small value of $\Delta J/J$ in order to still be valid, as assumed here. The Schrödinger-like equation (12a) gives rise to a reflection at the barrier which is dominated by the anisotropy fluctuations. Because of the random character of the impurities, the reflected waves badly interfere and so are practically lost. As a result the amplitude $|T_z|^2$ of the transmitted wave decreases at each inhomogeneity barrier. Quite obviously this is an exponential decrease and $|T_z|^2$ looks like

$$|T_z|^2\sim\exp[-(z/N_0)]\ ,$$

where the effective damping length N_0 is a coherence length which is determined by the one-impurity scattering t_1 and by the mean distance d_0 between the two-dimensional scatterers:

$$N_0=\frac{d_0}{1-|t_1|^2}=\frac{d_0}{|r_1|^2}\ . \tag{23}$$

In terms of the local fluctuations N_0, it reads

$$N_0=64d_0\pi^2\frac{J^2e^4}{\Delta^2(ZI)d^2a^4}n^2\ , \tag{24}$$

where n has been written instead of n', for the sake of simplicity. When the coherence length N_0 becomes of the same order of magnitude or smaller than the film thickness d, one has to deal with two complementary effects. Firstly, the number of scattering centers involved in Eq. (19) decreases, each of these scattering centers has a more or less strong contribution according to its z location in the sample; as a result $\delta\phi_n$ and W_n decrease from the value calculated with formulas (17a) and (21a). On the other hand, the coherence length N_0 involves a loss of memory of the phase shift for a distance N_0 and so a supplementary mean phase shift $\delta_0\phi_n$ for one run through the sample:

$$\delta_0\phi_n=\frac{\pi d}{N_0}\sim n^{-2}\ . \tag{25}$$

By application of Eq. (21), this induces an n^{-1} law for the additional W_0:

$$W_0\sim n^{-1}\Delta^2(ZI)\ . \tag{26}$$

This explains the strong increase of W near $n=0$, which is shown to be driven by the fluctuations of anisotropy.

c. $\Delta n/n$ not weak and $n>>n_0$: $n\gtrsim n_2$. There is no drastic condition (11) on $\Delta J/J$ in order to have undamped equations. However, Eq. (12a) shows a reflection at the barrier which arises from the fluctuations of the exchange. As in the previous paragraph the reflection simulates a penetration depth N_1 of the wave, which can be read

$$N_1=4d_0\frac{J^2e^4}{\Delta^2(Je^2)}\frac{d^2}{a^2}\frac{1}{n^2\pi^2}\ , \tag{27}$$

with a similar exponential damping. Since the observed spin waves in SWR do not have a very short wavelength, N_1 remains large compared to d. So the supplementary phase shift $\delta_1\phi_n$ introduced by the new interference condition $\delta_1\phi=\pi d/N_1$ in analogy with Eq. (25) is always negligible. However, the finiteness of N_1 means a weaker effective weight for the scatterers, and thus the total phase shift $\delta\phi_n$ is weaker than that derived from Eq. (17a). So we introduce the typical root-mean-square value $\Delta\phi_r$ of $\Delta\phi$ for a one-impurity layer. A straightforward calculation shows that the total phase shift $\delta\phi_n$ is proportional to $\Delta\phi_r$ and to $N_1^{1/2}$:

$$\delta\phi_n\sim N_1^{1/2}\Delta_r\phi\sim n\ , \tag{28}$$

where n is independent. Finally, the frequency linewidth W is proportional to n when n is large enough. This saturation effect of W_n is due to exchange fluctuations, but W_n becomes independent of ΔJ in the simple model of similar impurity layers, W_n is connected with the spatial density of defects, namely, proportional to $d_0^{1/2}$.

One can notice that for large n, i.e., for strongly damped spin waves, the damping terms included in Eq. (10) cannot be neglected. This means a complex behavior of n and ω which has already been assumed by some authors in order to explain the SWR experimental data.[12]

II. CONCLUDING REMARKS

This model describes the four parts of the curve $W(n)$ in agreement with the known data with more or less symmetrical contributions of anisotropy fluctuations and exchange fluctuations, as shown in Fig. 1. From the given analysis of the phase-shift widths $\Delta\phi$, one can derive the mean phase-shift drift $\delta\phi$, both for even and odd n modes. For the same reasons as before, four kinds of behavior are expected. Finally, this means that from the intensities of even modes in SWR one can measure the local fluctuations of exchange and anisotropy.

A mean feature of this theory is that long-range defects such as dislocations are neglected. This is probably valid for annealed amorphous thin films, and depends on the mechanical treatment. For crystalline samples, long-ranged defects are more probable and such defects are very efficient in long-wavelength spin-wave scattering. This is probably the reason for a larger linewidth in crystalline materials as observed by Vittoria *et al.*[1]

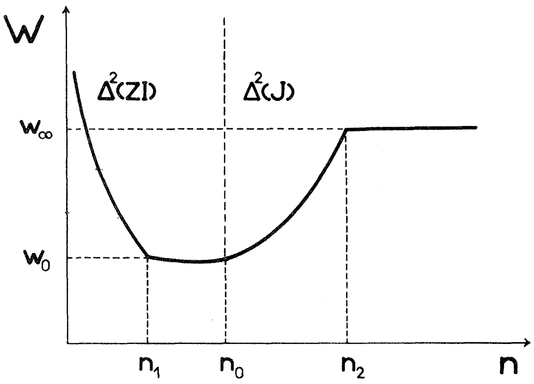

FIG. 1. Typical curve $W(n)$ linewidth versus mode number with the four regions of different behavior. The slope of $W(n)$ for large n has been chosen to be nearly zero for the sake of simplicity.

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