Using Ferromagnetic Resonance as a Sensitive Method to Study Temperature Dependence of Interlayer Exchange Coupling

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We have developed longitudinal pumping and angular dependent ferromagnetic resonance techniques to investigate the temperature dependence of the interlayer exchange field H_{ex} on a series of Co/Ru/Co trilayer films. Our results indicate, for the first time, that the variation of H_{ex} with temperature follows roughly the relationship $H_{ex} \propto (T/T_0)/\sinh(T/T_0)$ as predicted by theoretical models. In addition, no evidence of the biquadratic exchange term was observed in these samples.

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Understanding the interlayer exchange coupling mechanism in magnetic (nonmagnetic) multilayer films has been of great interest since the discovery of oscillations in the exchange coupling strength between successive ferromagnetic layers with respect to the nonmagnetic spacer thickness [1]. It was found that this oscillatory behavior exists in many magnetic multilayer structures with the oscillation period on the order of 10 Å [2]. For extremely good samples with sharp interfaces, another short oscillation period, on the order of two atomic layers, has also been observed [3–5]. Both the long and short oscillation periods have been qualitatively explained as a RKKY-type interaction determined by the detailed topology of the Fermi surface of the spacer material [6–8].

While most of the theoretical calculations were performed at T = 0, almost all of the experimental data were obtained at room temperature. It was found, however, that the exchange coupling strength can be significantly different between room temperature and low temperatures [9]. Using a single spin-split *d*-band model, Edwards *et al.* [10] suggested that the interlayer exchange coupling is based on spatial confinement of *d* holes in the spacer layer. With the assumption of a simple cubic tight binding band model and an analog of the de Haass-van Alphen effect, he predicted that the amplitude of the bilinear exchange coupling coefficient A_{12} increases with decreasing temperature and follows the relationship

$$A_{12} \propto (T/T_0)/\sinh(T/T_0)$$
. (1)

For multilayer films, the characteristic temperature, given by

$$T_0 = \hbar v_F / 2\pi k_B L \,, \tag{2}$$

is on the order of 100 K. Here v_F is the Fermi velocity and *L* is the spacer thickness. The same relation has also been predicted by Bruno and Chappert using a similar approach [11].

Ferromagnetic resonance (FMR) and Brillouin light scattering (BLS) have proved to be useful methods to evaluate the exchange coupling strength for both parallel and anitparallel coupled systems [12–16]. Based on the existing resonance theory [12], we have developed several

sensitive FMR methods to systematically study the temperature dependence of the interlayer exchange coupling in a series of Co/Ru/Co trilayer structures which allows us, for the first time, to compare experimental results to the theoretical models.

For a system consisting of two identical ferromagnetic layers antiparallel coupled with each other, the in-plane dispersion relation is shown in Fig. 1. Only the effective anisotropy field $H_{eff} = 2K_{u2}/M_s - 4\pi M_s$, which includes the demagnetization field and the out-of-plane uniaxial anisotropy field, and the bilinear exchange field $H_{ex} = 2A_{12}/dM_s$ are included in this model. Here *d* is the magnetic layer thickness. At any given field, two resonance modes can be obtained which correspond to the acoustic mode ($\uparrow\uparrow$) and the optic mode ($\uparrow\downarrow$). If the magnetization vector of each ferromagnetic layer is saturated at resonance, the acoustic mode, in which the magnetic

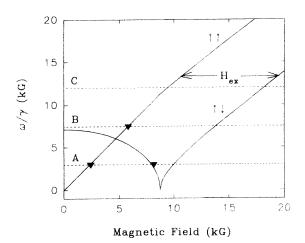


FIG. 1. In-plane dispersion relation for a symmetrical trilayer system. The parameters are $H_{ex} = 8.7 \text{ kG}$, $H_{eff} = 2K_{u2}/M_s - 4\pi M_s = -5.8 \text{ kG}$, $4\pi M_s = 17.6 \text{ kG}$, and g = 2.22, which are consistent with the observation for the Co(32 Å)/Ru(9 Å)/Co(32 Å) sample at room temperature. The solid triangles are experimental data at X-band (A) and K-band (B) frequencies. $\uparrow\uparrow$ and $\uparrow\downarrow$ correspond to acoustic mode and optic mode branches, respectively.

moments in both layers resonate in phase, is independent of the exchange energy; therefore, is degenerate with the resonance mode of noncoupled system. The optic mode, in which the magnetic moment in each layer resonates out of phase, reduces the exchange energy and is on the lower frequency or higher field side of the acoustic mode. The field separation between the optic mode and the acoustic mode is equal to the exchange field H_{ex} at a given frequency. As a result, the temperature dependence of the exchange coupling strength can be obtained if both the acoustic mode and the optic mode are observable in the FMR spectra.

