Layered Magnetic Structures: Evidence for Antiferromagnetic Coupling of Fe Layers across Cr Interlayers

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We investigated exchange coupling of Fe layers across Au and Cr interlayers by means of light scattering from spin waves. For Au interlayers we find a continuous decrease of this coupling to zero as the Au thickness is increased from 0 to ≈ 20 Å. For Cr interlayers of proper thickness we find antiferromagnetic coupling of the Fe layers. In small external fields such double layers order antiparallel with their magnetization perpendicular to the external field, in analogy to the spin-flop phase of antiferromagnets.

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Recently exchange coupling in superlattice structures consisting of infinitely many alternating ferromagnetic and antiferromagnetic films has been studied theoretically and a large variety of new magnetic phases was predicted.[1] For practical reasons we confine ourselves experimentally to magnetic double layers consisting of two Fe films separated by a thin Cr or Au film. The latter serves as a reference. For [100]-oriented growth there is a good match of the Fe, Cr, and Au lattices as was noted before.[2] More details on the preparation of such samples will be communicated at a later date. Fe-Cr-Fe and Fe-Au-Fe double layers have been investigated before.[3] In this case, however, growth was polycrystalline and not oriented as for the samples presented in this work.

The method of evaluating the effective interlayer exchange coupling from the mode spectra of such double layers was described elsewhere.[3] For the present purpose it is sufficient to know that the thicknesses of the magnetic films have been adjusted in such a way that two spin-wave modes can be conveniently measured by the light-scattering technique: one which depends on the strength of the interlayer exchange and one which does not. The latter for a two-film system with 100-Å individual film thickness is at the position of the Damon-Eshbach (DE) mode of a 200-Å-thick film and can be used to measure magnetization and anisotropy of the ferromagnetic films. In the following we want to call the other mode, which does depend on the interlayer exchange, the "exchange mode." It is at the position of the first standing spin-wave mode of a 200-Å-thick Fe film when the interlayer thickness is reduced to zero. This is the full-coupling limit. In the other limit, when exchange coupling across the interlayer can be neglected but the interlayer thickness d_0 is still very small, the exchange mode is in the position equivalent to the ferromagnetic resonance. This is a result of the dipolar coupling theory[3] under the assumption that $d_0 \ll k_y^{-1} = 580$ Å, where $k_y = 1.73 \times 10^5$ cm^{-1} is the in-plane wave vector of the spin waves seen by the light-scattering experiment. Hence in this limit the frequency of the exchange mode is given by

$$\omega = \gamma[(B_0 + B_{an})(B_0 + B_{an} + J)]^{1/2} \quad (1)$$

for B_0 along the easy axis [100], and

$$\omega = \gamma[(B_0 + B_{an}/2 + J)(B_0 - B_{an})]^{1/2} \quad (2)$$

for B_0 along [110]. Here B_0 is the external field, B_{an} is the anisotropy field, and J and d are magnetic polarization and thickness of the ferromagnetic films. We chose $\gamma = 1.85 \times 10^{11}$ T^{-1} s^{-1}, equivalent to a g factor of $g = 2.1$ for Fe.

In the limit of small d_0 and $B_0 \| [100]$ the DE mode is given by

$$\omega = \gamma[(1 - e^{-2k_y d})(J/2)^2 + (B_0 + B_{an})^2 + (B_0 + B_{an})J]^{1/2}, \quad (3)$$

and as mentioned is independent of the interlayer exchange. For $B_0 \| [110]$ the frequency of that mode has also recently been worked out[4] but for the present purpose it is sufficient to know that its shift from $B_0 \| [100]$ to $B_0 \| [110]$ is only about half the value of the shift displayed by the exchange mode. Hence for the two extreme cases of full and zero interlayer exchange coupling we can predict the mode positions as just indicated.

