Giant Near-90° Coupling in Epitaxial CoFe/Mn/CoFe Sandwich Structures

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Trilayer samples of CoFe/Mn/CoFe(001) have been prepared by molecular beam epitaxy and investigated by magnetometry and ferromagnetic resonance (FMR). Very strong near-90° coupling (up to 2.5 ergs/cm^2) between the CoFe layers, with no evidence for 180° coupling, was found in all but the thickest Mn-layer samples. The field dependence of the magnetization requires that the coupling energy have the algebraic form $F_c = C_+(\phi_1 - \phi_2)^2 + C_-(\phi_1 - \phi_2 - \pi)^2$ suggested recently for antiferromagnetic interlayers. The observed angular dependence of both the acoustic and optical FMR modes found can be described by this model.

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The electronic basis for the coupling between ferromagnetic layers through an intervening metal film has been pointed out by several authors [1–3] to depend on the relative energy alignment of the spin-split bands in the ferromagnet with the paramagnetic bands in the interlayer. In the Fe/Cr/Fe system, which exhibits very strong coupling, the unsplit bands in paramagnetic Cr are nearly aligned with the minority bands in Fe [2]. Since both Fe and Cr are bcc, we felt that if a new bcc layered system could be fabricated employing Co and Mn, respectively, there would be an opportunity to test this idea, since rigid band shifts would lead to a similar band alignment. The materials studied here (bcc CoFe alloy and bct Mn) provide a close approximation to the desired structure.

In this Letter, we present the experimental results on this system, single crystal epitaxial sandwich structures of CoFe/Mn/CoFe, which exhibit the largest near-90° type coupling ever observed without any detectable bilinear coupling. The form of the coupling is consistent with a recent theoretical model based upon the intervening layer being an ordered antiferromagnet [4].

A set of 15 trilayer samples was prepared by molecular beam epitaxy. The $Co_{75}Fe_{25}$ (001) alloy layers are all ~100 Å thick and were grown using methods described earlier [5]. They are bcc and exhibit reflection high-energy electron diffraction (RHEED) patterns indicating excellent crystalline quality. The lattice constant for this composition alloy is a nearly perfect 2:1 match to that of the ZnSe/GaAs(001) substrates used [5]. The Mn interlayer thicknesses range from 6 to 30 Å as determined by x-ray fluorescence. They are single crystal as shown by RHEED, while extended x-ray-absorption fine structure (EXAFS) measurements showed a bct structure with the tetragonal distortion along the growth direction [6]. Vibrating sample magnetometer (VSM) and SQUID magnetometry, as well as 35 GHz ferromagnetic resonance (FMR) angular dependence, were carried out in plane on all samples to measure their magnetic properties. All trilayers with $t(\mathrm{Mn}) < 30$ Å exhibit evidence of coupling between the FM layers but, for clarity, we discuss only one sample [sample A with $t(\mathrm{Mn}) = 11.2$ Å] in detail.

The low-field VSM magnetization data for sample A are shown in Fig. 1. Note that they seem to imply apparently easy ⟨100⟩ and hard ⟨110⟩ behavior in the (001) plane, exactly the reverse of the behavior of isolated CoFe

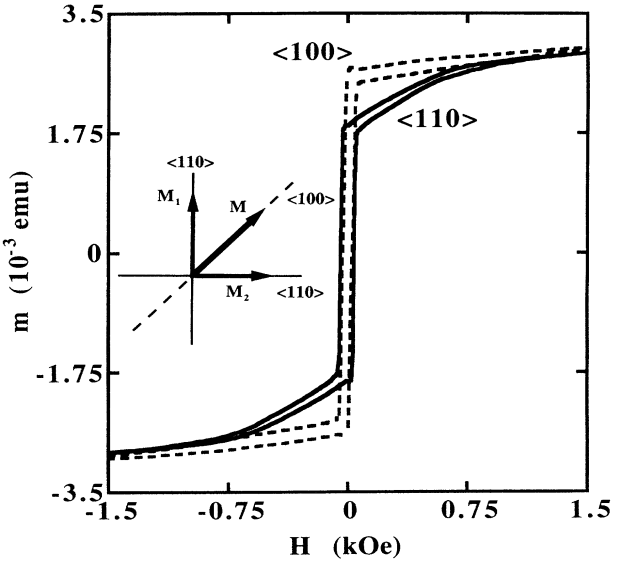

FIG. 1. Magnetic moment vs magnetic field data for a CoFe/Mn/CoFe(001) trilayer sample with $t(\mathrm{Mn}) = 11.2$ Å (sample A). The in-plane magnetic field is applied along the directions indicated. Inset: Moment directions in zero field.

