Novel Oscillation Period of the Interlayer Exchange Coupling in Fe/Cr/Fe Due to MgO Capping

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A novel period of the interlayer exchange coupling as a function of Cr thickness is observed in epitaxial Fe/Cr/Fe (001) sandwiches capped with MgO. This additional period, equal to 3 chromium atomic layers, vanishes when the capping is Cr. A strong oscillation of the magnetic coupling is also observed as a function of the thickness of the Fe layer next to the MgO capping layer. This effect is attributed to the formation of quantum well states in this Fe layer. It is believed that this confinement modifies the reflection coefficient at the Cr/Fe interface for electrons of a particular symmetry and leads to the new coupling period which is linked to the Fermi surface topology of chromium.

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Interlayer exchange coupling (IEC) between ferromagnetic films separated by a thin nonmagnetic metal layer has been the key for the discovery of giant magnetoresistance [1,2] and the development of magnetoresistive devices. Indeed, some of the main applications for spintronics rely on this effect that can couple or decouple the magnetization of two ferromagnetic films. The oscillatory behavior of the IEC with spacer thickness, on the other hand, was first observed by Parkin *et al.* [3] in Fe/Cr/Fe (001) multilayers. It allows tuning the strength of the coupling and thus the coercive fields of the ferromagnetic layers by choosing the appropriate spacer layer thickness.

The oscillatory behavior of the IEC is due to spin-dependent confinement of electrons in the spacer layer, leading to quantum interferences as a function of the spacer layer thickness. Its physical origin lies in the particular electronic properties of the spacer layer material. Indeed, all models developed so far to understand this phenomenon [4–8] show that the oscillation period of the IEC as a function of the spacer layer thickness is determined by the Fermi surface topology of the spacer material, more precisely by the presence of so-called stationary spanning vectors. However, since the strength of the electron confinement in the spacer layer depends on the electronic density of states in the ferromagnetic layers, the IEC is expected to depend also on the ferromagnetic layer thickness. A thickness variation of the latter should therefore modulate the coupling strength [6,9]. In fact, experiments in which the ferromagnetic layer thickness was varied showed weak oscillations of the coupling strength [10,11]. A similar but still weaker effect was observed as a function of the capping layer thickness [12]. However, these latter effects did not reveal a novel period of the IEC as a function of the spacer layer thickness.

Here we study Fe/Cr/Fe (001) sandwiches in which the bilinear magnetic coupling constant J_1 shows two oscillation periods as a function of the Cr thickness [13]: a short one of 2.1 monolayers (ML; 1 ML Cr corresponds to 0.144 nm) and a long one of about 12 ML. The short period can only be observed in films with very little roughness of the Cr spacer layer [14], whereas rougher films exhibit the long period. Things are in fact more complicated as both periods can be simultaneously observed and the biquadratic magnetic coupling constant J_2 has to be taken into account [15].

In the following, we show that capping the thin top Fe layer by an insulating MgO layer leads to the appearance of a novel period (3 ML) of the IEC as a function of the Cr thickness. The insulating layer reflects electrons and leads to the formation of quantum well states in the thin Fe layer next to it. It thus modifies the electronic density of states at the Fermi level in Fe and hence influences the reflection coefficient at the Cr/Fe interface. This in turn leads to a modification of the electron confinement in Cr, and in this way influences the IEC across Cr.

The epitaxial bcc (001) oriented heterostructures are deposited by molecular beam epitaxy in an ultrahigh vacuum chamber at a base pressure of 10^{-10} Torr. A 125 ML Fe buffer layer (1 ML Fe corresponds to 0.1435 nm) is deposited on a MgO substrate at room temperature and annealed at 450 °C for 1 h. In order to measure the IEC as a function of the Cr spacer thickness, wedge-shaped samples are prepared by continuously moving a shutter in front of the sample during deposition of Cr. Unless specified, the growth after the annealing of the Fe buffer layer is carried out at 80 °C. Then, about 42 ML of MgO are deposited before encapsulation with Pt. We note that recent transport measurements on epitaxial Fe/Cr/Fe/MgO/Fe samples prepared in this manner revealed symmetry-resolved quantum well states in Fe [16], thus proving the high epitaxial quality of the films.

