VOLUME 46, NUMBER 13

1 OCTOBER 1992-I

Oscillatory interlayer magnetic coupling of wedged Co/Cu/Co sandwiches grown on Cu(100) by molecular beam epitaxy

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Co/Cu/Co sandwiches were grown by molecular beam epitaxy onto a Cu(100) crystal with the intervening Cu layer fabricated to be wedge shaped. Electron diffraction was used to monitor the layer-bylayer growth. Films with 8, 14, and 20 monolayer- (ML) thick Co layers were investigated *in situ* by means of the magneto-optic Kerr effect. Characteristic hysteresis loops along the wedges, as a function of Cu thickness, unambiguously identify the oscillatory behavior in the sign of the exchange coupling between the two Co films. The oscillation periodicity (5.5 ML's of Cu) and, most importantly, the coupling strength are shown to be independent of the Co-layer thickness.

The investigation of the exchange coupling between magnetic films through nonmagnetic spacer layers has sparked great interest due to pioneering experiments on the Fe/Cr/Fe prototype.¹⁻⁴ The existence of antiferromagnetic (AF) interlayer coupling,¹ the negative magnetoresistance associated with such coupling,² and the oscillatory behavior between AF- and ferromagneticcoupling³ are now known to be characteristic of a whole new class of materials. $^{5-9}$ While most of the systems show oscillatory behavior with a periodicity of $\sim 10-15$ Å, short-period oscillations with periodicity of 2-3 monolayers (ML's) were observed for the Fe/Cr (Refs. 4, 10, and 11), Fe/Mn (Ref. 12), Fe/Au (Ref. 13), Fe/Mo (Ref. 14), and Co/Cu (Ref. 15) systems, which were fabricated by molecular beam epitaxy (MBE). In these latter cases the spacer layers were grown into a wedged shape to permit the systematic study of the interlayer coupling as a function of spacer-layer thickness by scanning a probe across a single sample. Theory suggests that the Ruderman-Kittel-Kasuva-Yosida (RKKY) interaction is responsible for the oscillatory behavior, ¹⁶ and that interfacial roughness can remove the short-period oscillations, while preserving the longer-period oscillations. Most theoretical models assume that the coupling occurs only at the interface between the magnetic and nonmagnetic layer and, therefore, constrain the thickness of the magnetic layer, for convenience, to be only 1 ML. The consequence of this assumption is that the interlayer coupling strength and the oscillation periodicity should be independent of the magnetic layer thickness. It is important to test this assumption experimentally, and that is the primary goal of this work. We report an investigation of the epitaxial Co/Cu/Co sandwich system grown onto Cu(100) by MBE with a wedge-shaped Cu spacer layer. MBE-grown samples of this system, but without wedged layers, have been studied previously.^{17,18} Miguel et al.¹⁷ made (100) sandwiches with the Cu layer between 2 and 10 ML's and reported one oscillation but could not obtain hysteresis loops for their AF-coupled samples. However, Heinrich et al.¹⁸ reported hysteresis loops for two AF-coupled samples: Co(4.3 ML's)/Cu(6

ML's)/Co(4 ML's) and Co(4 ML's)/Cu(10 ML's)/Cu(10 ML's). A more systematic study of this particular system is certainly worthwhile, especially because the growth mode has been well studied in the literature, $^{17-19}$ and Cu is a simple, noble metal that is frequently invoked by theorists as the spacer layers.¹⁶

The Co/Cu/Co sandwiches were grown by MBE onto a Cu(100) single-crystal substrate at room temperature in an ultrahigh vacuum (UHV) chamber of base pressure 2×10^{-10} Torr. The Cu(100) substrate is an ~3-mmthick disk with an \sim 10-mm diameter. The substrate surface was mechanically polished to an $\sim 0.1 - \mu m$ diamond-paste finish and ultrasonically cleaned in methanol before its introduction into the UHV chamber. Cycles of 3-keV Ar⁺ sputtering and annealing at 650 °C for ~ 30 min in UHV were used to clean the Cu(100) substrate surface and improve the surface quality. Auger electron spectroscopy confirmed the surface cleanliness. After this treatment a well-defined Cu(100) surface was formed as identified by reflecting high-energy electron diffraction (RHEED) and low-energy electron diffraction (LEED) [Fig. 1(a)]. The sharp streaks and Kikuchi lines in the RHEED pattern and sharp spots in the LEED pattern indicate that the substrate surface is flat on an atomic scale.

