Unambiguous Evidence of Oscillatory Magnetic Coupling between Co Layers in Ultrahigh Vacuum Grown Co/Au(111)/Co Trilayers

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The first direct evidence of oscillatory coupling in Co/Au(111)/Co trilayers grown in ultrahigh vacuum is reported. For samples with identical cobalt layer thicknesses, up to three peaks of magnetoresistance, corresponding to antiferromagnetic coupling between the cobalt layers, are observed with increasing gold spacer layer thickness, t_{Au}. By growing trilayers with different cobalt coercive fields, a direct measurement of both ferromagnetic and antiferromagnetic coupling as a function of t_{Au} was possible. The experimental data are in good agreement with theoretical predictions.

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The oscillatory coupling between ferromagnetic layers through a nonmagnetic (NM) metallic spacer layer, first observed on rare-earth [1] and transition metal [2] multilayers, has been shown to be a fairly general behavior [3].

There has recently been a theoretical treatment of the relation between the oscillatory behavior and the Fermi surface of the NM spacer [4]. More precisely, an oscillation period, Λ_i, exists corresponding to each wave vector, q_i, parallel to the reciprocal of the growth direction and connecting points on the Fermi surface with antiparallel Fermi velocities. The oscillatory behavior thus depends on the nature of the NM metal and on the growth direction of the multilayer. Quantitative prediction can be made for noble metal spacers: Cu, Ag, Au, based on experimentally determined Fermi surfaces of the bulk metals.

Predictions based on such a model have been unambiguously verified for the following NM spacers and orientations: Cu(100) and (110) [5–7], Ag(100) [8], and Au(100) [9–11]. However, a controversy still remains for the (111) orientation of each of the aforementioned NM spacers. Indeed, for this orientation clear magnetic coupling oscillations have only been observed for sputtered Co/Cu [12,13] and Ni/Ag [14] samples which are essentially (111) textures. Co/Cu(111) samples grown in ultrahigh vacuum (UHV) have exhibited contradictory results. While both clear oscillations of magnetoresistance (MR) [15] and oscillatory coupling evidence from neutron measurements [16] have been reported recently, other samples of high crystalline quality have shown no coupling [17,18]; for others, only the first MR peak [19–23] is observed or the oscillation periods are very different from the predicted ones [24]. Only indirect evidence of coupling oscillations has been reported recently for UHV grown Fe(110)/Ag(111) multilayers [25], and only the first antiferromagnetic coupling peak has been observed for Fe(110)/Au(111) multilayers [11]. To our knowledge, we report here the first unambiguous evidence of oscillatory coupling in UHV grown Co/Au(111)/Co samples.

The samples are Co/Au(111)/Co trilayers, grown at room temperature, in UHV ($<5\times10^{-10}$ hPa), on a 25 nm thick Au(111) buffer layer deposited on float glass platelets. Detailed studies of the growth and crystalline structure of identically grown samples have been published elsewhere [26]. After annealing at 175 °C for 1 h, the Au buffer layer is polycrystalline, 100% textured with perpendicular [111] orientation, the lateral size of the crystallites being about 200 nm. The surface is atomically flat and made of (111) terraces about 30 nm wide, separated by monatomic steps. As tested by cross transmission electron microscopy and scanning tunneling microscopy measurements, this flatness extends across grain boundaries. Samples have been grown in two different UHV units, the main difference being the Co evaporation method, either thermal evaporation from tungsten boat or *e*-beam evaporation. In both cases deposition rates were kept below 1 atomic layer (AL) per min.

Flat, abrupt interfaces are a necessary condition for the observation of magnetic coupling. The abruptness of the interfaces is ensured by the fact that Au and Co do not alloy. Under the evaporation conditions described above, Co does not exactly grow layer by layer. However, it has been shown recently [27] that the deposition of about 1 AL (=0.235 nm) of Au coverage smooths the surface. Moreover, at least for the Au spacer thicknesses investigated here, it has been verified by atomic force microscopy that there is no significant increase in roughness with further growth of the Au spacer. Furthermore this study was restricted to thin Co layers with perpendicular magnetization easy axis where monodomain configurations can be achieved [28]. The Au/Co system thus seems a good candidate for the study of magnetic coupling.

Finally, to insure maximum precision and reproducibility of the Au interlayer thickness variation, a moving shutter is used to grow stepped-wedge shaped Au spacer layers, with up to 10 different Au thicknesses on the same sample [15,17].