The concept developed so far describes very well the situation which we find in the Fe-Au-Fe double layers. Spectra with increasing Au thickness are displayed on the right-hand side of Fig. 1. For each thickness two spectra were taken—one with B_0 along [100] (e=easy) and one with B_0 along [110] (h=hard). B_0 is always in the plane of the film. Clearly for B_0 along [100] both modes shift to higher frequencies which is in agreement with the fact

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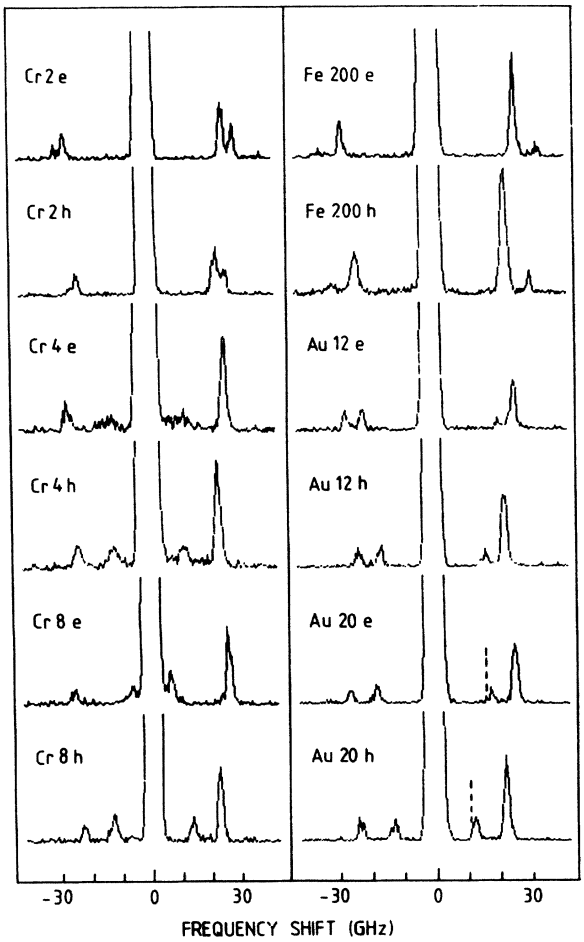

FIG. 1. Light-scattering spectra from spin waves in 200-Å-thick single layers of Fe (upper two traces of left-hand side) and Fe double layers where the interlayer material and thickness is marked on the various traces (e and h indicate direction of external field along easy and hard axis of Fe). From traces Fe 200e and Fe 200h we evaluate $J=2.0$ T and $B_{an}=0.035$ T which yields the position of the exchange mode in the decoupled case marked by dashed lines in traces Au 20e and Au 20h. $B_0=0.1$ T in all cases.

that here the anisotropy field assists the external field. The observation that these films show the expected magnetic anisotropy of Fe confirms their epitaxial growth. From the position of the DE mode and the formalism described above we evaluate the magnetization J and anisotropy field B_{an}. We find $J=2.0$ T and $B_{an}=0.035$ T. Next we use these parameters to predict the position of the exchange mode for vanishing interlayer exchange. In the lower traces on the right-hand side of Fig. 1 these positions are indicated by dashed lines. They agree well with the actual line positions. The slight disagreement can be attributed to the fact that the interlayer thickness d_0 is not really zero, which for modes with $k_y\neq 0$ causes some dipolar coupling and a slight upshift of the exchange mode. For the same reason the DE mode is now at a somewhat lower frequency than in the Fe 200 traces. Anyway we conclude that in double layers with Au interlayers of thickness $d_{Au}=20$ Å the Fe layers are exchange decoupled. This is also confirmed by the observation that a further increase of the Au thickness (not shown) slowly shifts the mode to ever higher frequencies, in agreement with the dipolar theory.

Next we turn to the spectra from the Fe-Cr-Fe double layers shown on the left-hand side of Fig. 1. Again for each film we confirm the expected magnetic anisotropy and hence epitaxial growth. The DE mode as before shows only small dependence on the interlayer thickness. The exchange mode, however, now behaves quite differently than in the Fe-Au-Fe reference. Let us first discuss the spectra for $B_0\|[100]$ (e). For $d_{Cr}=4$ Å we see that the exchange mode has already dropped *below* the value expected for the exchange-decoupled case (compare to the lowest traces on the right-hand side). At the same time there is appreciable line broadening. When the Cr thickness is increased the mode stays at a low frequency but becomes more narrow. The fact that this mode is now observed at a frequency which is lower than the value corresponding to zero interlayer exchange can only be interpreted such that the effective coupling has not become antiferromagnetic. This is also in agreement with the phenomenological theory.[5] The line broadening at $d_{Cr}=4$ Å could, for example, be due to a nonuniform Cr thickness, which would cause strong variations in the effective interlayer exchange.

