Oscillations of Interlayer Exchange Coupling and Giant Magnetoresistance in (111) Oriented Permalloy/Au Multilayers

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The existence or not of oscillatory interlayer exchange coupling of ferromagnetic layers via (111) oriented copper spacer layers is controversial. We present evidence from magnetic and giant magnetoresistance studies of well-defined antiferromagnetic interlayer coupling in single crystalline (111) permalloy/Au multilayers. Four oscillations in the coupling are observed as the Au spacer layer thickness is increased. The oscillation period is $\simeq 10$ Å which is significantly shorter than the period of $\simeq 11.5$ Å predicted in Ruderman-Kittel-Kasuya-Yosida based models.

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Thin layers comprised of any of the ferromagnetic $3d$ transition metals are magnetically coupled via spacer layers of many nonferromagnetic transition metals [1]. The coupling oscillates between ferromagnetic and antiferromagnetic (AF) as the spacer layer thickness is varied. The noble metals, Cu, Ag, and Au, have relatively simple electronic band structures and associated Fermi surface topologies. Thus, these form excellent systems for testing predictions of theoretical models introduced to account for the oscillatory interlayer exchange coupling in metallic magnetic multilayers. These models include extensions of the Ruderman-Kittel-Kasuya-Yosida (RKKY) theory [2–4], spin-dependent quantum confinement of electrons in spin-dependent potential wells [5–7], and detailed first principles spin-polarized multilayer calculations [8]. Although the phenomenon of long-range oscillatory interlayer exchange coupling is now well established in polycrystalline Cu [9,10], as well as (110) or (100) oriented Cu [9,11–15], Ag [16], and Au [17–19], no clear-cut experimental evidence of long-range oscillatory coupling has been found in (111) oriented Co/Cu or Fe/Cu [20]. This had led to considerable controversy as to the existence or not of coupling in (111) oriented noble metal layers. In this Letter we present definitive evidence of oscillatory exchange coupling mediated by (111) oriented Au spacer layers in permalloy/Au multilayers.

The films were grown in a V. G. Semicon 80M molecular beam epitaxy (MBE) system equipped with low energy electron diffraction (LEED), reflection high energy electron diffraction (RHEED), and angle-resolved x-ray photoemission spectroscopy. The system base pressure was 4×10^{-11} mbar and during growth the pressure remained below $\simeq 2\times10^{-10}$ mbar. Three electron-beam sources were used to evaporate Pt, Fe, and $Ni_{1-x}Fe_x$ ($x\simeq0.15$). The permalloy (Py) layers were grown by codeposition of Fe and $Ni_{1-x}Fe_x$ ($x\simeq0.15$). By adjusting the evaporation rates from these sources the Ni-Fe composition could be varied. A temperature-stabilized effusion cell was used for Au. Typical growth rates were $\sim$0.05 to 0.4 Å/sec. The (111) crystalline was established using a thin Pt seed layer, $\sim$20 Å thick, which was deposited onto a (0001) sapphire substrate at 600 °C. The Pt seed layer grows epitaxially [21] on sapphire (0001) with Pt(111)‖Al_2O_3(0001) and Pt(110)‖Al_2O_3(10$\bar{1}$0). Two twin orientations of Pt, differing by a rotation of 180° about the [111] axis, are observed, leading to sharp sixfold LEED patterns. Twelve Py layers separated by eleven Au layers were subsequently grown, at 100 °C, via computerized shutter control. A final capping film of 20 Å Pt was deposited onto the topmost Py layer at $\simeq$70 °C. LEED and RHEED confirmed that the Py and Au layers grow epitaxially oriented with respect to the Pt seed layer. In order to examine the variation of exchange coupling and associated giant magnetoresistance with Au thickness in a consistent set of specimens, the source or substrate geometry was arranged to give a large and nearly linear variation in Au spacer thickness across the long ($\simeq$5 cm) sapphire substrates. We have previously exploited this feature to prepare Co/Cu(111) wedged multilayers [22]. The Py layer thickness was nominally constant ($\simeq$30 Å) for all epitaxial runs and the Ni:Fe composition along the length of a wedge varied by less than $\simeq$4%. After growth, the prescribed substrates were cleaved into samples, $\simeq 2\times11$ mm^2 in size, for magnetoresistance (MR), magnetic, and x-ray measurements. The resistance was measured using a standard 4-in-line contact geometry with gold plated pressure contacts and a low frequency ac lock-in technique.