For the MR measurements, two such stepped-wedge

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samples have been grown, with Co layers of identical thickness (1.2 nm), and Au spacer layers with eight equally spaced thicknesses varying by steps of about 1 AL from 0.51 to 2.34 nm and from 1.87 to 3.62 nm for samples (1) and (2), respectively. Inside each terrace of uniform Au spacer thickness, well separated 1 mm wide stripes are then obtained by scribing the sample with a diamond-tip stylus along the step direction. The experiments were performed at room temperature with current in-plane and perpendicular applied magnetic field. The symmetrical configuration of the Co films was chosen to insure the maximum ratio between the values of the MR amplitudes for antiferromagnetic (AF) and ferromagnetic (F) coupling. When the two magnetic films are identical, a significant MR value is indeed only observed if they are antiferromagnetically coupled. Moreover, in that case, the AF coupling *should* manifest itself clearly by a splitting of the hysteresis loop, easily observed by magneto-optical (MO) measurements.

The MR curve versus the gold spacer layer thickness t_{Au} shows unambiguously, and for the first time in this system, three oscillations (Fig. 1). The maximum MR values are obtained for t_{Au} values of about 5, 9, and 14 AL (1.2, 2.1, and 3.3 nm), leading to a mean oscillation period of 4.5 AL, in good agreement with the theoretical prediction of 4.83 AL [4] for oscillating coupling through Au(111). The MR ratio $\Delta R/R=[R_{max}-R(H_s)]/R(H_s)$, where H_s is the saturation field, is around 2% at the maxima. At the minima, $\Delta R/R$ is smaller by about 2 orders of magnitude. Unexpectedly, the amplitude of the first peak at $t_{Au}=5$ AL is smaller than that of the two others.

The same stepped-wedge samples have then been investigated by polar magneto-optical effect. Faraday ellipticity hysteresis loops are shown in Fig. 2 for a few values of the Au spacer thickness. Relevant changes in the shape of the loops are observed versus t_{Au} [29]. For t_{Au} values corresponding to low MR, perfectly square hysteresis loops are obtained, whereas broadened ($t_{Au}=5$ AL) or even split ($t_{Au}=9$ or 14 AL) hysteresis loops are obtained for t_{Au} values around the MR maxima. This provides already strong evidence for an oscillating F-AF interlayer coupling in our samples. However, the symmetrical trilayer configuration, with two identical Co layers, does not allow quantitative determination even of AF coupling. Indeed, in these high quality perpendicularly magnetized Co layers, reversal of magnetization happens through easy propagation of domain walls, after very few domain nucleations at well separated centers: The coercive field corresponds to a nucleation field H_N, higher than the propagation field H_P [28]. In our symmetrical trilayers, where both Co layers have nearly identical coercivities, magnetization reversal in one layer might induce some unwanted nucleation in the other layer, for instance around areas with ferromagnetic coupling, expected to occur through spacer thickness fluctuations even for t_{Au} values where the average coupling is antiferromagnetic. As $H_P<H_N$, the two layers will then reverse their magnetization in parallel, which could explain the broadened loop of Fig. 2 ($t_{Au}=5$ AL) and the corresponding reduced MR value around the first maximum.

To directly measure the coupling between the Co layers, we devised an original technique, based on our ability to produce perpendicularly magnetized Co layers with square hysteresis loops and very different coercive fields [27,30]. The experiment was made in a different UHV

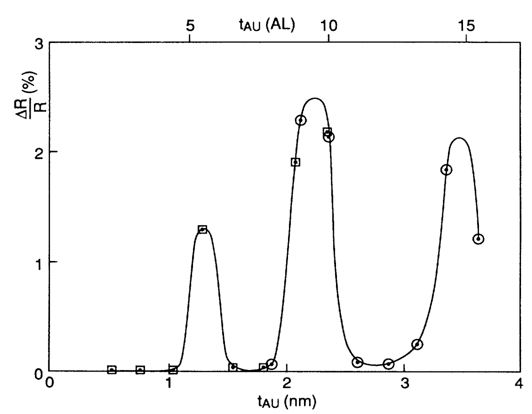

FIG. 1. Magnetoresistance versus gold spacer thickness t_{Au} at room temperature, for Au/Co/Au/Co/Au(111) samples with two Co layers of equal thickness 1.2 nm. The MR is defined by $[R_{max}-R(H_s)]/R(H_s)$ where H_s is the saturation field. The different symbols correspond to two different stepped-wedge samples, each one with eight different gold spacer thickness t_{Au}. The uncertainty on the absolute value of t_{Au} is $\pm 5\%$.