A remarkable behavior is also observed when B_0 is now brought over to the [110] direction (h). There is a clear frequency upshift for the exchange mode—opposite to what is expected from the Fe anisotropy (note that the DE mode shifts according to the Fe anisotropy). This could be due to an anisotropy within the Cr film. If the Cr moments want to stay aligned along a $\pm$ [100] direction then the coupling to the adjacent Fe layers becomes weaker when we force the Fe moments into a [110] direction. Hence the overall antiferromagnetic coupling of the two Fe films also decreases, which now means a frequency upshift.

The low frequency of the exchange mode as displayed, e.g., in trace Cr 8e of Fig. 1 suggests that this mode might go soft when we decrease the external field which should cause it to shift to even lower values. We have tested this. The result is displayed in Fig. 2. Again we discuss first the behavior of our Fe-Au-Fe reference system displayed on the right-hand side of Fig. 2. Here we have chosen a sample with an Au thickness of 20 Å. When the external field B_0 is decreased all modes shift gradually downwards but stay in the same position for $B_0\lesssim 0.05$ T ($\triangleq$500 Oe). In agreement with Eqs. (1)–(3) this is due to the effect of the anisotropy field of $B_{an}=0.035$ T (350 Oe) of these Fe films. At the lowest applied fields shown, there is still enough remanence to hold the magnetization in the same position as in higher

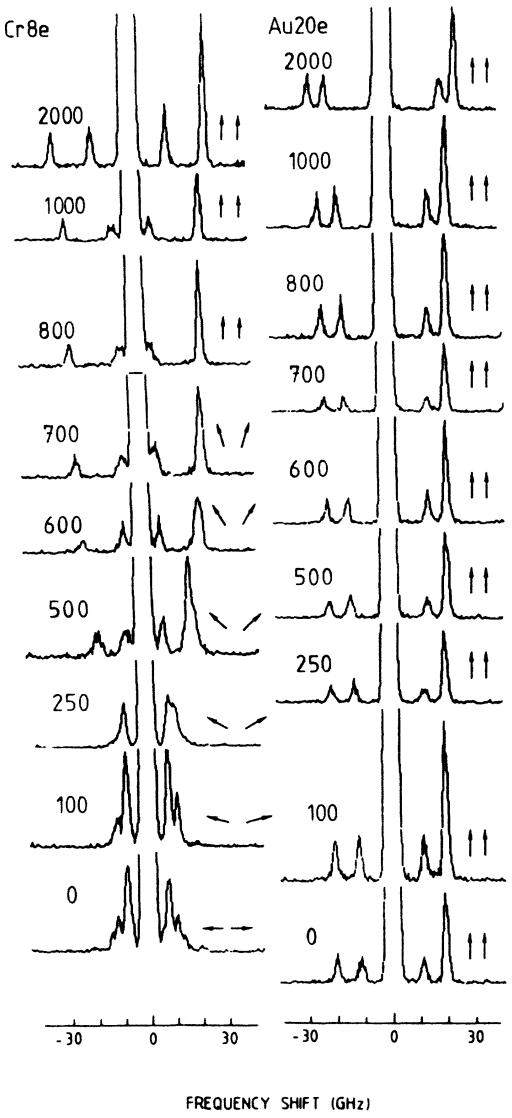

FIG. 2. Spectra from samples Cr 8 and Au 20 with B_0 along the easy axis of Fe. Numbers on the traces mark values of external field in units of 0.1 mT $\triangleq$1 Oe. The arrows indicate the suggested magnetization directions on the two Fe layers where B_0 is supposed to point up. Observed spin-wave propagation then is along a horizontal line.

