Interlayer Magnetic Coupling Interactions of Two Ferromagnetic Layers by Spin Polarized Tunneling

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Magnetic interactions involving ferromagnetic layers separated by an insulating barrier have been studied experimentally on a fully epitaxial hard-soft magnetic tunnel junction: Fe/MgO/Fe/Co. For a barrier thickness below 1 nm, a clear antiferromagnetic interaction is observed. Moreover, when reducing the MgO thickness from 1 to 0.5 nm, the coupling strength increases up to $J = -0.26\ \mathrm{erg \cdot cm^{-2}}$. This behavior, well fitted by theoretical models, provides an unambiguous signature of the interlayer exchange coupling by spin-polarized quantum tunneling.

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After the first observation of an antiferromagnetic (AF) interaction of Fe films separated by a Cr spacer [1], the interlayer exchange coupling (IEC) has been subsequently studied with a large variety of metallic spacers [2]. In these systems the oscillation of the coupling strength with spacer thickness has been observed and attributed to the topology of the spacer metal Fermi surface. Theoretically, various models based on either a total energy calculation or models of a Ruderman-Kittel-Kasuya-Yosida–type have been elaborated [2]. Furthermore, a generalization of the IEC theory to nonmetallic (insulating) spacers has been proposed [3–5] by introducing the concept of a complex Fermi surface. However, in the latter case, the IEC, either ferromagnetic or antiferromagnetic, should show a monotonic nonoscillatory variation of its strength with spacer thickness. Experimentally, in the case of nonmetallic spacers, the IEC has been reported for only one system [6,7], namely, a Si based spacer between Fe magnetic layers. In this system for which both a bilinear and a biquadratic coupling are observed [8], the analysis of the contradictory results is rather complex. Moreover, with a semiconductor spacer, the coupling can be related directly to the conduction charge carrier in the Fe-Si spacer layer thermally or optically generated, which may communicate spin information between the Fe layers. Finally, the formation of metallic silicide could explain the observation of oscillatory coupling [9].

In this Letter we provide experimental evidence of room temperature antiferromagnetic coupling between two ferromagnetic (F) layers across a very thin insulating tunnel barrier. Here the spin information and the coupling are carried out across the spacer by equilibrium quantum tunneling of spin-polarized electrons. Our study is performed on the hard-soft magnetic tunnel junction architecture, namely, MgO(100)/Fe/MgO/Fe/Co/V. The materials and the thickness of the layers of our multilayer system were chosen in order to achieve a net AF coupling, as estimated theoretically. The sign of the IEC is a major condition for performing an unambiguous analysis of the interlayer exchange coupling variation when reducing the spacer thickness. Otherwise, a corresponding strong augmentation of a ferromagnetic coupling would be difficult to decorrelate from the direct coupling effects associated with ferromagnetic pinholes in ultrathin spacers.

Theoretically, several model types have been developed to explain the IEC effects, relating to the charge and spin-current transmission between the ferromagnetic (F) layers across an insulating spacer. In the spin-current Slonczewski's model [3,4], the coupling is derived from the torque produced by rotation of the magnetization from one F layer relative to another and is described in terms of a spin-flip current probability calculated from the stationary wave functions of the free-electron Schrödinger equation. The quantum interference model of Bruno [5], associates the coupling with the interferences of the electron waves in the barrier due to the spin reflections at the interfaces. The coupling is expressed in terms of the spin asymmetry of the reflections. This model extends for both metallic and insulating spacers by introducing the concept of complex Fermi surface in the case of insulators. It predicts the temperature variation of the coupling which reduces to the Slonczewski's spin-current model for $T = 0$ K. In addition, we may cite the more sophisticated models implicating the nonequilibrium Keyldysh formalism [10,11] developed to calculate the spin-polarized tunnel current and its connection to the interlayer exchange interaction in thin planar junctions out of equilibrium. They have shown that a nonequilibrium bias across a tunnel junction system may significantly alter the amplitude and the sign of the coupling and that there is a component of the interaction energy between the ferromagnets proportional to their thickness. However, in the absence of external bias, when the ferromagnetic/insulator/ferromagnetic trilayer lies in the equilibrium state, these models reduce again to the equilibrium Slonczewski's spin-current model. Indeed, within the framework of this last model, which has a high physical transparency, the coupling strength J is directly correlated to intrinsic physical parameters of the insulating barrier (width d, height u) and to the free-electron band structure parameters of the ferromagnetic/insulating/ferromagnetic trilayer system: the Fermi energy E_F, the wave vectors of spin up ($k_\uparrow$) and spin down ($k_\downarrow$) electrons in the ferromagnets and in the insulating layer (k), the Stoner splitting in the ferromagnets Δ, and the effective mass of the electron m_{Fe}. When a two-band model is used to describe the ferromagnets, the coupling strength is