The structure of the films was examined *in situ* with LEED and RHEED and *ex situ* with x-ray diffractometry. A typical high-angle x-ray specular (θ-2θ) scan along the (001) direction is shown in Fig. 1(a), recorded using a high-resolution 4-circle diffractometer. Intense, narrow features near $q_z=2.9$ and 5.8 Å^{-1} are due to the (0006) and (00012) reflections from the (001) sapphire substrate. Clusters of satellites from both (111) and (222) reflections from the multilayer are present, though these are well resolved only for the (111) region. No evidence of diffraction from other crystallographic orientations is seen. Fringes in the (111) region between the superlattice satellites may be thickness fringes originating from the Pt seed and cap films. The inset to Fig. 1(a)

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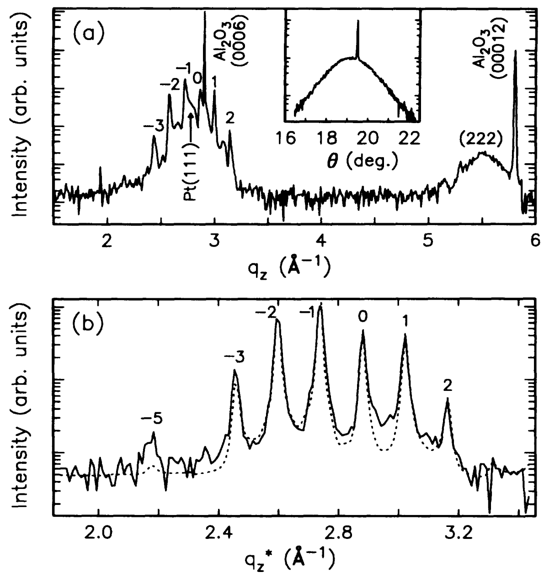

FIG. 1. X-ray data corresponding to a Py/Au sample from the second AF peak with a Au layer thickness of 20.2 Å. (a) $\theta/2\theta$ scan. The inset shows the rocking curve through the -1 (111) satellite peak; (b) $(\theta+0.15)/2\theta$ off-specular scan. The smooth curve corresponds to the measured data and the broken line is a fit to these data as described in the text.

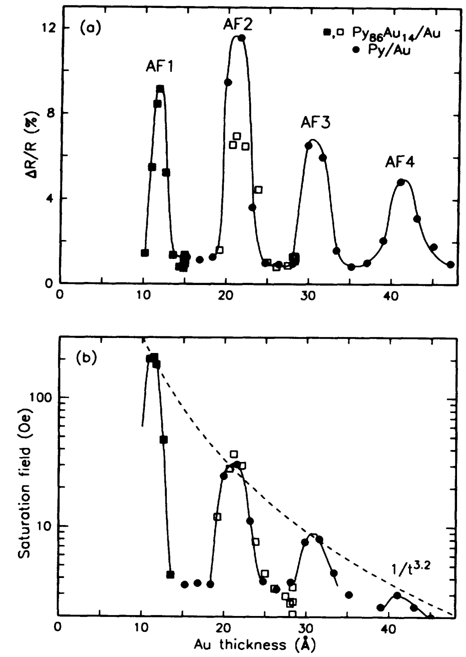

FIG. 2. Room temperature saturation magnetoresistance (a) and saturation field (b) versus Au spacer layer thickness at 295 K for three series of samples corresponding to three wedged superlattices. The open and filled boxes correspond to two wedges grown with $Py_{86}Au_{14}$ magnetic layers and the filled circles to a wedge prepared with Py layers. Four peaks in MR and saturation field are observed in the Au thickness range spanned, as indicated by AF1, AF2, AF3, and AF4.

shows a rocking curve across the (111) multilayer $n=-1$ satellite. The curve comprises a very narrow resolution-limited peak, 0.042° full width at half maximum (FWHM), and a broader peak, $\simeq 2.5°$ FWHM. The sharp peak is associated with the thin Pt seed layer and the broad peak is due to the multilayer. The width of the latter suggests that the Py/Au interfaces are semicoherent as a result of the large ($\sim 14.5\%$) misfit between the permalloy and Au layers. Figure 1(b) shows a slightly off-specular ($\Delta\theta=0.15°$) θ-2θ scan along the (111) direction which eliminates scattering from the substrate and Pt layers. These data were fitted using a multiparameter refinement procedure [23]. A good fit is obtained as shown in the figure. The Py and Au layer thicknesses could be precisely determined and were found to be in excellent agreement with thicknesses inferred from electron microprobe and x-ray fluorescence analyses. Consistent with the broad x-ray rocking curve, the permalloy and Au lattice parameters were found to be close to their respective bulk values. Good fits were obtained assuming the Au layers have constant lattice parameters throughout the layer but with some compression (expansion) of the Py out-of-plane (in-plane) lattice constants near the Py/Au interfaces. The best fits were obtained by introducing an intermixed region at the Py/Au interfaces of approximately 3 monolayers. Whether this corresponds to roughness or interdiffusion remains to be determined.

