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Interface mode absorption line in ferromagnetic resonance of antiferromagnetically coupled bilayer films

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Abstract. It is believed that the high-field line of the double-peak ferromagnetic resonance spectrum observed in exchange coupled ferromagnetic bilayer films is due to an out-of-phase mode. By applying rigorous microscopic FMR theory to such systems we show that this mode is in fact due to an in-phase combination of sublayer modes, similar to the next (second) mode peak. We also show that when interface exchange coupling J^{AB} is antiferromagnetic, this high-field side peak is due to the interface-localized mode (in contrast to the bulk character of the next mode peak), and its intensity decreases when negative J^{AB} increases in absolute value. This explains the experimentally observed fact that some double-peak resonance spectra exhibit an inverted pattern of the peak intensities (i.e. $I_2 > I_1$). Finally, we also show that ferromagnetic interface coupling generates only single peak resonance, irrespective of the strength of the coupling. It is concluded that when surface and/or interface bilayer spectra is antiferromagnetic.

1. Introduction

In recent years, ferromagnetic resonance (FMR) in exchange coupled bilayer films has been the subject of numerous papers. From the experimental viewpoint, this FMR is characterized by the fact that (1) some specimens show single resonance, whereas (2) others show double resonance. Moreover, double resonance can exhibit a regular pattern in which the high-field (HF) line possesses an intensity greater than that of the low-field (LF) line, or it can exhibit an inverted pattern with a HF line less intense than the LF line. Whereas there is no agreement among the authors regarding the occurrence of single resonance as distinct from double resonance (opinions vary strongly-some authors say that single resonance is the result of strong interface coupling [1] while others, on the contrary, believe it to be caused by weak coupling), there is general agreement that the inverted FMR pattern arises because the HF line is an out-of-phase (and the LF line is an in-phase) composition of the individual sublayer modes. Here, this consensus seems to be due to the circumstances that out-of-phase modes, naturally, have a lower net magnetization associated with them; this is invoked as an explanation of why the HF resonance line has an intensity lower than the LF line. However, a perusal of the literature shows that this interpretation of the inverted FMR pattern lacks rigorous proof; its basic assumption-that between the two lines observed the HF line is out-ofphase and the LF line in-phase—has never, to our knowledge, been proved; moreover,

from a more general standpoint, that interpretation raises the objection that an in-phase composition of the sublayer modes implies their symmetrical combination whereas an out-of-phase composition is obviously antisymmetric. Now, elementary quantum mechanics states that a symmetric combination is always endowed with an energy lower than the respective antisymmetric combination—a rule that would make one assume the HF line as in-phase, in contradiction to the interpretation given by experimenters.

This divergence between a basic result of quantum mechanics and the widely accepted interpretation of the experimenters stimulated us to take a closer look at the whole problem. The theoretical explanations of ferromagnetic resonance in bilayers are, as a rule, based on phenomenological equations of motion of the spins. However, Erickson and Mills [2] note that the use of such equations for ultra-thin films like the ones we are studying is by no means fully justified since, in applying the long-wave approximation, we omit certain essential terms expressing the energy of the spins (for details, we refer the reader to the original paper [2]). Thus, one might be led to think of the above divergence as due to the application of approximate methods of calculation of the phenomenological approach, and that a completely microscopic theory would have been more appropriate.

In this paper, we accordingly propose a theory of FMR in ultra-thin bilayers based throughout on a microscopic approach. Our theory introduces order into the problems concerning the nature of the resonance lines observed in bilayer FMR. We show that the HF line always corresponds to an in-phase mode which is interface localized; its intensity decreases with growing strength of its localization on the interface and, when localization becomes sufficiently strong, its intensity becomes less than that of the next mode, which is of the bulk type. This is an effect similar to that observed many years ago [3] in singlelayer film, where the inverted pattern of spin-wave resonance (SWR) was shown to be due to surface-localization of the high-field lines.