$$J = \frac{(U - E_F)}{8\pi^2 d^2} \frac{8k^3(k^2 - k_\uparrow k_\downarrow)(k_\uparrow - k_\downarrow)^2(k_\uparrow + k_\downarrow)}{(k^2 + k_\uparrow^2)^2(k^2 + k_\downarrow^2)^2} e^{-2kd}. \tag{1}$$

For the estimations of the coupling strength, we use bulk band structure parameters [12] for Fe: $k_\uparrow \simeq 1.09$ Å^{-1} and $k_\downarrow \simeq 0.43$ Å^{-1} are extracted from $k_\sigma = \sqrt{(E_F + \Delta\sigma)2m_{\mathrm{Fe}}/\hbar^2}$ (where $\sigma = \pm 1/2$), which correspond to $E_F \simeq 2.6$ eV and $\Delta \simeq 3.6$ eV.

With these values, the IEC coupling is expected to be antiferromagneticlike (AF) when $k^2 < k_\uparrow k_\downarrow = 0.469$ Å^{-2}. By using a reasonable value for the effective mass of the electron in the barrier, m_i, and an experimental determination [13] of the barrier height $u = U - E_F$, the above equation, and the relation $k = \sqrt{(U - E_F)2m_i/\hbar^2}$, a net AF coupling in the Fe/MgO/Fe system is predicted.

Within the same range of parameters, the temperature variation of the coupling strength estimated using Bruno's model,

$$J(T) = J(0\ \mathrm{K}) \frac{2\pi m k_B T d/\hbar^2 k_F}{\sinh(2\pi m k_B T d/\hbar^2 k_F)}, \tag{2}$$

predicts no significant difference of the coupling strength between $T = 0$ K and the room temperature; k_B is the Boltzmann constant, m is the mass of electron, T is the temperature, and $k_F = ik$ is the complex wave vector of the electron in the insulating layer. Thus, the quantitative analysis of the experimental variation of the coupling strength with t_{MgO} obtained at room temperature, can be achieved within the framework of the interlayer exchange theories [3,5].

The epitaxy of metal/insulator superlattice MgO(100)/Fe in ultrahigh vacuum is very well established [14–16]. By using molecular-beam epitaxy (MBE) a two-dimensional growth mode of MgO on Fe was obtained with high quality ultrathin layers without pinholes and with very flat surfaces. The growth conditions have been detailed in our previous study [17]. Briefly, after annealing the MgO substrate at 500 °C for 20 min, first a 50-nm-thick Fe layer is deposited, then annealed at 450 °C for 15 min. Then, the thin MgO insulating layer is subsequently deposited by means of an electron gun. We observe a two-dimensional layer-by-layer growth of MgO up to 10 to 15 monolayers asserted by reflection high-energy electron diffraction (RHEED) intensity oscillations and oscillations of the in-plane lattice parameter [18]. The observation of clear RHEED intensity oscillations (Fig. 1) gives access to a precise determination of t_{MgO} with a low absolute uncertainty, certainly below ± 0.05 nm, and even better relative accuracy. The second magnetic electrode is a bilayer composed by a 5-nm-thick Fe layer, epitaxially grown on the top of the MgO barrier magnetically hardened by a 50-nm-thick Co layer deposited on the top of it. The continuity of the insulating MgO layer has been previously checked down to 0.8 nm thickness, at different spatial scales by means of morphological (high resolution transmission electronic microscopy), electrical (the local impedance), magnetoresistance measurements, and down to 0.5 nm in the present work by magnetic measurements. As a similar example, MgO(100)/Fe/MgO/Fe/Co/Pd tunnel junctions have shown tunnel magnetoresistance up to 17% for a 1 nm thick MgO layer [17].