The Co and Cu layers were grown at room temperature by evaporating a Co wire and a Cu rod from alumina crucibles surrounded by W heating wire. The typical evaporation rate for both Co and Cu, as monitored using a quartz-crystal oscillator, was ~ 0.5 Å/min, and the pressure during growth remained $< 3 \times 10^{-10}$ Torr. The growth mode was studied by monitoring the RHEED intensity. The results obtained during growth for both Co on Cu and Cu on Co are shown in Fig. 2. The oscillations in the RHEED intensity demonstrate that both Co on Cu and Cu on Co follow predominantly a layer-bylayer epitaxial grow mode. We also found that the RHEED line spacing for Co on Cu(100) is the same within experimental uncertainty as that of Cu(100), which indicates that the fcc Co structure is formed on Cu(100) with the in-plane lattice spacing of Cu (3.61 Å).

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Although we did not obtain the interlayer spacing, Schneider *et al.*¹⁹ performed a LEED intensity analysis for the same system and reported a 1.74-Å spacing, which is $\sim 3\%$ smaller than the corresponding in-plane value. Thus, the resultant fcc structure of Co on Cu(100) is tetragonally compressed along the surface normal.

The Co/Cu/Co sandwiches were grown in a similar manner to that we used in growing Fe/Mo/Fe sandwiches.¹⁴ A Ta mask was placed above and in front of the specimen to block part of the evaporated beam. During the growth of the Cu spacer layer, the specimen was slowly translated behind the mask along the [011] crystal direction to form the Cu wedge. The slope of the wedge was controlled by the specimen translation speed and the Cu evaporation rate to be ~ 1 ML/mm. Three series of samples were made in this manner with Co thicknesses of 8, 14, and 20 ML's. Each series included several samples to cover the Cu thickness range of 2-20 ML's. The RHEED and LEED patterns for the three series of samples are shown in Figs. 1(b)-1(d). The LEED patterns show no noticeable differences from those of the Cu(100) substrate, and the Kikuchi lines in the RHEED

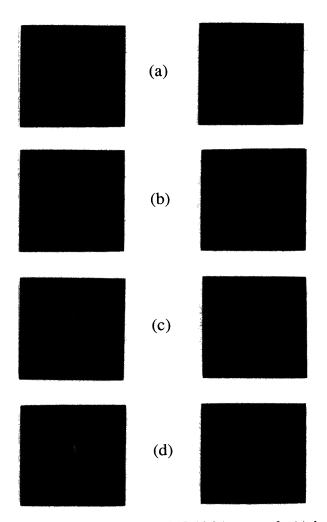


FIG. 1. RHEED (left) and LEED (right) patterns for (a) the Cu(100) substrate and for Co/Cu/Co sandwiches with (b) 8-ML-, (c) 14-ML-, and (d) 20-ML-thick Co layers.

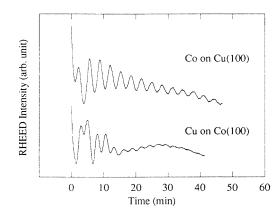


FIG. 2. RHEED oscillations for Co grown on Cu(100) and Cu grown on Co(100) at room temperature.

pattern persist, although they are somewhat weakened. Thus, we conclude that the sandwich structures are almost as flat as the Cu(100) substrate.

The magnetic coupling at room temperature between the two Co films across the Cu spacer layer was studied in situ by means of the surface magneto-optic Kerr-effect (SMOKE) technique. An external magnetic field H was applied in the film plane, in the plane of incidence of the light (longitudinal Kerr effect), and along the $[01\overline{1}]$ axis of the sample, which is perpendicular to the length of the wedge. It has been shown¹⁷ that the $[01\overline{1}]$ and [011] axis are the easy magnetization axes of Co on Cu(100). The incident beam ($\sim 17^{\circ}$ incident angle) emanated from a He-Ne laser, was focused onto the sample surface by an optical lens and possessed a beam size of ~ 0.15 mm. Schematic illustrations of the SMOKE apparatus appear in previous papers.^{14,20} Hysteresis loops are obtained for different thicknesses of Cu by scanning the laser along the length of the wedge.