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films with the above composition [5]. If, however, the Mn thickness is increased to 30 Å, the usual isolated film behavior is observed in both VSM and FMR. We call attention to the fact that the $t(\mathrm{Mn}) = 30$ Å FMR data show explicitly that, in this no-coupling limit, the two CoFe layers have indistinguishable anisotropy parameters equal to those of an isolated film.

The above results can be understood easily if the magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$ of the CoFe layers of sample A lie nearly perpendicular to one another in zero field, and along the expected in-plane $\langle 110\rangle$ easy directions, so that the net magnetization $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$ is along $\langle 100\rangle$ (see inset in Fig. 1). For strong nearly-90° coupling, as found here, $\mathbf{M}_1$ and $\mathbf{M}_2$ initially rotate as a rigid unit in an attempt to align $\mathbf{M}$ with $\mathbf{H}$ when a field $\mathbf{H}$ is applied along $\langle 110\rangle$, and they come together very slowly for $\mathbf{H}$ applied parallel to $\mathbf{M}$ along $\langle 100\rangle$. This type of behavior will be found as long as the coupling is large compared to both the in-plane anisotropy and $-\mathbf{M}\cdot\mathbf{H}$ energy density term, as explicit calculations show. The SQUID magnetization data for $\mathbf{H} \parallel \langle 110\rangle$ are shown over a much more extended range in Fig. 2. Note that after the magnetization break near 1 kOe (when the above rotation is complete), M continues to increase as the field causes $\mathbf{M}_1$ and $\mathbf{M}_2$ to come together, and M approaches saturation only slowly above 20 kOe.

A quantitative fit to this end (and the corresponding $\mathbf{H} \parallel \langle 100\rangle$) data requires one to write down the free energy per unit area,

$$F = F_c + F_a - \mathbf{H}\cdot(\mathbf{M}_1 + \mathbf{M}_2)t(\mathrm{CoFe}), \quad (1)$$

where F_c is the coupling energy/area, F_a is the corresponding anisotropy term, and $t(\mathrm{CoFe})$ is the thickness of a FeCo layer. Based on previous FMR measurements on isolated CoFe films [5], we expect

$$F_a = t(\mathrm{CoFe})\sum_i \{K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_u\cos^2(\phi - \phi_u)\}_i, \quad (2)$$

with K_1/M and $|K_u/M|$ about -0.25 and 0.05 kOe, respectively. Here K_1 is the usual cubic anisotropy and K_u is an in-plane uniaxial anisotropy, which distinguishes one in-plane $\langle 110\rangle$ from the other [5]. The expression $\phi_i - \phi_{ui}$ is a measure of the deviation of the in-plane angle ϕ_i from the preferred $\langle 110\rangle$. The above then permits one to determine the magnitude and form of the coupling term F_c by minimizing the energy.

Almost universally, the coupling which has been found experimentally in the many types of magnetic trilayer and superlattice samples studied to date has the form

$$F_c = -2A_{12}\hat{\mathbf{M}}_1\cdot\hat{\mathbf{M}}_2 - 2B_{12}(\hat{\mathbf{M}}_1\cdot\hat{\mathbf{M}}_2)^2, \quad (3)$$

where A_{12} and B_{12} are the so-called bilinear and biquadratic coupling contants and $\hat{\mathbf{M}}_i$ is a unit vector in the direction of $\mathbf{M}_i$. The values of A_{12} and B_{12} generally oscillate and decrease in magnitude as the interlayer thickness increases [7]. For dominant A_{12}, $\mathbf{M}_1$ and $\mathbf{M}_2$ antialign for $A_{12} < 0$. However, if $|B_{12}| \gg |A_{12}|$ and $B_{12} < 0$, the configuration $\mathbf{M}_1 \perp \mathbf{M}_2$ is favored. Both Fe/Cr/Fe and Fe/Al/Fe trilayers exhibit this 90° configuration for some interlayer thicknesses [8–11], but this is not the usual behavior for coupled magnetic films.