In order to observe the effect of the capping layer on the IEC, we deposited on a Fe(125 ML)/Cr(13 ML)/Fe(14 ML) sample, a Cr capping layer on one part of the sample and a MgO capping layer on another by using a shadow mask during evaporation. Magneto-optical Kerr

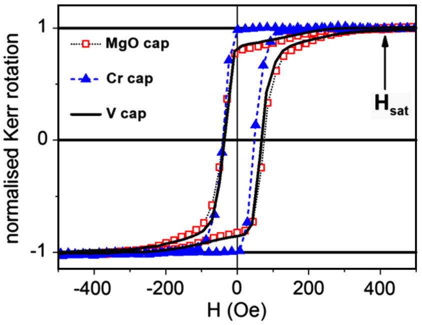

FIG. 1 (color online). Magneto-optical Kerr rotation of a Fe(125 ML)/Cr(13 ML)/Fe(14 ML)/cap epitaxial sample: (squares) with MgO capping layer, (triangles) Cr capping layer, (full line) V capping layer. The measurements are performed in the longitudinal configuration with the magnetic field applied along the Fe [100] easy magnetization direction.

measurements (Fig. 1) show that in the Cr-capped part the Fe layers are ferromagnetically coupled. This is as expected, since the maximum of the ferromagnetic coupling strength in the case of the long period oscillations is at around 13 ML Cr spacer thickness [3]. On the other hand, the MgO-capped part of the sample clearly shows an antiferromagnetic coupling between the Fe layers, with a saturation field H_{sat} close to 400 Oe. Thus, the capping by MgO influences the coupling strength so strongly that it leads even to a sign change.

Systematic measurements were performed as a function of the Cr spacer thickness with both kinds of capping layers. Figure 2(a) shows H_{sat} measured along the Cr wedge of a MgO-capped sample. A Fourier analysis of the data reveals the existence of two periods. Indeed, assuming two contributions for the bilinear coupling as a function of the Cr thickness d [5,6]:

$$J_1 = \frac{A_2}{d}\sin\left(2\pi\frac{d}{T_2}+\phi_2\right)+\frac{A_3}{d^2}\sin\left(2\pi\frac{d}{T_3}+\phi_3\right), \quad (1)$$

with T_2 and T_3 the periods of a 2.1 ML- and a 3 ML-period sinusoid, respectively, ϕ_2 and ϕ_3 the corresponding phases, and A_2 and A_3 the corresponding amplitudes we reproduce qualitatively well the main oscillation effects observed in our measurements. We assumed a $1/d$ dependence of the amplitude of the 2.1 ML oscillations and a $1/d^2$ dependence of the 3 ML oscillations as expected for oscillations associated to a nesting and a stationary spanning vector, respectively [6]. However, the fit does not reproduce well the amplitude of the oscillations and we cannot rule out the presence of the 12 ML long period. In fact, the latter cannot be distinguished from the envelope of the sum of both short period oscillations.

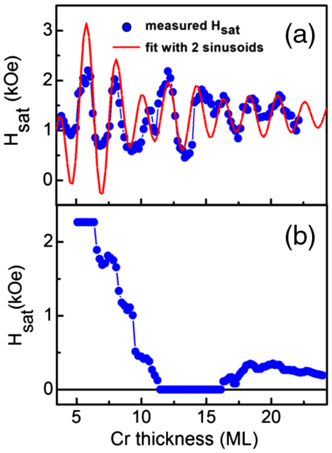

FIG. 2 (color online). Saturation field H_{sat} measured along the Cr wedge of a Fe(125 ML)/Cr(x ML)/Fe(14 ML)/cap sample. (a) With a MgO capping layer. The fit (full line) is performed with H_{sat} proportional to $2 \times (J_1 + 2J_2)$, J_2 being taken constant, and J_1 being given by Eq. (1). The parameters used are: $T_2 = 2.1$ ML, $\phi_2 = 1$ rad, $T_3 = 3.04$ ML, $\phi_3 = 1.23$ rad, and $A_3/A_2 = 8.7$ (b) With a Cr capping layer.