For certain ranges of Au layer thicknesses the Py/Au multilayers display enhanced magnetoresistance and saturation fields characteristic of antiferromagnetic coupling of the permalloy layers. The resistance of the structure is higher for antiparallel arrangement of neighboring magnetic layers compared to parallel alignment of these layers [24–27]. The magnitude of the saturation magnetoresistance versus Au layer thickness is plotted in Fig. 2(a) for three families of multilayers associated with three distinct wedges. Related saturation fields determined from the magnetoresistance curves are shown in Fig. 2(b). They correspond to the field at which the MR is half that of the saturation MR. Four well-defined oscillations in saturation magnetoresistance and saturation field are found as the Au layer thickness is increased, indicated by AFn, $n=1$-4 in Fig. 2(a). Typical resistance versus in-plane magnetic field curves are shown in Fig. 3 for samples corresponding to the four maxima in Figs. 2(a) and 2(b). Note that a small amount of Au was deliberately inserted in the permalloy layers in two of the wedges depicted in Fig. 2 by coevaporation of the Py and Au sources with fluxes in the ratio $\simeq 6$ to 1. This was an attempt to take advantage of the likely surface segregation of the Au from the Py layers during growth so as to reduce the possibility of magnetic pinholes between the Py layers. Similar samples grown without Au in the Py layers show suppression of the MR peak near $t_{Au}\simeq 11$ Å

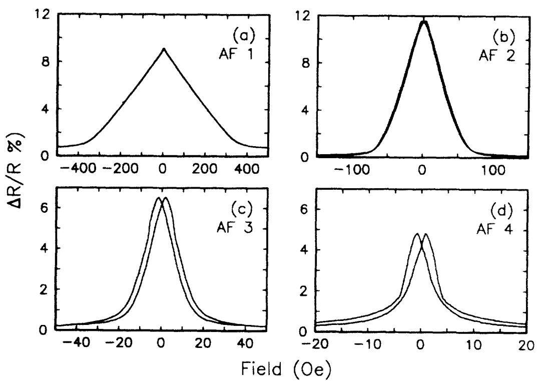

FIG. 3. Room temperature resistance versus in-plane magnetic field curves for four samples corresponding to the four maxima in MR shown in Fig. 2 with Au layer thicknesses of (a) AF1, 11.7 Å; (b) AF2, 21.5 Å; (c) AF3, 29.8 Å; and (d) AF4, 41.0 Å. The corresponding resistivities of these samples at room temperature are $\simeq 43$, 19, 13, and 10 $\mu\Omega$ cm, respectively, in fields large enough to saturate the magnetization of the samples.

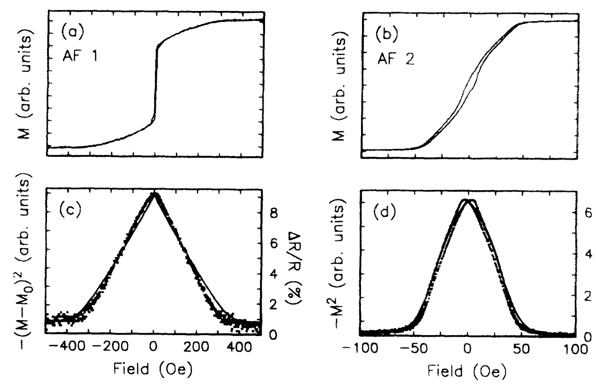

FIG. 4. Magnetization versus field curves, (a) and (b), for two Py-Au/Au multilayers from the AF1 and AF2 peaks shown in Fig. 2. In (c) and (d), corresponding resistance versus field curves (full lines) and plots of $-(M-M_0)^2$ versus field (filled circles) are shown. M_0 is the remanent magnetization.

(AF1). An analogous "surfactant" effect has been reported for copper surface segregation through cobalt during epitaxy of exchange-coupled Co/Cu(111) multilayers [22]. Note that alloying the permalloy with Au has little effect on the properties of the system for thicker Au layers except that the resistivity of the multilayers is increased. For example, at the second MR peak the resistivity is increased by about 30%, consistent with the decreased MR of these samples (see Fig. 2, open squares) relative to otherwise similar samples.