The dashed line in Fig. 2 results from a least squares fit to the data of Fig. 2 via a minimization of F at each field yielding the following parameters: $2A_{12} = 0$ and $2B_{12} = -3.0$ ergs/cm^2, $K_1/M = -0.31$ kOe, and $K_u/M = 0.03$ kOe. Note that this calculation produces a rather poor fit above the break in the curve and shows an abrupt rather than a slow approach to saturation. This type of behavior is inherent to the A_{12}, B_{12} coupling form of Eq. (3). We stress the fact that the $|A_{12}/B_{12}|$ ratio is quite small and this is found for all the coupled samples studied here. Such behavior is very unusual as is the large magnitude required for B_{12}. Previously, the largest value of $2B_{12}$ found is -0.3 erg/cm^2 for Fe/Al/Fe [11]. For the $\mathbf{H} \parallel \langle 100\rangle$ magnetization data, this standard coupling form yields a similarly inadequate fit.

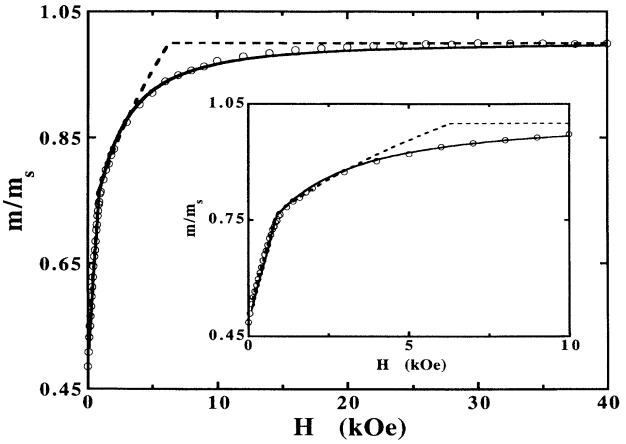

FIG. 2. Field dependence of the magnetization of sample A over an extended field range with $\mathbf{H} \parallel \langle 110\rangle$. Inset: Details of the low field region. The data are indicated by the open circles while fits based on the coupling expressions of Eqs. (3) and (4) in the text are shown by the dashed and solid curves, respectively.

Slonczewski [4] proposed a new form of coupling in which strong deviations from bilinear coupling are attributed to quasiantiferromagnetic exchange coupling within the interlayer itself. Due to competition between areas of differing interlayer thickness which favor either alignment or antialignment of the FM layers, the mean coupling has the following anticipated form,

$$F_c = C_+(\phi_1 - \phi_2)^2 + C_-(\phi_1 - \phi_2 - \pi)^2, \quad (4)$$

where C_+ and C_- are the FM and antiferromagnetic coupling constants and ϕ_i is the in-plane orientation angle of $\mathbf{M}_i$. For $C_+ \approx C_-$, it is easy to show from Eq. (4) that the favored configuration [4] has $\mathbf{M}_1$ and $\mathbf{M}_2$ nearly

90° apart. Note that Eq. (4) is algebraic rather than trigonometric [as Eq. (3) is] in the orientation angles.

The solid line in Fig. 2 is a fit to the data using the coupling form of Eq. (4) with the following least squares parameters: $C_+ = 0.95$ and $C_- = 1.07$ ergs/cm^2, $K_1/M = -0.34$ kOe, and $K_u/M = 0.03$ kOe. This is clearly a much improved representation of the magnetization data, especially in terms of the deficiencies noted earlier for the usual coupling form. Note that $C_+ \approx C_-$ as expected for the zero field orientation found. The fit for the $\mathbf{H} \parallel \langle 100 \rangle$ data using the same parameters (not shown) is similarly improved.

The above data thus offer strong evidence that Eq. (4) is the proper coupling for CoFe/Mn/CoFe and, furthermore, imply that the Mn interlayer is antiferromagnetically ordered, at least for small t(Mn). Recent soft x-ray magnetic circular dichroism and polarized neutron diffraction measurements on a superlattice of CoFe/Mn, with t(Mn) chosen for very strong coupling, support the existence of Mn moments [12] as well as the near-90° orientation of adjacent CoFe layer moments [13]. At present, we do not know whether the bct Mn AFM order is intrinsic at room temperature or if it is stabilized by the adjacent CoFe layers.