If the two ferromagnetic layers are exactly identical, the optic mode cannot be observed in the FMR spectra. In order to overcome this difficulty, a commonly used method is to make an asymmetrical structure so that the anisotropy energy within each ferromagnetic layer is different from one to the other [15]. However, even with the condition that the thickness of the second ferromagnetic layer is about half that of the first magnetic layer, which is on the order of 10 Å, the intensity of the optic mode is still relatively weak [15], making it difficult to sensitively evaluate the temperature dependence of the exchange coupling strength.

We have developed a longitudinal pumping technique for which the optic mode can be observed even for the symmetrical trilayer structures as shown in Fig. 2(a). Consider the case where the magnetization vectors of the two ferromagnetic layers are unsaturated at resonance due to their antiparallel exchange coupling. In transverse pumping, the microwave field \mathbf{h} is applied normal to the bias field orientation which results in coupling with the acoustic mode and decoupling from the optic mode. However, if \mathbf{h} is applied along the bias field orientation (longitudinal pumping), the optic mode is coupled with \mathbf{h} and produces a noticeable absorption at resonance. When \mathbf{h} is applied at an arbitrary angle with respect to the bias field orientation, both modes are observable in the FMR spectra as shown in Fig. 2(b).

From Fig. 2(a), it is clear that, in the unsaturated state, the dispersion relation of the acoustic mode also depends on the interlayer exchange coupling field H_{ex} because the magnetization vector in each layer no longer stays at a constant angle with each other during the precession. As a result, the interlayer exchange coupling strength can also be evaluated by just using the resonance field of the acoustic mode if the effective internal anisotropy energy and the g value are known for each ferromagnetic layer. These parameters can be accurately determined using angular dependent FMR measurements at one or more frequencies as shown in Fig. 3. Over a certain frequency range (near frequency B in Fig. 1), the angular dependence of the resonance field is very sensitive to small changes in the exchange field and/or the effective anisotropy field as shown in Fig. 3(a), and, therefore, is especially suitable for the temperature dependence measurements and an accurate evaluation of the biquadratic exchange contribution if present.

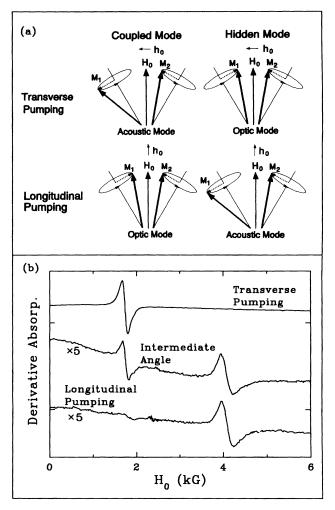


FIG. 2. (a) A schematic diagram of longitudinal and transverse pumping in the FMR setup. h_0 and H_0 are microwave pumping field and bias field, respectively. M_1 and M_2 are magnetization vectors in magnetic layers 1 and 2. (b) In-plane FMR spectra of the sample Co(32 Å)/Ru(10 Å)/Co(32 Å) at X-band frequency and room temperature (using the TE₁₀₂ cavity). The rf field is applied at 90° (top), 20° (middle), and 0° (bottom) with respect to the bias field.

A series of Co(32 Å)/Ru(t_{Ru})/Co(32 Å) trilayer films were prepared in ultrahigh vacuum by evaporation on freshly cleaved mica substrate. Structure analysis using reflection high-energy electron diffraction (RHEED) [17] and transmission electron microscopy (TEM) [18] techniques indicates that the layers are epitaxially grown and have an hcp structure with the c axis normal to the film plane. The high-angle x-ray diffraction measurements on a series of Co/Ru multilayer films show a slight spread of the *c*-axis orientation of about 1.5° [17]. In addition, nuclear magnetic resonance [19] experiments and magnetization measurements [20] on a series of single Co layers deposited on the Ru buffer layer, with the Co layer thicknesses varying from 4 to 30 Å, suggest an intermixing of Co and Ru across two atomic planes at the Co/Ru interface, in agreement with the structure analysis [17].