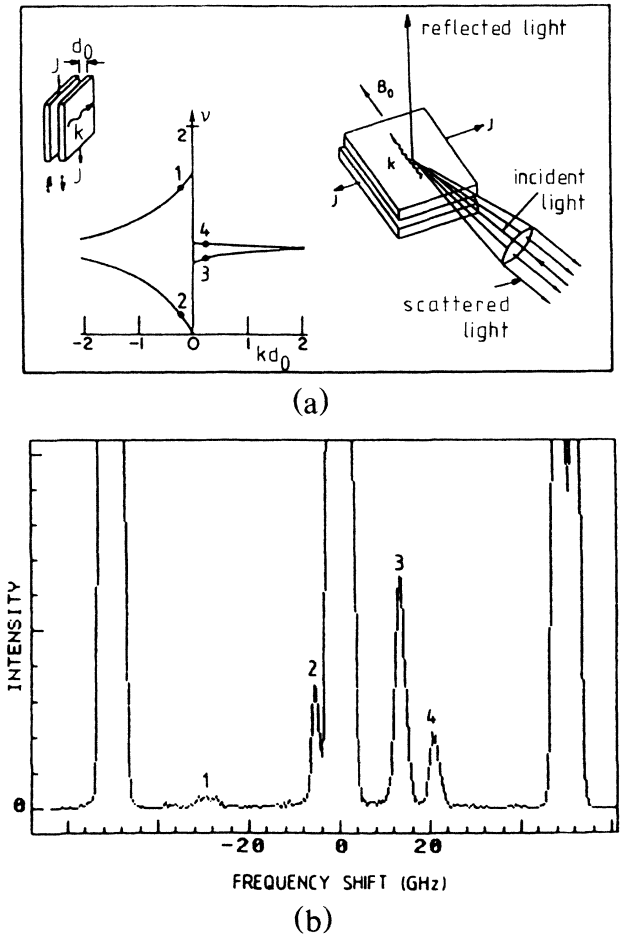

FIG. 3. (a) Dispersion curves of spin waves in antiparallel layers (from Ref. 6) and the employed scattering geometry. (b) The resulting light-scattering spectrum. The numbers indicate the correspondence to the branches in (a). The spectrum proves that the magnetizations J in Cr 8e for small external fields are aligned as proposed in Fig. 2.

fields; otherwise the modes would shift. Opposite to this, for the Fe-Cr-Fe system as B_0 is decreased the exchange mode goes through a minimum at B_0=0.08 T (800 Oe) and comes up again slightly as B_0 is decreased further. At the same time the DE mode starts to shift down which indicates a change in the direction of the sample's magnetization. It is very likely that this has to do with the softening of the exchange mode which makes the previous magnetization configuration unstable. However, as a result of this change the soft mode again increases its frequency. The correct interpretation, however, could also be that the soft mode disappears from the spectrum and another mode comes up instead.[6] To gain further information on the behavior of the static magnetization the net moment of the sample was measured with a Faraday balance as a function of external field. It was found that for decreasing B_0 below 0.05 T (500 Oe) the net moment gradually disappeared.[7] Now since we have seen antiferromagnetic coupling dynamically we expect that it manifests itself also statically. Hence we expect that in low fields the two Fe layers align antiparallel. Final evidence for this comes again from spin-wave mode spectra.

We repeat the low-field experiment of Fig. 2 on the Fe-Cr-Fe sample but change the light-scattering geometry such that we now observe modes which propagate in the direction of the external field. The result is displayed in Fig. 3. From comparison with theory[3,8] we find that this unique pattern occurs for a double layer with antiparallel orientation of the magnetization in a very small

field and modes which travel transverse to the magnetization. Note that opposite to the spectra shown in Figs. 1 and 2, modes occurring on the Stokes and the anti-Stokes sides now have different frequencies. This comes from the fact that an antiparallel layer by symmetry is a truly nonreciprocal system.[8] Here the remarkable feature is that not only do we prove by this that the magnetization of the two Fe films is *antiparallel* but also that it is *perpendicular to the small external field.* This is in complete analogy to the spin-flop phase of an antiferromagnet.

In order to demonstrate the "normal" case were an interlayer of increasing thickness simply decouples the two magnetic films we have chosen here Fe-Au-Fe layers. Since neither Auger nor reflection high-energy electron-diffraction investigations during growth were available to us, it is not certain whether in this system the main coupling mechanism is via pinholes in the Au or, for example, via conduction electrons as in the Ruderman-Kittel-Kasuya-Yosida interaction.[9] For the results from the Fe-Cr-Fe layers the question of the pinholes is not so important because it is hard to imagine that coupling across pinholes could be anything else but ferromagnetic.

Here our goal was to demonstrate the possibility of antiferromagnetic coupling between ferromagnetic films in layered structures. New magnetic materials might evolve from this with interesting physical properties, thanks to the possibilities of the layering.

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