In comparison, the Cr-capped sample shows a dominant 12 ML long period [see Fig. 2(b)] with an additional very weak short period of 2.1 ML. Another striking feature is that the 3 ML period disappears in MgO-capped samples when the growth temperature goes beyond 150 °C. In this case, the dependence of H_{sat} as a function of the spacer thickness (not shown) is the same as in the case of Cr-capped samples grown at 80 °C.

Furthermore, in the case of MgO-capped samples grown at 80 °C, a strong biquadratic coupling constant J_2 can be inferred from the magnetic hysteresis cycles. Indeed, absolute measurements of the magnetization by alternating gradient force magnetometry show that the magnetization of the thin top Fe layer at remanence is oriented almost perpendicularly with respect to the thick bottom Fe layer [see inset in Fig. 3(a)], which is corroborated by micromagnetic calculations [17]. Moreover, the linear dependence of $M(H)$ close to $H = 0$ is also consistent with a perpendicular magnetic configuration of the Fe layers. This biquadratic coupling leads to an increase of the saturation field proportional to J_2 [18]. Since the biquadratic coupling does not oscillate with Cr thickness [14], it thus explains the offset observed in Fig. 2(a), which makes H_{sat} always positive. Without biquadratic coupling, H_{sat} would change sign and the system would thus oscillate between the ferromagnetic and the antiferromagnetic configuration.

To further investigate the effect of the capping layer on the IEC, we also studied H_{sat} as a function of the top Fe layer thickness. Here again, with a MgO capping layer, oscillations are observed [Fig. 3(a)], having a surprisingly strong amplitude that decreases very slowly with increasing Fe layer thickness. The average period of the oscillations is 5.8 ML. The presence of a strong biquadratic coupling as in Fig. 2(a) also prevents H_{sat} from changing sign. A similar sample, but capped with Cr, only shows a ferromagnetic coupling between the Fe layers (not shown), as expected in the case of a dominant long period for a

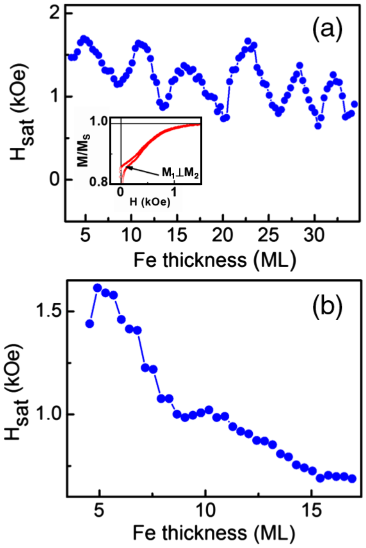

FIG. 3 (color online). Saturation field $H_{\rm sat}$ measured along a Fe wedge: (a) on a Fe(125 ML)/Cr(13 ML)/Fe(x ML)/MgO sample. Inset: part of the alternating gradient force magnetometry hysteresis cycle showing a perpendicular configuration of the thin top Fe layer ($x = 15$ ML). Note the linear behavior of $M(H)$ close to $H = 0$. (b) on a Fe(125 ML)/Cr(8 ML)/Fe(x ML)/Cr sample.

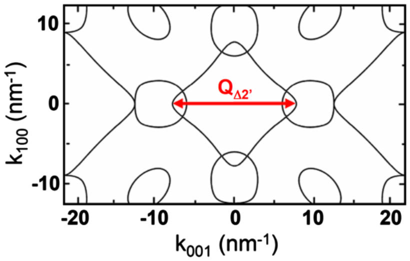

FIG. 4 (color online). Cross section of the bulk Cr Fermi surface perpendicular to the [010] direction in bulk Cr. The stationary spanning vector relative to $\Delta_{2'}$ electrons is labeled $Q_{\Delta_{2'}}$.