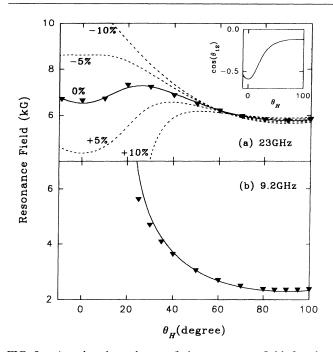


FIG. 3. Angular dependence of the resonance field for the sample Co(32 Å)/Ru(9 Å)/Co(32 Å) at K-band (a) and X-band (b) frequencies. θ_H is the angle between the external field orientation and the z axis which is normal to the film plane. The solid lines are a theoretical fit using the same parameters as in Fig. 1. The broken lines in (a) are predictions corresponding to $\pm 5\%$ and $\pm 10\%$ change in the exchange field H_{ex} . The inset of (a) is the cosine of θ_{12} as a function of θ_H , where θ_{12} is the angle between the magnetization vectors of the two Co layers at resonance. A constant A_{12} used in this fit indicates that the biquadratic exchange term B_{12} is negligible in this sample.

The interlayer exchange coupling as a function of the Ru layer thickness t_{Ru} has been investigated for these Co/Ru/Co trilayer structures at room temperature [21]. To summarize our previous results, we have found that the magnetization vectors of the Co layers are strongly antiparallel coupled in the region $t_{Ru} \le 12$ Å, parallel coupled in the region $14 < t_{Ru} < 18$ Å, and weakly antiparallel coupled in the region $18 < t_{Ru} < 28$ Å.

Four samples from this series have been investigated at low temperatures with $t_{Ru} = 9$, 10, 20, and 24 Å, respectively. All of these samples are antiparallel coupled films at room temperature as their in-plane magnetization curves indicate a linear increase of the induced magnetic moment with the applied field below the saturation value $H_{sat,\parallel} = H_{ex}$. FMR was performed at both the X-band (9.2 GHz) and K-band (23 GHz) frequencies with the temperature ranging from 10 to 300 K. In the longitudinal pumping experiments, the bias field is always applied in the film plane.

For the $t_{Ru} = 20$ and 24 Å samples, the exchange coupling field is about 0.5 kG at room temperature; therefore the magnetic moment in each layer is already saturated at resonance (both *X*-band and *K*-band frequencies). Both the acoustic mode and the optic mode were observed in the in-plane FMR spectra due to a slight difference in the effective anisotropy field between the two Co layers $|H_{eff,1} - H_{eff,2}| \neq 0$ which originates from a slight difference in the growth quality of each Co layer [21]. The analysis from room temperature measurements (angular dependent FMR measurements at X-band and K-band frequencies) suggests that the field separation between the acoustic mode and optic mode in the in-plane resonance spectra at X-band frequencies is close to the exchange field H_{ex} : therefore, is used as an approximation of H_{ex} at each temperature.

For the $t_{Ru} = 10$ Å sample, the exchange field is about 5 kG at room temperature; therefore, the film is not saturated at the X-band resonance condition. The longitudinal pumping technique was applied to this sample and the optic mode was observed with the linewidth of ~ 250 Oe (see Fig. 2), only 5% of the exchange field H_{ex} value. This suggests that even with the existence of surface roughness, the exchange coupling strength is quite uniform across the film, in agreement with the inplane magnetization curve which shows a nearly straight line below the saturation field. The resonance fields of both the acoustic mode and the optic mode increase upon decreasing the temperature and were used to evaluate H_{ex} as well as H_{eff} .

