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Antiferromagnetic coupling by spin polarized tunneling

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By performing magnetic studies on Fe/MgO/Fe magnetic tunnel junctions, we provide experimental evidence of room-temperature antiferromagnetic coupling between two ferromagnetic layers across a very thin insulating barrier. Epitaxial growth of the MgO barrier on a very flat Fe layer leads to an extremely low ‘‘orange peel’’ magnetic coupling. Then, antiferromagnetic coupling is observed for MgO thickness, t_{MgO}, below 0.8 nm. The strength of this coupling increases abruptly when reducing t_{MgO} down to 0.5 nm. The shape of the variation of experimental coupling strength J with t_{MgO}, the quantitative value of $|J|$, and finally, the thickness range of t_{MgO} for which the antiferromagnetic coupling is observed are in good agreement with the equilibrium interlayer exchange theory by the spin polarized quantum tunneling of electrons between the ferromagnetic layers. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1540175]

Interlayer exchange coupling has been observed with a large variety of metallic spacers.[1,2] In these systems the oscillation of the coupling strength with the spacer thickness is attributed to the topology of the spacer metal Fermi surface.[2] Generalizations of the theory to insulating spacers have then been proposed. In the spin-current model of Slonczewski,[3,4] the coupling is derived from the torque produced by rotation of the magnetization from one ferromagnetic (F) layer relative to another, being 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] introduces the concept of complex Fermi surface to extend the interlayer exchange coupling (IEC) theory for insulators (I). It predicts the temperature variation of the coupling and reduces to the Slonczewski's spin-current model for $T=0\,\mathrm{K}$. More-sophisticated models implicating the nonequilibrium Keyldysh formalism[6,7] 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 absence of external bias, these models reduce again to the equilibrium spin-current model of Slonczewski. Within the framework of this model, coupling strength J is given by

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

where, d and U are the width and the height of the insulating barrier; E_F the Fermi energy of the F/I/F system; and k, $k_\uparrow$ and $k_\downarrow$ are the wave vectors of electrons in the insulating layer and of spin-up and spin-down electrons in the ferromagnets, respectively.

Our study is performed on a hard–soft magnetic tunnel junction architecture, namely, MgO(100)//Fe/MgO/Fe/Co/V, elaborated in ultrahigh vacuum by molecular beam epitaxy (MBE). After annealing the MgO substrate at 500 °C for 20 min, first a 50-nm-thick Fe layer has been deposited using a Knudsen cell at a 0.7 nm/min rate. The iron layer grows pseudomorphically on the MgO(100) substrate, and the lattice mismatch is 3.7% when the Fe unit cell is turned by 45° with regard to the MgO unit cell. To improve the surface quality, the Fe layer was annealed at 450 °C for 15 min. The surface rms roughness after annealing, estimated from atomic force microscopy (AFM) images, was about 0.3 nm. A thin MgO insulating layer was subsequently deposited by means of an electron gun at 0.5 nm/min. We found that insulating barriers of thickness from 0.5 to 3 nm grew epitaxially on the Fe layer. Two-dimensional layer-by-layer growth[8–10] was observed up to several monolayers by means of reflecting high energy electron diffraction (RHEED) intensity oscillations. The observation of in-plane lattice parameter oscillations (Fig. 1) is further evidence of high quality layer-by-layer growth.[11] The second magnetic 5-nm-thick Fe layer has been epitaxially grown on the top of the insulating MgO barrier. Then, a Co layer with a thickness of 50 nm was deposited on the top of iron using an electron gun at 3 nm/min. RHEED images indicate clearly epitaxial growth of Co on Fe. To prevent the *ex situ* oxidation of the top Co layer and to protect it during the subsequent patterning steps of the lithography, we have used a 10-nm-thick V capping layer. The continuity of the insulating MgO layer has been also checked, at different spatial scales by high resolution transmission electronic microscopy (HRTEM) (Fig. 2), by magnetic measurements (Fig. 3), and by local impedance and magnetoresistance measurements.[12]

Magnetization versus field loops have been performed on continuous multilayer films with lateral sizes above a few millimeters, in order to avoid spurious antiferromagnetic di-