Our theory, moreover, explains the hitherto unclarified problem of the conditions for the occurrence of single, or double FMR. This problem hinges on the nature of the exchange coupling acting between the sublayers. It is now well known from numerous observations [4] that the coupling can be ferro- or antiferromagnetic, and that the nature of the interface interaction can vary with the thickness of the spacer and can oscillate between ferromagnetic and antiferromagnetic coupling. According to our theory, singlepeak absorption results from ferromagnetic coupling on the interface and this feature is altogether independent of the strength of the coupling; here, our conclusions are in agreement with the work of Layadi and Artman [5], who used a phenomenological approach. On the other hand, a multi-peak absorption spectrum results from resonance due to antiferromagnetic coupling at the interface which, moreover, creates the interface-localized mode. Since, however, the predominant intensities belong to the first two modes, experimenters are apt to notice these two lines only. Our theory predicts a regular intensity pattern or an inverted one depending on the strength of the antiferromagnetic coupling at the interface (an inverted pattern is a symptom of strong antiferromagnetic coupling).

2. Microscopic treatment of interface exchange coupling

Our microscopic theory of ferromagnetic resonance in bilayer ferromagnetic film applies the Heisenberg model of localized spins on the assumption that the two ferromagnetic sublayers are exchange coupled by way of their interface. The sublayers are crystallographically and magnetically identical. The interface coupling can be arbitrarily strong, of either sign (we admit ferromagnetic, or antiferromagnetic, interface coupling). The Hamiltonian contains an exchange term (J_{bulk} is the exchange integral between nearest neighbours) and a Zeeman term; for the sake of simplicity, we assume the external field to be applied perpendicularly to the film surface. Since we shall concentrate on effects originating in coupling at the interface, we assume in this paper that the system is free of pinning from surface anisotropy on the outer surfaces of the film as well as of intrinsic surface anisotropy on its internal interfaces, so that the only inhomogeneity in the magnetic properties of the system is assumed to come from the difference between the interface exchange integral J^{AB} and the bulk integral J_{bulk} .

We take the Hamiltonian of reference [6] (consisting of standard exchange and Zeeman energies) and diagonalize it using the method proposed earlier [7]. We then obtain the analytical solution by applying our earlier proposed method [8, 9] of rescaling the interface conditions. Since the external field is orientated perpendicularly to the film the ground state of the system of spins in the bilayer can be assumed to be strictly collinear because, under conditions of perpendicular resonance, the external field is sufficiently strong to cause saturation of the magnetic specimen. Moreover, the choice of a perpendicular external field presents the advantage of precluding terms from elliptical precession of the spins from appearing in the diagonalization procedure so that the latter involves no approximations (except for the well-justified quasi-saturation assumption). The results we give further on are thus exact results, for a range of temperatures which are low in comparison with the Curie point of the material.

It is worth re-stating that, in our approach, all the spins of the bilayer are oriented parallel to the external field, which is perpendicular to the plane of the film. This is so, not only if the interface integral J^{AB} is ferromagnetic but also if it is antiferromagnetic, since the external field has, in fact, been assumed to vanquish the tendency to spin reversal at the interface arising from negative J^{AB} .

Our simplification assuming symmetric boundary conditions on the bilayer will facilitate our interpretation of the results, which will nonetheless turn out to be close to experiment. It will obviously mean that the spin-wave solutions obtained within the above framework will be either purely symmetric or purely antisymmetric. It follows from the diagonalization procedure that the wave-functions of magnon modes $u_l(k)$ are solutions of the following set of difference equations:

$$(x-1)u_0 - u_1 = 0 l = 0 -u_0 + xu_1 - u_2 = 0 l = 1$$

$$-u_{N-2} + (x - b)u_{N-1} - \rho u_N = 0 \qquad l = N - 1$$

$$-\rho u_{N-1} + (x - b)u_N - u_{N+1} = 0 \qquad l = N$$

$$-u_N + xu_{n+1} - u_{N+2} = 0 \qquad l = N + 1$$
 (1)

$$-u_{L-3} + xu_{L-2} - u_{L-1} = 0 \qquad l = L - 2$$

$$-u_{L-2} + (x - 1)u_{L-1} = 0 \qquad l = L - 1$$

where we have introduced the following notation:

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$$x = 2 + (g\mu_{\rm B}H^{\rm eff} - E)/2SJ_{\rm bulk}z$$
(2a)

$$b = 1 - \rho \tag{2b}$$

$$\rho = J^{\rm AB} / J_{\rm bulk}. \tag{2c}$$

The meaning of the quantities occurring in the above formulae is the following: $L \equiv 2N$ is a thickness of the bilayer film (in lattice units), S is the spin number (in units of \hbar), z is the number of nearest neighbours situated in the adjacent plane, E is the energy of a given spin-wave mode, b denotes the interface-pinning parameter, whereas ρ is the interface-coupling parameter. One sees that the pinning as well as the coupling interface parameters are functions of the interface exchange integral J^{AB} . By inserting $x = 2 \cos k$ into (2a) one gets the mode energy E expressed by the wave-number k:

$$E(k) = 4SJ_{\text{bulk}}z(1 - \cos k) + g\mu_{\text{B}}H^{\text{eff}}.$$
(3)

The set of (1) has already been solved strictly by applying the recurring interface rescaling approach [9]. Here, we shall be using the functions $u_l(k)$, explicitly expressed by the wave-number k in the form given in our paper [9], which also contains the respective characteristic equation quantizing the mode-number k.

It is instructive to analyse the characteristic equation graphically. With this aim in mind we rewrite the equation given in [9] in the form of the following two equations:

$$r(k) = \cos[(2N+1)/2]k$$
 [1 (4a)

$$T(k) = \frac{1}{\cos[(2N-1)/2]k} - \left(1 - 2J^{AB}/J_{bulk}\right).$$
(4b)

These two equations jointly lead to 2N allowed values of the variable k. The set of these values is obviously dependent on the bilayer thickness 2N; however, concerning the effect of the interface coupling, we see that only half of the permitted k values is affected by the interface exchange coupling integral J^{AB} . This is a fact of considerable importance, as we shall see later on.

The model-number $k \operatorname{can}$, in general, be complex; only three types of k are permitted, to each of which there corresponds a different kind of spin wave:

(i) k real(bulk modes)(ii) k = it(acoustical interface modes)(5)(iii) $k = \pi + it$ (optical interface modes)

where t is a real, positive number. On inserting (5) into (3) one obtains formulae which show that the acoustical and optical interface modes have energies lying, respectively, below and above the energy 'band' of the bulk spin-wave modes. It is easily verified that all solutions $u_i(k)$ are obtained on restriction to the interval $k \in (0, \pi)$ for bulk modes, and $t \in (0, +\infty)$ for interface modes. The roots of (4a) can be found analytically; they are:

$$k_n = n(\pi/2N)$$
 $n = 0, 2, 4, \dots, (2N-2).$ (6)

There is no general analytical method available for obtaining solutions of (4b) for arbitrary values of the normalized interface exchange integral (J^{AB}/J_{bulk}) . In order to gain some essential information on the k-spectrum, we will proceed graphically. In

Interface bilayer mode in ferromagnetic resonance



Figure 1. Accessory graph for the discussion of the characteristic equations (4): (a) for ferromagnetic interfacial coupling; (b) for antiferromagnetic coupling. We denote here: S—symmetric modes, A—antisymmetric modes. Bilayer thickness of 22 monoplanes is assumed. The roots of the characteristic equation are obtained graphically by searching for the points of intersection of the two straight lines and the curves. In the central part one obtains bulk modes; the root corresponding to the acoustic interface mode is obtained from the graph to the left, while the optical interface mode from the graph to the right.

figure 1 we have plotted the function F(k) defined in (4) (in the middle for bulk modes, to the right and left for optical and acoustical interface modes, respectively). On fixing some value of the interface parameter J^{AB}/J_{bulk} , the roots of (4b) are found by searching for the points of intersection of the straight line parallel to the axis of abscissae and the curves F(k).