The magnetic properties have been investigated by a superconducting quantum interference device and alternating gradient field magnetometers. Magnetization versus field loops have been performed on continuous multilayer films with lateral sizes above a few millimeters, in order to avoid spurious antiferromagnetic dipolar coupling introduced by patterning of small size devices. In these films, the MgO thickness ranges from 0.4 to 2.5 nm. Because of the epitaxial growth, both soft and hard layers present fourfold symmetries [17], with the same directions for the easy axis. The contrast between their coercive fields is significant: $H_c = 40$ Oe for the soft layer and $H_c > 350$ Oe for the hard layer. This will define in the hysteresis loop a large field window where one of the magnetic layers is magnetically rigid, while the other layer can easily be turned by a small external field. Therefore, the interlayer

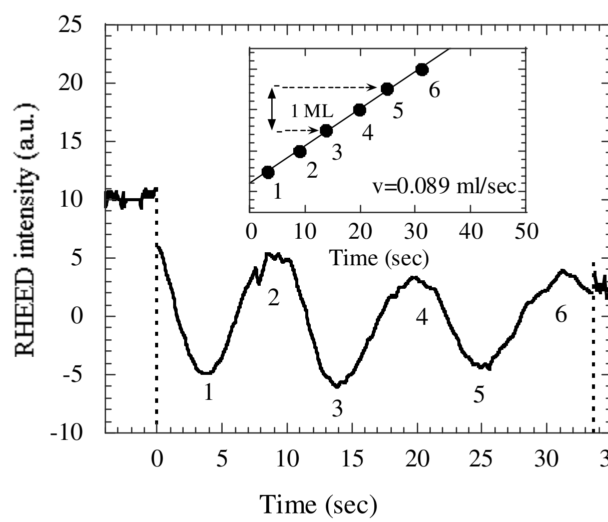

FIG. 1. RHEED intensity during the deposition of 79 nm thick MgO. The period of oscillations corresponds to the growth of 1 monolayer. In order to determine the rate of the growth we plot (inset) the positions of the maxima and minima as a function of time.

magnetic coupling can be extracted from the shift of the minor hysteresis loops, taken for the soft magnetic layer in a field window where the hard layer is magnetically ''locked'' by a previous magnetization saturation.

For a spacer thickness $t_{\mathrm{MgO}} < 0.8$ nm, we observe clearly (Fig. 2) a net positive shift of the M-H minor loop. Such a shift can be explained by the IEC through the insulating layer, but it could also be attributed to an exchange biasing of the first Fe layer by a possible antiferromagnetic/ferrimagnetic oxide layer at the interface between the bottom Fe layer and the oxide insulating barrier. The exchange bias hypothesis would lead to a coupling mainly independent of the insulating spacer thickness and/or should also be present in samples without the second top hard magnetic layer. However, in our samples we observe a fast dependence of the measured AF coupling strength J with the spacer thickness, as discussed below. The rapid variation of the coupling with the thickness of the spacer is directly illustrated in the inset of Fig. 2, where we can see that by increasing the spacer thickness from 0.5 to 0.63 nm the shift reduces drastically from 58 to 7.5 Oe. Moreover, on simplified samples where we excluded on purpose the hard (top) layer: MgO(100)/Fe/MgO, we observe no shift of the M(H) loops. At least the shift is below the uncertainty of the measurement setup (1 Oe), whereas for the Fe/MgO/Fe/Co multilayer a shift up to 133 Oe, has been obtained for the $t_{\mathrm{MgO}} = 0.5$ nm layer. Consequently, we can exclude the occurrence of the AF biasing. Therefore, the observed field shift of the M-H minor loops can be unambiguously attributed only to interlayer coupling effects.