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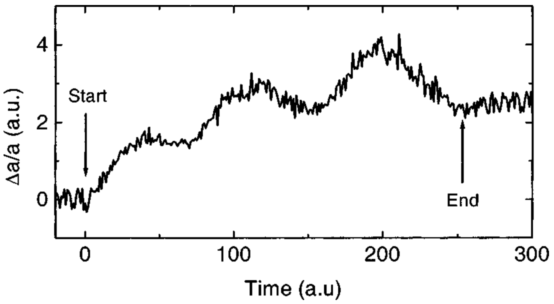

FIG. 1. Lattice parameter oscillations during the MgO insulator growth.

polar coupling introduced by patterning of small size devices. For a spacer thickness $t_{\mathrm{MgO}}<0.8\ \mathrm{nm}$, we observe clearly a net positive shift of the $M-H$ minor loops (Fig. 3). Experimental results presented in Fig. 3 are obtained on the same sample with an identical MgO layer ($t_{\mathrm{MgO}}=0.58\ \mathrm{nm}$). Three different Fe thicknesses have been obtained for different parts of the sample by using a set of shadowing masks during the growth of the soft magnetic layer. Clearly, the coupling field, H_{ex}, is observed to increase when the thickness, t_{Fe}, of the soft magnetic layer decreases. Indeed, with a surface interaction we expect a linear increase of H_{ex} with t_{Fe}^{-1}. The observation of such linear variation (inset of Fig. 4) on samples obtained on three different epitaxies with the same spacer thickness (namely, $t_{\mathrm{MgO}}=0.62\ \mathrm{nm}$) is then a signature of the interfacial antiferromagnetic (AF) coupling.

Due to the fourfold magnetic symmetries and the significant contrast between the coercive fields of "hard" and "soft" layer, coupling energies J have been extracted from the $M-H$ minor loops.[13] The value of J is then calculated as the product between the field offset of the minor $M-H$ curves and the magnetization of the soft magnetic layer. Conventionally, we associated the sign of J to the type of the coupling: AF ($J<0$) and F ($J>0$). Three regimes can be clearly distinguished. First, an AF coupling is measured for $t_{\mathrm{MgO}}<0.8\ \mathrm{nm}$, with a very fast increase of amplitude ($|J|$) when the thickness of the spacer is reduced from $t_{\mathrm{MgO}}=0.8-0.5\ \mathrm{nm}$. Second, 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 explain also a rounded shape of the magnetic hysteresis loops observed within this thickness range. For thicker insulating layers, even if we cannot completely exclude the occurrence of pinholes, the measured minor hysteresis loops are square. Therefore, we can reasonably assume that above 0.5 nm the contribution of the direct coupling via ferromagnetic pinholes is certainly much smaller that the one of the AF exchange interaction. Finally, 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,[14] associated with the correlated roughness of the ferromagnetic/insulator interfaces. The fluctuation length of the roughness is estimated by HRTEM to be above 10 nm. So, we can consider the orange peel coupling to be basically constant, and equal to the average value observed for spacer thickness above 1.2 nm, namely, 0.02 erg/cm^2.

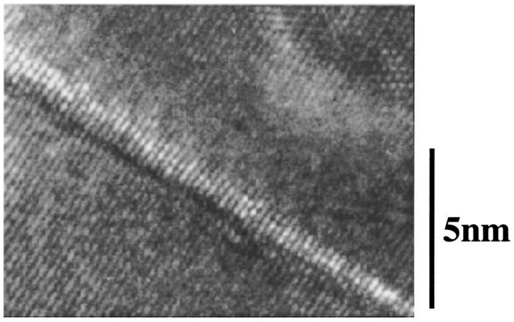

FIG. 2. Cross section high resolution TEM image for a 0.45-nm-thick MgO barrier illustrating the pseudomorphic epitaxial growth of Fe/MgO/Fe.