3. Results

A fundamental rule states that (isolated) sublayer modes coalesce into (complex) bilayer modes when coupling between the sublayers is 'switched on'. This is illustrated in figure 2, where the phenomenon of composition of single-layer modes to give multi-layer modes is shown separately for ferro- and antiferromagnetic coupling. The middle part of figure 2 shows the modes of the single-layer film; the lowest mode corresponds to the uniform mode because we have assumed the sublayers to possess no intrinsic surface anisotropy (according to the terminology applied in our earlier paper [3]: the boundary conditions correspond to natural pinning). One notes immediately that the coalescence of 'sublayer modes' to 'bilayer modes' leads to results that differ depending on whether

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Figure 2. Formation of bilayer coupled modes as dependent on the nature of the interfacial exchange coupling of the sublayers. For ferromagnetic (antiferromagnetic) interfacial coupling symmetric (antisymmetric) modes are unaffected by the coupling.

the exchange integral J^{AB} is positive or negative; however, the initial mode goes over into two complex modes by way of the following two combinations: in-phase, or out-ofphase. Obviously, the modes which arise by way of in-phase combinations are symmetric modes (on the whole cross-section through the bilayer film), whereas those formed in 'out-of-phase' combinations are antisymmetric modes. Within a pair of modes with opposite symmetries, the in-phase partner always possesses an energy lower than its out-of-phase partner.

We note, moreover, that the eigen-modes of a single layer with natural boundary (pinning) conditions are highly characteristic. Its symmetric modes (n = 3, 5, ...) present the following property: if halved, they become strictly antisymmetric in either of the resulting two halves of the layer. On taking two sublayers, both of them natural, and on subjecting them to exchange coupling mutually, the resulting bilayer will exhibit an eigen-mode spectrum in which one half of the modes are natural modes of the resulting bilayer (thus, they correspond to quantization $k = n\pi/2N$; with 2N being the number of monoplanes in the film). However, these 'natural' modes of the bilayer fail to possess a univocally-determined type of symmetry; their symmetry is dependent on the nature of the exchange coupling of the sublayers (figure 2); if it is antiferromagnetic, the natural modes are antisymmetric, whereas if it is ferromagnetic they are symmetric. In the latter case half the spectrum of the bilayer consists of its natural symmetric modes, i.e. presenting the property of being *strictly* antisymmetric in either of the two halves of each sublayer.

Figure 3 shows how the energy of the individual bilayer modes is dependent on the sign and value of the coupling J^{AB}/J_{bulk} . It is highly interesting that, irrespective of the sign at J^{AB}/J_{bulk} , every second mode of the spin-wave spectrum does not change its energy with varying J^{AB}/J_{bulk} . We shall refer to the modes with constant energy (i.e. independent of the interface coupling) as 'non-affected' modes. For ferromagnetic coupling $(J^{AB}/J_{bulk} > 0)$ the non-affected modes are those labelled with odd numbers (n = 1, 3, ...) whereas in the case of antiferromagnetic coupling $(J^{AB}/J_{bulk} < 0)$ they are the even modes (n = 2, 4, ...). We refer to the other modes (those with an energy varying with J^{AB}/J_{bulk}) as 'affected' modes. Among the affected modes, the special case of the mode with n = 1 for antiferromagnetic coupling merits particular attention: its





energy decreases steeply as the interface coupling J^{AB} increases. As we shall see, this is the mode localized on the interface; it thus possesses a great amplitude in the region where the coupling J^{AB} is most active and this explains its exceptionally strong sensitivity to changes in J^{AB} .

Figure 4(a) shows profiles of the unaffected modes for ferro- and antiferromagnetic coupling, respectively; these profiles are preserved as J^{AB} varies. Figures 4(b) and (c) show how the profiles of affected modes change with growing interface coupling, for ferro- (figure 4(b)) and antiferromagnetic (figure 4(c)) coupling. Figure 4(c) convincingly illustrates the changes undergone by the profile of the mode n = 1—from almost uniform at $J^{AB} \approx 0$ to interface-localized for $J^{AB}/J_{bulk} \ll 0$.