Typical magnetization versus in-plane magnetic field are shown in Figs. 4(a) and 4(b) for two Py/Au multilayers from the AF1 and AF2 peaks. The magnetization loops are consistent with the resistance curves shown in Fig. 3 and indicate antiferromagnetic coupling of the permalloy layers. The degree of antiferromagnetic coupling can be inferred from the remanent magnetization near zero field. Figure 4 demonstrates almost perfect antiferromagnetic alignment at the AF2 peak but incomplete ($\simeq 45\%$) AF coupling at the AF1 peak. The detailed relationship of magnetization and MR is explored in Figs. 4(c) and 4(d). Since permalloy is a soft magnetic material whose magnetization is saturated in small fields, the simplest model of giant MR can be used in which the resistance is proportional to the cosine of the angle between the magnetic moments of neighboring layers [28]. Then the resistance of the structure is expected to vary as $-M(H)^2$, where $M(H)$ is the magnetization parallel to the applied field, H. Figures 4(b) and 4(d) show a comparison of resistance and $-M^2$ versus field curves. Excellent agreement is obtained confirming the expected relationship. Note that for the AF1 sample the MR is compared with $-(M-M_0)^2$, where M_0 is the residual magnetization in small fields. M_0 represents the portion of the sample ($\simeq 55\%$) containing ferromagnetically coupled Py layers which does not contribute to the magnetoresistance. If there were complete AF coupling of the Py layers, these data suggest the magnitude of the MR at the AF1 peak would be much higher (MR$\simeq$20%) for multilayers containing Py-Au magnetic layers and yet higher still for pure Py layers.

The strength of the AF interlayer coupling, J_{AF}, is related to the magnetization, M, and thickness, t_F, of the permalloy layers and the field, H_S, needed to rotate the magnetic moments of the Py layers parallel to one another, by the relation $J_{AF} \simeq H_S M_S t_F/4$ [29]. Thus J_{AF} is $\simeq 0.02$ erg/cm^2 at the AF1 peak. This is approximately 5 times larger than found in sputtered (111) textured Py/Au multilayers [30] but is similar in magnitude to that found in sputtered Py/Cu multilayers [10] at room temperature. Assuming J_{AF} is proportional to M^2, the magnetic coupling through epitaxial (111) Au is about an order of magnitude smaller than the coupling found at AF1 in epitaxial (111) Cu [31]. Note that the strength of the coupling falls off as approximately $1/t_{Au}^n$, where $n = \simeq 3.2$, as shown in Fig. 2(b). This is faster than predicted in simple RKKY models ($n=2$) [32] although similar, for example, to that for single crystalline (110) Co/Cu [14].

Following the observation of oscillations in interlayer coupling and giant magnetoresistance in sputtered (111) textured Co/Cu multilayers [9,26] there has been considerable controversy as to whether this coupling is intrinsic to the (111) orientation or whether it arises from grains oriented along other directions [20]. Early studies on high quality MBE grown (111) Co/Cu found no evidence for coupling or enhanced MR [33]. Subsequent work on multilayers of nominally similar structural quality found evidence for AF coupling but only at the first AF peak and only partial AF coupling [22,31,34–37]. More recently, neutron reflectivity studies suggest the possible existence of oscillations in coupling for (111) Co/Cu, although only a tiny fraction ($\simeq 5\%$) of the films was found to be AF coupled [38] making the interpretation ambiguous. In contrast, the presence of oscillatory coupling in (110) and (100) oriented Cu was readily observed by many groups [11,12,20,34,35,39]. Note that even for sputtered Co/Cu the presence of AF coupling is very sensitive to the growth of the multilayers [26]. It was hypothesized that this was a result of structural defects such as "pinholes" through the Cu layers which could give rise to strong ferromagnetic coupling of the magnetic layers [9,26,37]. Since the interlayer magnetic coupling is weak compared to the intralayer exchange coupling within the ferromagnetic material, its observation is dependent on the growth of high quality structures. Recently, a detailed mechanism for the formation of pinholes in (111) oriented Co/Cu has been suggested [40]. The observation of oscillatory coupling in Py/Au suggests similar structural defects are less important in Py/Au.

In summary, we have observed clear-cut oscillations in interlayer coupling of permalloy layers mediated via (111) oriented Au spacer layers. Wedged (111) Py/Au multilayers exhibit oscillatory interlayer coupling with an oscillation period of $\simeq 10$ Å. The observed oscillation period is $\simeq 15\%$ shorter than that predicted within RKKY models. The strength of the coupling is about 10 times weaker than that via (111) oriented suggesting that the systematic trends in interlayer coupling strength found for coupling via transition metals [1] also apply to the noble metals.

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Note added.—Another group has independently found evidence for oscillatory coupling via (111) Au in (111) Co/Au sandwiches [41].

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