For the $t_{Ru} = 9$ Å sample, H_{ex} is more than 8 kG and the film is not saturated even at *K*-band frequencies. Angular dependent FMR measurements at *K*-band frequencies were used to determine the temperature dependence of H_{ex}

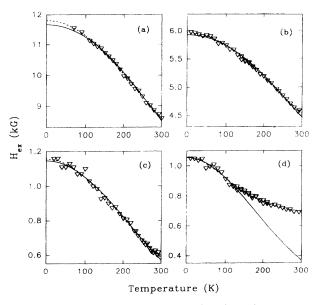


FIG. 4. The exchange field H_{ex} as a function of temperature for various Co(32 Å)/Ru(t_{Ru})/Co(32 Å) structures with t_{Ru} = 9 Å (a), 10 Å (b), 20 Å (c), and 24 Å (d), respectively. The solid lines are best fits using $H_{ex} = H_{ex}^0(T/T_0)/\sinh(T/T_0)$ (theoretical model) and the broken lines are best fits using $H_{ex} =$ $H_{ex}^0(T/T_0)/\sinh(T/T_0) + H_{ex}^\infty$ (modified theoretical model) with the parameters listed in Table I.

TABLE I. Fitting parameters of the interlayer exchange coupling field H_{ex} as a function of temperature for the symmetrical Co(32 Å)/Ru(t_{Ru})/Co(32 Å) structures. These parameters are defined in the caption of Fig. 4. T_0^{th} is the prediction using Eq. (2) with the Fermi velocity $v_F = 1.7 \times 10^7$ cm/s for the theoretical model and $v_F = 1.1 \times 10^7$ cm/s for the modified theoretical model.

| | Theoretical model | | | Modified theoretical model | | | |
|------------------------|-------------------|----------------------------------|-----------------|----------------------------|-------------------------|---------------------|-------------------------------|
| t _{Ru} (Å) | T_0 (±20 K) | $\frac{T_0^{\rm th}}{({\rm K})}$ | H_{ex}^0 (kG) | T_0 (±20 K) | T_0^{th} (K) | $H_{\rm ex}^0$ (kG) | $H_{\rm ex}^{\infty}$ (kG) |
| 9 | 210 | 238 | 11.7 | 145 | 152 | 6.8 | 5.0 |
| 10 | 225 | 218 | 5.9 | 145 | 139 | 3.1 | 2.9 |
| 20 | 135 | 105 | 1.15 | 115 | 67 | 0.94 | 0.22 |
| 24 | 108ª | 87 | 1.07 | 60 | 56 | 0.40 | 0.67 |

^aObtained from the data at low temperatures.

and H_{eff} for this sample (the room temperature data are shown in Fig. 3). The fitting results also agree with the evaluation from the longitudinal pumping technique at X-band frequencies.

From Fig. 4, it can be seen that the exchange field H_{ex} increases with decreasing temperature for all samples, suggesting that the oscillatory exchange coupling in the symmetrical Co/Ru/Co trilayer structures does not change its oscillation period or phase with decreasing temperature, but rather changes its oscillation amplitude. Except for the $t_{Ru} = 24$ Å sample, H_{ex} can be fit by the same relationship [Eq. (1)] predicted by the theoretical models [10, 11] (shown by the solid lines in Fig. 4). For the $t_{Ru} = 24$ Å sample, the experimental data above 120 K are significantly larger than the theoretical predictions. This behavior is opposed to the expectation due to the softening of the interfacial magnetism at high temperatures and has not been fully understood. A better fit to the experimental data can be achieved for all the samples if a constant term A_{12}^{∞} (or H_{ex}^{∞}) was added to the right hand side of Eq. (1) as shown by the dotted lines in Fig. 4. A positive H_{ex}^{∞} was obtained in each sample; however, there is a large error in H_{ex}^{∞} due to the lack of experimental data at T significantly higher than T_0 .

The fitting parameters using both methods are listed in Table I. T_0 decreases with increasing t_{Ru} as expected from the theoretical models [10, 11]. By choosing suitable v_F , the predicted values of T_0 from Eq. (2) agree reasonably well with the experimental evaluation as shown in Table I.

In summary, we have developed sensitive FMR methods and applied them to evaluate the temperature dependence of the interlayer exchange coupling on a series of symmetrical Co/Ru/Co trilayer structures. For the first time, we have proved that the temperature dependence of the interlayer exchange field H_{ex} follows the relationship predicted by the theoretical models and the characteristic temperature T_0 decreases with increasing Ru layer thickness. The excellent fit to the angular dependent FMR data in Fig. 3 along with the in-plane magnetization measurements also suggests that the amplitude of the biquadratic exchange coupling coefficient $|B_{12}|$ is much smaller than $|A_{12}|$ in our Co/Ru/Co trilayer films which have an hcp crystal structure.

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