We are now able to proceed to an interpretation of the swR calculated within the framework of our theory. In figure 5 we show swR spectra, calculated for different values of the ratio J^{AB}/J_{bulk} . For ferromagnetic coupling we always obtain a single-peak absorption spectrum irrespective of the value of J^{AB}/J_{bulk} . If, however, J^{AB}/J_{bulk} is antiferromagnetical, the resonance spectrum is of the multi-peak kind and its structure changes with varying J^{AB}/J_{bulk} . The most essential change is the fact that the intensity of the first mode (n = 1) decreases (with respect to that of the next mode n = 3) with growing antiferromagnetic coupling; this is so because the mode n = 1 is interface-localized (IM). Note that for some critical value of the ratio J^{AB}/J_{bulk} (between -0.2 and -0.3) the intensities of the first two modes (n = 1, 3) become inverted, as indeed is observed in various experiments. The findings of this figure are easily understood by referring them to figure 6, where profiles of all modes (both resonant and non-resonant) are shown for each strength of the interfacial coupling considered here.

We have assembled in figure 6 the profiles of the six energetically lowest modes for different values of J^{AB}/J_{bulk} . The result concerning the out-of-phase modes is simple:

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Figure 4. (a) Bilayer modes unaffected by the interface coupling; (b) and (c) modes affected in their dependence of the ferromagnetic and antiferromagnetic interface coupling parameter, respectively. Note that when negative J^{AB}/J_{bulk} increases in absolute value the interface localization of the (n = 1)-mode becomes stronger.

NORMALIZED SWR SPECTRA CALCULATED FOR VARIOUS VALUES OF INTERFACE EXCHANGE COUPLING J^{AS} : (a) FERROMAGNETIC, (b-e) ANTIFERROMAGNETIC COUPLING



Figure 5. Stick spin-wave resonance spectra calculated for various values of the ratio of the interface exchange coupling integral and the bulk exchange integral J^{AB}/J_{bulk} , i.e. for various strengths of the interfacial coupling of the bilayer AB. The spectra exhibit only peaks corresponding to the resonant symmetric modes (of odd numbers n = 1, 3, 5, ...). The calculations are performed for the case when both sublayers A and B have equal thicknesses (11 monolayers each). For the ferromagnetic type of interface coupling (a) the spectrum always consists of only one single FMR peak, irrespective of the strength of the coupling; this mode corresponds to the uniform mode (UM). For the antiferromagnetic type of interface coupling (b)-(e) the multi-peak spectra always exhibit the presence of the interface-localized mode (IM), which is located on the low energy side of the spectrum. The units of the horizontal axis are proportional to the normalized energy; the peak intensities have also been normalized by assuming the intensity of the highest peak as unity (in each spectrum separately).

they are always antisymmetric and, as such, have no magnetization associated with them; thus, they contribute no lines to the resonance spectrum. The results concerning the symmetric modes are much more interesting: we note that in the case of ferromagnetic coupling (figures 6(a), (b)), among all the symmetric modes only the energetically lowest uniform mode (n = 1) is able to absorb energy from the microwave field, whereas the other symmetric modes (n = 3, 5, ...) are *non-resonant* since the integrated area under the respective bilayer mode profile gives a net result equal to zero. This is the reason why, always, irrespective of the value of the ferromagnetic integral J^{AB}/J_{bulk} , one obtains but one resonance line.

In the case of antiferromagnetic coupling (figures 6(c), (d)) the symmetric modes are resonant since, due to their sensitivity to changes in J^{AB}/J_{bulk} (affected modes) they are deprived of the property of pure antisymmetricity in the individual sublayers so that summation of their amplitudes over the cross-section through the film gives a result different from zero. Thus, the swR spectrum now consists of a set of lines, corresponding to the resonant symmetric modes. Since the interface-localized mode is symmetrical, the resonance spectrum contains the corresponding line (n = 1, the line is denoted by IM). If its localization is very strong (this corresponds to a high negative value of J^{AB}/J_{bulk}) its resonance absorption is decidedly less than that of the next symmetric mode (n = 3), which is of the bulk type; the respective intensity ratio I_3/I_1 is plotted separately versus J^{AB}/J_{bulk} in figure 7, where the 'inversion' of the intensities of the first two modes is shown to exist for continuously varying J^{AB}/J_{bulk} . Regarding figure 7, note moreover that as J^{AB}/J_{bulk} tends to zero so does I_3/I_1 ; this results from the circumstance that, in the limit, the mode n = 3 becomes non-resonant symmetrical.