13 ML thick Cr layer [3]. If the Cr spacer thickness is decreased to 8 ML, still with a Cr capping layer, we obtain an antiferromagnetic coupling with very weak oscillations, the period being again close to 5.8 ML [see Fig. 3(b)].

We thus can sum up our experimental results as follows: the bilinear magnetic coupling constant is modified in Fe/Cr/Fe trilayers when capped with MgO. Strong oscillations appear with a 3 ML period as a function of Cr thickness and oscillations with a 5.8 ML period appear as a function of Fe thickness. However, the novel period of 3 ML Cr vanishes when the growth is performed at a slightly higher temperature or when the capping layer is Cr.

What is the origin of the novel period of 3 ML Cr? Since oscillation periods of the IEC as a function of the spacer layer thickness are determined by stationary spanning vectors Q of the Fermi surface of the spacer material [4–8], we have to look for a stationary spanning vector of the Cr Fermi surface along the [001] direction that yields a period of 3 ML. Indeed, such a stationary spanning vector can be identified in Cr (see Fig. 4). It links two points of the $\Delta_{2'}$ band and is associated with a period of 0.4 nm, i.e., 2.8 ML of Cr, as already calculated by Stiles for paramagnetic Cr [7]. No other stationary spanning vector is likely to explain the observed 3 ML oscillation period. Moreover, the calculations by Stiles (see Fig. 5 in Ref. [7]) confirm a nonvanishing reflection coefficient $r_{\Delta_{2'}}$ for the $\Delta_{2'}$ electrons at the Cr/Fe interface, allowing the formation of quantum well states in Cr. It thus clearly suggests that the 3 ML oscillation period is due to $\Delta_{2'}$ electrons in our samples. We note that a 3 ML period was also observed in Fe/Mo/Fe samples by Qiu *et al.* [19]. The analogy is striking since Mo and Cr have very similar electronic band structures [20].

Why does the 3 ML period appear in our experiments, whereas it has never been observed so far in the Fe/Cr/Fe system? The answer lies in the confinement of electrons in the thin top Fe film below the MgO capping layer. As shown in Fig. 3(a), the bilinear magnetic coupling constant strongly oscillates with the top Fe layer thickness. Such oscillations with a period of 5.8 ML were already observed by Okuno and Inomata[11] in Fe/Cr/Fe samples capped with Cr, but they were much weaker as in our MgO-capped samples. Thus, MgO capping dramatically increases the influence of the electron confinement in the Fe layer on the magnetic coupling. Consistent with what we observe as a function of Cr thickness, we believe that majority $\Delta_{2'}$ electrons in the Fe layer are involved, while minority $\Delta_{2'}$ electrons exhibit too weak a reflection coefficient to lead to a significant oscillation amplitude [7]. The corresponding stationary spanning vector $Q_{\rm maj}$ in Fe yields a period that is given by $T_{\rm Fe} = (1 - \frac{Q_{\rm maj}}{k_{ZB}})^{-1}$ ML with k_{ZB} the wave vector of the boundary of the Fe Brillouin zone along the ΓH direction. A look on the band dispersion curve for $\Delta_{2'}$ electrons [7] suggests values of this period between 5 and 8 ML. We note that a different explanation for the 5.8 ML period in Fe has been put forward by Okuno and Inomata [11]. They suggest that the involved stationary spanning vector connects Δ_1 and $\Delta_{2'}$ Fe majority bands at the Fermi level. However, this would be surprising as such a spanning vector connects bands of different symmetries.