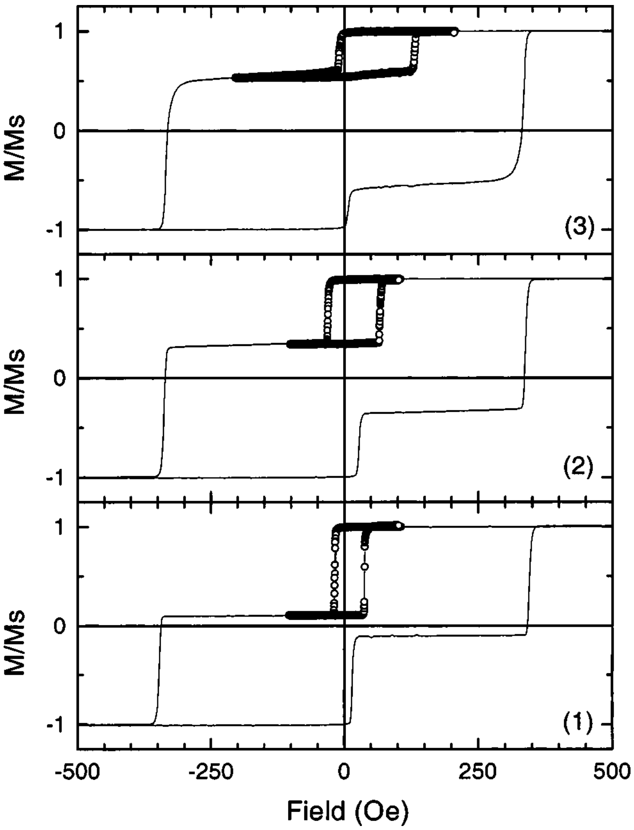

FIG. 3. Magnetization curve along the easy axis for MgO(100)/Fe(x)/MgO(0.58 nm)/Fe(5 nm)/Co(55 nm). The bottom Fe is deposited using a shadowing mask to obtain on the same sample three different Fe thicknesses: $x=50$ nm (1), 33.5 nm (2), and 16.5 nm (3), for the same insulator and top Fe/Co thicknesses. The minor loops (-○-) are taken after a positive saturation of the whole system, in a field window where the hard Fe/Co bilayer is magnetically rigid.

The variation of the coupling strength J with t_{MgO}, estimated from the above equation[3] is also plotted in Fig. 4. For the calculation we have used: (i) bulk band structure parameters for Fe, namely,[15] $k_{\uparrow}\simeq 1.09\ \mathrm{\AA}^{-1}$ and $k_{\downarrow}\simeq 0.43\ \mathrm{\AA}^{-1}$; (ii) reasonable parameters for the insulating barrier: a barrier

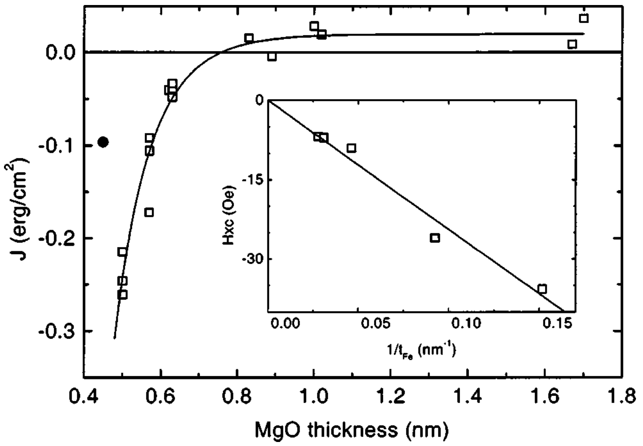

FIG. 4. Variation of coupling strength *J* with the insulator thickness. Experimental data are represented by empty square features. Theoretical estimation of *J*, performed within the framework of the spin polarized tunneling of Slonczewski, is illustrated by the filled line. For $t_{\mathrm{MgO}}=0.45$ nm (point represented by a black 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 for $t_{\mathrm{MgO}}=0.62$ nm.

height of $U-E_F=1$ eV and an effective mass in the barrier $m_{\mathrm{eff}}=0.4\,m_0$; and finally, (iii) a constant positive "coupling offset" of 0.02 erg/cm^2 to describe the orange peel coupling. It appears clearly on Fig. 4 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, where $J\alpha e^{-2kd}/d^2$. In conclusion, our experimental results strongly support the equilibrium interlayer exchange theory by the spin polarized quantum tunneling of electrons between the ferromagnetic layers.

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