Finally, we briefly consider the FMR modes found in the strongly coupled trilayers. The angular dependence of the two modes found is shown via the points in Fig. 3. The high field mode is identified as an in-phase (acoustic) mode and the low field one as an out-of-phase mode (optical) by means of their expected polarization behaviors. The in-phase mode is strong when the microwave magnetic field $\mathbf{h}_1 \perp \mathbf{H}$ and the out-of-phase mode is strong when $\mathbf{h}_1 \parallel \mathbf{H}$. A fit to the angular dependence of these modes was made using the same energy density techniques which we made explicit earlier for Fe/Cr/Fe coupled trilayers [14]. We note that the demagnetizing term $-2\pi(M_{1z}^2 + M_{2z}^2)t$(CoFe) must be appended to the areal energy density F for this calculation.

Using the standard [Eq. (3)] coupling term, one cannot obtain even a qualitatively acceptable fit to these FMR modes. On the other hand, using the algebraic coupling of Eq. (4), one easily obtains the quantitatively correct fits to the average fields of both modes and the qualitatively correct angular dependence of the two modes (including their striking phase reversal) as shown by the solid lines in Fig. 3. This fit has an extra coupling term $C_z(\theta_1 - \theta_2)^2$ added to allow for a different coupling along the tetragonal distortion direction of the Mn layer. Although the distinctive qualitative behavior of the FMR modes is well reproduced, we do not consider the quantitative fit completely acceptable for the out-of-phase mode nor can we fully justify the small C_z/C_+ ratio of 0.67 needed. Rather we suggest that the model used is oversimplified for treating the FMR modes in that it assumes one can treat the FM layers as rigid entities characterized by

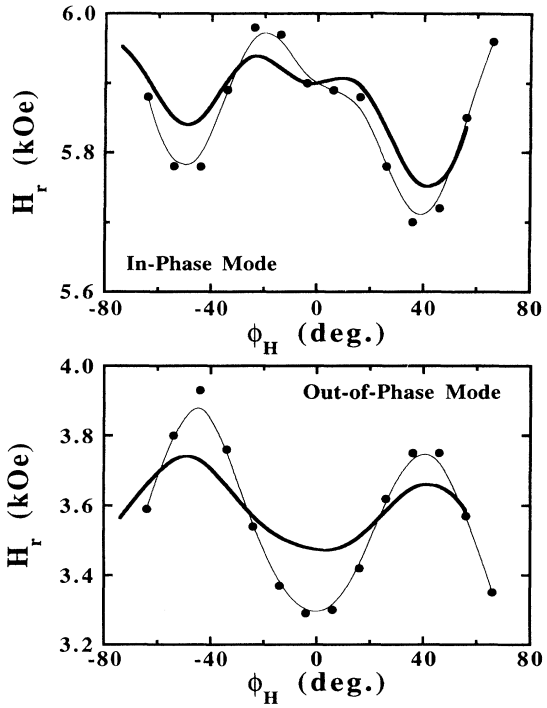

FIG. 3. The in-plane angular dependence of the in-phase and out-of-phase FMR mode positions for sample A. The open dots are the experimental data and the thin line is a guide to the eye. The heavy solid line fit with C_+ ($= C_-$) and C_z equal, respectively, to 1.01 and 0.67 erg/cm^2 is discussed in the text.

unique magnetization angles and that the Mn layer can be replaced by simple interface coupling constants. It seems likely that a more sophisticated model explicitly dealing with the change in magnetic moment direction throughout the Mn layer and into both alloy films ultimately will be needed for a precise treatment of the FMR mode behavior.

The observed strong variation of the in-phase mode angular dependence on t(Mn) can be used to obtain the dependence of the coupling constants on t(Mn). Using this approach, we have found evidence of oscillations in the coupling amplitude along with the usual overall decrease as the interlayer thickness increases. The M vs H fits independently yield very similar coupling constants for the cases considered. A complete report on these thickness studies (including a justification of the analysis used) plus the temperature dependence is planned for a more extended publication.

We believe that this system, which employs metastable bct Mn to couple the ferromagnetic layers, represents a new departure for studies of coupled layers, and the strong near-90° coupling at room temperature may prove useful for technological applications. The existence of this antiferromagnetically mediated coupling in CoFe/Mn suggests that the Fe/Cr system should be reexamined with this in mind.

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