The appearance of the novel period in IEC as a function of the Cr spacer layer thickness can finally be explained as follows: capping the thin top Fe layer with MgO leads to a reflection coefficient equal to 1 at the MgO/Fe interface and enables the formation of quantum well states in this Fe layer for $\Delta_{2'}$ electrons. This in turn modifies the density of states for these electrons at the Fermi level in Fe. As a consequence, the reflection coefficient $r_{\Delta_{2'}}$ of $\Delta_{2'}$ electrons in Cr at the top Cr/Fe interface is modified, therefore changing the confinement of $\Delta_{2'}$ electrons in Cr. The appearance of the 3 ML Cr period suggests that $r_{\Delta_{2'}}$ is increased in the studied systems. This was anticipated by Stiles who showed that the reflection coefficient at the Cr/Fe interface for states along the ΓH direction depends on the Fe thickness when Cr is paramagnetic [7]. In the case of Cr capping, the reflection coefficient is less than 1 at both Fe interfaces, which is less favorable to the formation of quantum well states in Fe and thus to a subsequent modification of $r_{\Delta_{2'}}$ at the Cr/Fe interfaces. Nevertheless, as shown by Okuno and Inomata [11], there is still an influence of the Fe layer thickness on the coupling, suggesting that the 3 ML period should even be present in the case of Cr capping, but with a much weaker intensity.

Finally, in order to confirm the role of $\Delta_{2'}$ electrons, we capped the Fe/Cr/Fe system with epitaxial bcc vanadium instead of MgO. Since no $\Delta_{2'}$ states are available at the Fermi level in V [21], it should play the same role as MgO. Indeed, we also observed that the V capped part of the sample exhibits an antiferromagnetic coupling (see Fig. 1) as in the case of a MgO-capped sample. This confirms that $\Delta_{2'}$ states are involved in the appearance of the novel oscillation period and that V plays the same role as an insulator regarding the magnetic coupling.

We now have to address the role played by the quality of the Fe/Cr interfaces which seems crucial to observe the 3 ML period. In the case of very smooth Fe/Cr layers grown on a Fe whisker at 300 °C, the coupling is fully dominated by the 2.1 ML oscillation period [15]. As pointed out by Heinrich *et al.* [14], there is then no noticeable influence of the Fe thickness on the magnetic coupling constant. On the contrary, rougher samples grown at room temperature, which do not exhibit the 2.1 ML period, show a weak but noticeable oscillation of the coupling with the Fe thickness [11]. If the roughness is too high, the 3 ML Cr period cannot be observed. Here we are apparently in an intermediate regime where the 2.1 ML oscillations do not dominate, but where the roughness is sufficiently low to enable us to observe the 3 ML period. This is consistent with the observation of both the long and short (2.1 ML) period in our Cr-capped samples (Fig. 2(b)]. The sensitivity to the growth temperature also shows the role played by the interfaces: increasing it above 150 °C prevents the appearance of the 3 ML period. This suggests that interdiffusion at the Fe/Cr interfaces suppresses the contribution of $\Delta_{2'}$ states to the coupling relative to the other periods [15], probably due to a higher sensitivity of $r_{\Delta_{2'}}$ to alloying. This is consistent with the observation that oscillations of J_1 as a function of the Fe thickness only appear in layers grown at low temperature [14]. The difference in roughness might also explain that the 3 ML period was not observed by Unguris and co-workers [13], although their Fe/Cr/Fe samples were uncapped. Vacuum should have played the role of a perfect barrier as MgO in our case, thus revealing the 3 ML period.

In conclusion, we observed the appearance of a novel period (3 ML) of the IEC as a function of Cr thickness in epitaxial Fe/Cr/Fe (001) sandwiches capped with MgO. This effect is explained by the creation of quantum well states for $\Delta_{2'}$ electrons in the Fe layer next to the MgO capping layer which in turn modifies the confinement of $\Delta_{2'}$ electrons in the Cr spacer layer. Other systems might show similar effects provided some conditions are fulfilled: at least one electronic band should be present at the Fermi level in both the spacer and the ferromagnetic layers, but not in the capping layer. As a consequence, the capping and the spacer layers should be made of two different materials. Moreover the reflection coefficient at the spacer-ferromagnetic interface for electrons of this band should be nonzero and be sensitive to the density of states in the ferromagnetic layer. Finally, one might speculate that this effect could be useful in tailoring the IEC by an appropriate choice of the nonmagnetic capping layer material.

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