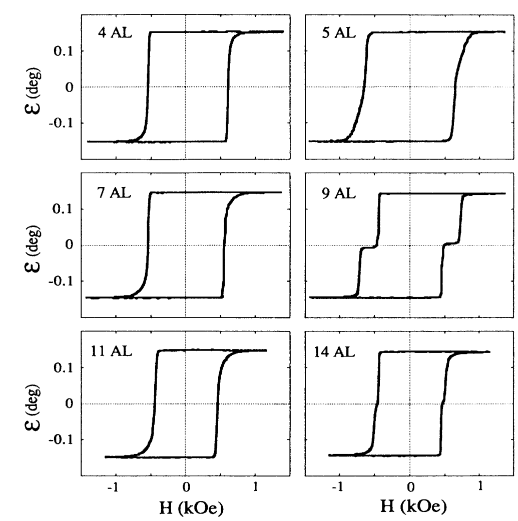

FIG. 2. Hysteresis loops measured by magneto-optical Faraday ellipticity on the samples used for magnetoresistance measurements. Note the different shapes of the hysteresis loops for Au thicknesses corresponding to minima (4, 7, and 11 AL) and to maxima of the magnetoresistance (5, 9, and 14 AL), attributed to, respectively, ferromagnetic and antiferromagnetic interlayer coupling.

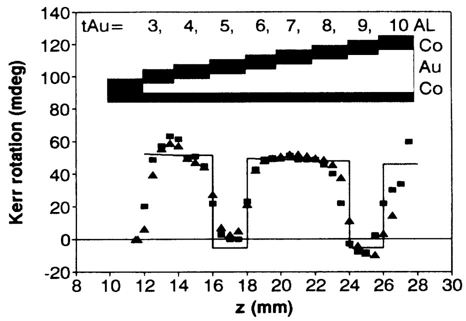

FIG. 3. Experimental recording in null field of the magneto-optical (MO) polar Kerr signal versus laser spot position, for the vacuum/Co/Au/Co/Au(111) schematized in the inset. Full line shows the MO simulation assuming perfect either parallel or antiparallel alignment of the perpendicularly magnetized Co layers, and infinitely small laser spot. The experimental zero reference is given by the signal on the 6.6 AL uncovered Co terrace, which exhibits in plane easy magnetization.

system than that used to grow the samples discussed above. In this new system *in situ* polar Kerr magnetic loops can be obtained in perpendicular magnetic fields up to 1.4 kOe [27].

On a Au buffer layer, a 3 AL Co film was grown and covered by a stepped-wedge shaped Au spacer layer with ten 2 mm wide terraces of $t_{Au}=0,3.27,4.36,5.44,6.54,7.63,8.71,9.80,10.89,11.98$ AL (cf. inset of Fig. 3). As shown previously [27], a 3 AL Co film, covered with 3 AL or more of Au, exhibits a high perpendicular anisotropy and displays a very square hysteresis loop with a coercive field larger than 800 Oe. This Co film is magnetized to saturation in a 1.4 kOe field, and a second Co layer of thickness 3.6 AL is evaporated on top. At this thickness, the second uncovered Co layer on Au is still perpendicularly magnetized, with square hysteresis loops, but has a low coercive field, around 100 Oe. The second Co layer has thus grown under the influence, through the Au spacer, of the magnetically saturated first Co layer. At terraces where the coupling through Au is ferromagnetic, parallel alignment of the second Co layer with the first is expected, and respectively antiparallel alignment for antiferromagnetic coupling. To test that, immediately after deposition the sample is swept in null field in front of the laser beam, while the MO signal is recorded versus sample position (Fig. 3). Two dips in the curve, for spacer thicknesses of 5.44 and 9.80 AL, are indeed observed, in excellent agreement with the MR results displayed in Fig. 1. As can be seen from Fig. 3, a full magneto-optical calculation using optical and MO indices previously determined on Au/Co/Au simple sandwiches, and assuming perfect either antiparallel or parallel configuration of the magnetic layers in the regions of respectively AF and F coupling, is in excellent agreement with the experimental data [31]. This gives thus direct evidence of AF coupling through Au(111). Moreover, it is to our knowledge the first observation of perfect antiparallel alignment in AF coupled systems through a

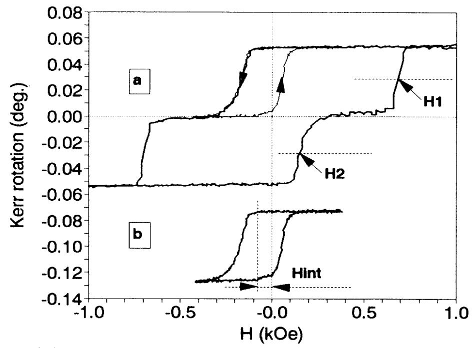

FIG. 4. (a) Full hysteresis loop and (b) minor hysteresis loop measured in UHV by magneto-optical Kerr rotation on the terrace with $t_{Au}\sim6.54$ AL of the trilayer schematized in Fig. 3. In (a), the jump at H_1 (H_2) corresponds to the magnetization reversal of the buried (uncovered) Co layer.

(111) oriented noble metal.