The coupling energies, J, have been extracted from the M-H minor loops for all the samples. J is calculated as the product between the field offset of the minor M-H curves (see Fig. 2) and the magnetization of the soft magnetic layer. Conventionally, we associated the sign of J with the type of the coupling: antiferromagnetic ($J < 0$) and ferromagnetic (F) coupling ($J > 0$). Three regimes can be clearly distinguished: An AF coupling ($J < 0$) is measured for $t_{\mathrm{MgO}} < 0.8$ nm, with a very fast increase of amplitude ($|J|$), when the thickness of the spacer is reduced from $t_{\mathrm{MgO}} = 0.8$ to 0.5 nm (Fig. 3).

Below 0.5 nm, we observe unambiguously a modification of the shape of the magnetization reversal, and a decrease of the apparent coupling strength. Indeed, with such a low interlayer thickness, we expect the occurrence of pinholes, and consequently a direct ferromagnetic coupling competing with the AF exchange coupling studied here. This leads to significant deviations from the pure bilinear coupling interaction and can be simulated by a biquadratic interaction, which could also explain the shape of the magnetic hysteresis loops. For thicker insulating layer, we cannot exclude the occurrence of any pinholes. However, for thicker insulators the measured minor hysteresis loops are square. Therefore, we can reasonably assume that above 0.5 nm the contribution of direct coupling via ferromagnetic pinholes is certainly much smaller than the one of the AF exchange interaction.

On the other hand, for larger spacer thickness, namely, above 1 nm, we observe always a net ferromagnetic coupling. We may easily attribute this F coupling to the well known ''Orange Peel'' interaction [19], associated with the

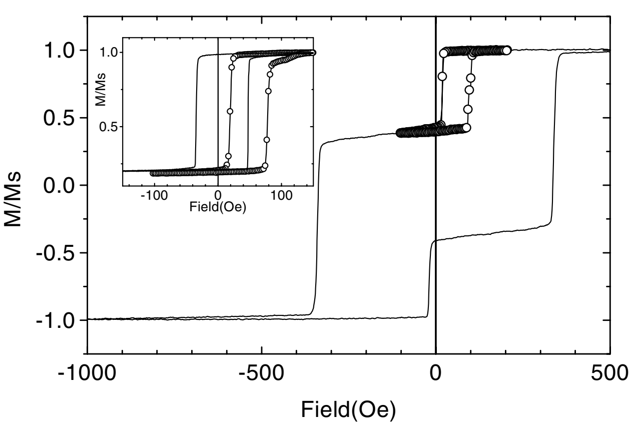

FIG. 2. Magnetization curve along the easy axis for MgO(100)/Fe/MgO(0.5 nm)/Fe/Co. The minor loop (- ∘ -) is taken after a positive saturation of the whole system, in a field window where the hard Fe/Co bilayer is magnetically rigid. In this sample, the positive shift of 58 Oe in the minor loop is the signature of a strong AF coupling. Inset: The rapid variation of the coupling strength with the thickness of the insulator is reflected by two minor loops, (—) $t_{\mathrm{MgO}} = 0.63$ nm and (- ∘ -) $t_{\mathrm{MgO}} = 0.5$ nm.