4. Final remarks

We have based our considerations on a bilayer film taking into account *intrinsic* interface coupling only so as to have the resonance effects at the interface stand out in pure form.

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Figure 6. It is highly instructive to collect profiles of the low-energy spin-wave resonance modes into respective separate sets corresponding to different values of the ferromagnetic (a), (b) or antiferromagnetic (c), (d) interface coupling. The consecutive modes correspond to the respective resonance lines of the SWR spectrum shown at the bottom of each figure. Notations used: A—antisymmetric mode, RS and NRS—resonant and non-resonant symmetric mode, respectively; BM—bulk mode, IM—interface mode; N_i —number of monoplanes in the *i*th sublayer. From this figure we derive the following rule for observations of SWR: two identical sublayers (with natural surface pinning conditions) subjected to ferromagnetic coupling still fail to exhibit a SWR spectrum (irrespective of the strength of the coupling), but do exhibit a SWR spectrum if coupled antiferromagnetically.



Figure 7. Calculated intensity ratio of the third (bulk) and first (interface) modes versus antiferromagnetic interfacial coupling.

However, it should be added that—as shown by us earlier [6, 10, 11]—intrinsic interface coupling exerts some effective pinning on the interface spins; we have taken this effect duly into account in the course of this investigation. Furthermore, we are able to take into consideration both intrinsic (outer) surface spin pinning and interface (internal) pinning; the respective calculations have been carried out and will be published separately; they confirm the principal thesis of this paper: only escalation of antiferromagnetic coupling at the interface leads to IM arising, whereas ferromagnetic coupling produces a tendency to the contrary.

The reason why antiferromagnetic (and not ferromagnetic) interface coupling leads to localization of the spin wave on the interface is quite obvious. What is the physical meaning of the spin-wave amplitude? The square of its modulus determines the probability of reversal of a spin at a given node of the spin lattice (this reversal then becomes a collectivized spin-wave process). In our case, the spins at the interface, which are all disposed vertically to the film surface, are mutually parallel due to the ordering action of the external magnetic field. If, in addition, ferromagnetic coupling between them becomes enhanced, their pinning in the direction of the external field undergoes an enhancement too so that it is now more 'difficult' for them to deviate from the field direction. Inversely, if interface coupling is antiferromagnetic, the natural tendency of this kind of coupling is to array the spins antiparallel, so that the pinning of the interface spins in the external field direction weakens. Thus, the interface spins achieve some sort of freedom owing to the antiferromagnetic coupling between them, and their reversal (the onset of the collective spin-wave process) becomes easier. This is equivalent to an increase in probability of reversal at the interface and an enhancement of the interface spin wave amplitude, apparent experimentally as the interface mode.

The presence in the SWR spectrum of a resonance line due to IM is a potential source of information concerning the interface. Thus, for example, figure 7 proves that by fitting the intensity ratio $I_{\text{bulk}}/I_{\text{interface}}$ derived from our theory to the experimental data, one can easily determine the value of the interface exchange integral. Obviously, the bilayer used here is suitable for quantitative studies only in rather specific cases, i.e. when interface coupling is the predominant factor influencing the spin dynamics. In the most general case, corresponding to real specimens, one will have to take into consideration the surface and interface anisotropies as well as the eventuality that these anisotropies may cause the boundary conditions to be asymmetric [12]. Also, the external field may be conceived of as tilted at an arbitrary angle to the surface of the film. Calculations for these cases are now under way.

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