The technique we used to determine the coupling amplitude is illustrated in Fig. 4, with hysteresis loop measurements on a terrace with $t_{Au}=6.54$ AL (F coupling). The full hysteresis loop [Fig. 4(a)] displays as expected two jumps at fields H_1 and H_2, corresponding to magnetization reversal in respectively the first and second (uncovered) Co layers. If, starting from saturation in positive field, we decrease the field to a value between $-H_2$ and $-H_1$ to complete the magnetization reversal of only the second layer, and then increase the field again, we obtain a minor loop, displaced by a field H_{int} corresponding to the magnetic interaction between the two layers [Fig. 4(b)]. Naming the thickness of the second Co layer by t_2, the saturation magnetization in the layer by M_s and the interlayer coupling per unit surface by J, we have

$$H_{int}=-J/M_s t_2 . \quad (1)$$

Moreover, J can also be determined from H_2, with the equation

$$H_2=H_{c2}+J/M_s t_2 , \quad (2)$$

where H_{c2} is the intrinsic coercive field of the second Co layer, determined, for instance, from the minor loops ($H_{c2}=113\pm3$ Oe). Both determinations agree well within experimental precision. Note that in the second Co layer, where perpendicular anisotropy is weak [27], the magnetization reversal happens through nucleation only [28], and thus coupling fluctuations will have no other effect than slight broadening of the field range for magnetization reversal.

Figure 5 displays the resulting experimental variation of J versus t_2. Clearly an oscillating coupling is observed, with AF interaction for t_{Au} around 5 and 9 AL in agreement with results discussed above. The overall decrease rate, however, is much higher than the theoretically predicted $1/t_{Au}^2$ dependence [4]. Such a fast decrease, with an attenuation length t_c of about 4 AL, has actually been predicted for the Co/Au(111) system, on the basis of both the specific character of the corresponding wave vector and the expected presence of a high density of inter-

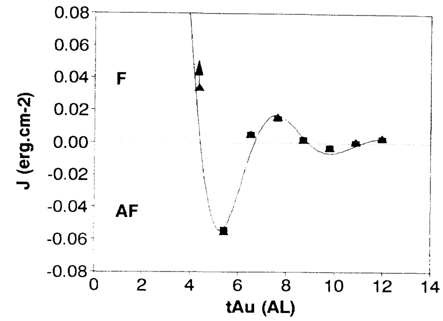

FIG. 5. Dependence of the exchange coupling J between Co layers versus the thickness t_{Au} of the Au(111) interlayer. J was determined in the trilayer schematized in Fig. 3: (square) from field shift in the minor hysteresis loops [cf. Fig. 4(b)], and (triangle) from the field H_2 in the full hysteresis loops [cf. Fig. 4(a)]. The arrow for $t_{\mathrm{Au}}=4.36$ AL means that, due to imperfect separation between magnetization reversals in the two layers, the value reported here must be considered as a lower estimate. Continuous line: theoretical fit of experimental data to Eq. (3) (RKKY model), with $I_0=33.8$ ergs cm^{-2}, $\Lambda=4.5$ AL, $\psi=0.11$ rad, $t_c=5$ AL, and $m^*/m=0.16$.

face dislocations, due to the large lattice mismatch between Co and Au. A plausible way of introducing such a decrease, at least at thicknesses of the order or above t_c, is to include a factor $\exp(-t_{\mathrm{Au}}/t_c)$. The experimental data have been compared with the function proposed by Bruno and Chappert [4]:

$$J=-(3I_0m^*/m)\sin[2\pi(t_{\mathrm{Au}}/\Lambda)+\psi]\exp(-t_{\mathrm{Au}}/t_c)/t_{\mathrm{Au}}^2\,. \quad (3)$$

As can be seen from Fig. 4, an excellent agreement is obtained by taking $I_0=11.3$ ergs cm^{-2}, $\Lambda=4.5$ AL, $\psi=0.11$ rad, and $t_c=5$ AL, using the predicted value $m^*/m=0.16$. As mentioned by Bruno and Chappert [4], simple Ruderman-Kittel-Kasuya-Yosida calculations cannot predict the amplitude and phase of the coupling, and the comparison between the experimental values and the predicted ones (about 13 ergs cm^{-2} and $\pi/2$ for I_0 and ψ, respectively) is not significant. On the contrary, for the period, Λ, and the attenuation length, t_c, the agreement with the predicted values, respectively 4.83 and 4 AL, can be considered very good.

In conclusion, the oscillatory interlayer couplings between Co films across Au(111) have been observed by different experimental techniques, on samples prepared in two different UHV units. Both the oscillation period and the dependence of the coupling strength on the spacer layer thickness are in good agreement with theoretical predictions based on a RKKY model.

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