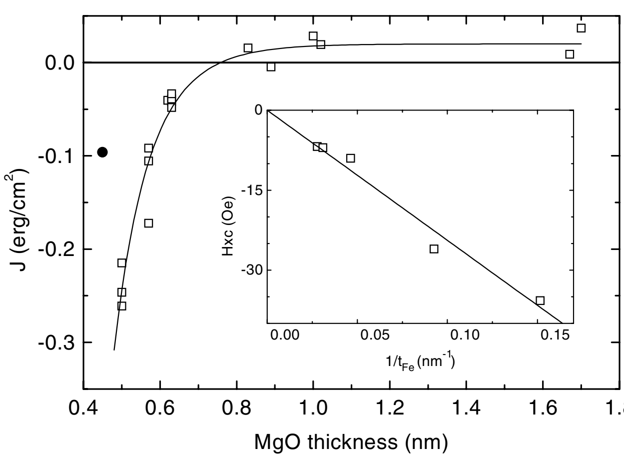

FIG. 3. Variation of the coupling strength J with the insulator thickness. The experimental data are represented by empty square features. Theoretical estimation of J, performed within the framework of the spin-polarized tunneling of Slonczewski, is illustrated by the filled line. For $t_{\mathrm{MgO}} = 0.45$ nm (point represented by a filled circle) the net coupling is still AF, but it is reduced by the ferromagnetic pinholes contribution. Inset: Variation of the exchange field with the thickness of the soft ferromagnetic layer.

correlated roughness of the ferromagnetic/insulator interfaces. Having in view the large fluctuation length of the roughness determined by high resolution transmission electron microscopy in our epitaxially grown layers (> 10 nm), the orange peel coupling is basically constant in the thickness range involved in our study. Moreover, as we already discussed in a previous paper [17], because of the high quality of the two-dimensional growth, this coupling is small, i.e., lower than 0.04 erg/cm^2.

With a surface interaction, we expect a linear variation (linear increase) of the coupling field with $t_{\rm Fe}^{-1}$, where $t_{\rm Fe}$ is the thickness of the soft magnetic layer. Experimental results presented in the inset of Fig. 3, and obtained on three different epitaxies with the same spacer thickness $t_{\rm MgO} = 0.62$ nm, are in good agreement with this expectation. In one of the epitaxies, three different Fe thicknesses have been obtained for the same MgO layer, by using shadow masks during the growth of the soft magnetic layer. Therefore, we confirm that the observed shift is due to a surface interaction. Moreover, since the dependence of the AF coupling with $t_{\rm MgO}$ is abrupt as discussed below, the reproducibility, and then the relative determination, of the spacer thickness is very good.

We present also in Fig. 3, the theoretical variation of J with $t_{\rm MgO}$, estimated from Eq. (1) (Full line). For the calculation we have used first the bulk Fe band structure parameters, ($k_\uparrow \simeq 1.09$ Å^{-1}, $k_\downarrow \simeq 0.43$ Å^{-1}, and $E_F \simeq 2.6$ eV) [12], and, second, reasonable parameters for the insulating barrier: a barrier height of $U - E_F = 1$ eV and an effective mass in the barrier $m_{\rm eff} = 0.4m_0$. Indeed, through a determination of the prefactor and the exponential decay length in Eq. (1), we could expect an independent determination of $u = U - E_F$ and $m_{\rm eff}$. However, it would require an even greater ''accuracy'' in evaluating the insulating layer thickness $t_{\rm MgO}$. Finally, the orange peel coupling is described in terms of a constant positive "coupling offset" of 0.02 erg/cm^2, which corresponds to the average value observed for spacer thickness above 1.2 nm, and it also represents a reasonable assumption having in view the roughness fluctuation length in our epitaxial samples. From Fig. 3, we can conclude that the experimental variation of the coupling strength with the insulating spacer thickness is well fitted in the framework of the Slonczewski's spin-current model. Moreover, we obtain an estimation of the relationship between the barrier height and the effective mass in the barrier: namely, $(U - E_F)m_{\rm eff} = 0.44$ eV. Finally, let us note that the experimental data cannot be fitted by a simple exponential law $J\alpha e^{-2kd}$. The observation of a faster variation, namely, $J\alpha e^{-2kd}/d^2$, is a clear signature of the equilibrium spin-current IEC model.

In summary, antiferromagnetic interlayer coupling through an insulating spacer has been unambiguously evidenced. The shape of the variation of the experimental coupling strength J with the insulating spacer thickness $t_{\rm MgO}$, the quantitative value of $|J|$, and finally the thickness range of $t_{\rm MgO}$ for which the antiferromagnetic coupling is observed represent an experimental proof of the interlayer exchange theory [3,5] by the spin-polarized quantum tunneling of electrons between the ferromagnetic layers.

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