Typical hysteresis loops of ferromagnetically and AFcoupled Co(20 ML's)/Cu/Co(20 ML's) are shown in Fig. 3. It should be mentioned that it is not possible to distinguish between ferromagnetic coupling and the noncoupling case from the shape of the hysteresis loops.²¹ Thus, we implicitly include zero coupling when we refer to ferromagnetic coupling. The hysteresis loop for AF coupling has a plateau in low fields and a rather abrupt switching to the saturation at high field, resulting in two hysteresis loops shifted from zero field by $+H_S$ and $-H_{\rm s}$. The plateau is due to the AF coupling, which aligns the magnetic moments of the two Co films in an antiparallel fashion to give a zero net magnetization. At high field, the AF coupling is overcome and the magnetic moments of the two Co films become aligned parallel to each other, which yields a net magnetization equal to its saturation value. The complete cancellation of the Kerr signal at the plateau indicates that the thicknesses of the two Co films are virtually identical. Another interesting feature in the AF-coupling hysteresis loop is that there are only two switching regions (at $+H_S$ and $-H_S$) separated by the plateaus with zero net magnetization. This result implies that there are only two stable states corresponding, respectively, to parallel and antiparallel

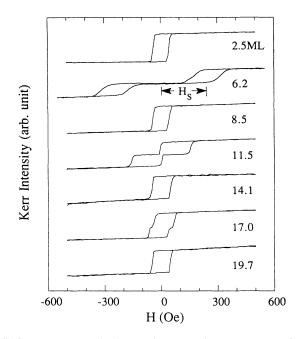


FIG. 3. Hysteresis loops for Co(20-ML)/Cu/Co(20-ML) sandwiches for different thicknesses of the Cu spacer layer. The Cu thickness values, denoted to the right, represent a linear vernier of the location of the laser probe along the wedge.

alignment of the magnetic moments in the two Co films. For the Fe/Au and Fe/Al systems¹³ multiple steps have been observed in the hysteresis loops associated with additional intermediate switching states in which the magnetization of the two layers are separated by 90°. The additional metastable states have been attributed to quadratic terms in the expression for the interlayer coupling en-

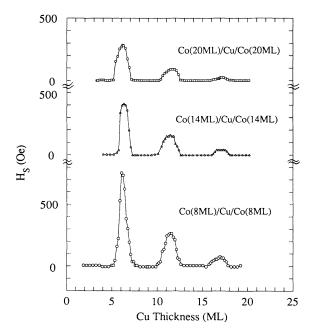


FIG. 4. The switching field, which is the offset of the loop from zero field, vs Cu thickness,

ergetics.¹³ The absence of additional structure in the loops we present indicates that 90° orientations of the two Co films and quadratic exchange coupling are negligible features in this system. Detailed discussions about the shape of the hysteresis loop can be found elsewhere.²²

We use the field H_S as an approximate measure of the AF-coupling strength. For ferromagnetic coupling $H_s = 0$. In Fig. 4, we display H_s as a function of Cu thickness for all three series of samples. Three oscillations in the exchange coupling appear in Fig. 4 with an oscillation periodicity of ~ 5.5 ML's (9.9 Å) of Cu, independent of Co thickness. The 5.5-ML periodicity agrees very well with one period predicted by Bruno's and Chappert's calculation. However, the other period $(\sim 2.6 \text{ ML's})$ predicted by the theory was not observed in our experiment but was observed recently by Johnson et al.¹⁵ We tried different conditions (e.g., changing the substrate temperature and/or homoepitaxially smoothing the substrate), but all films showed the same ~ 5.5 -ML periodicity. The absence of short-period oscillations may be attributed to interfacial roughness. It is interesting to note that the interface roughness we previously observed for the Fe/Mo system appears to be greater than that of this system, as indicated by RHEED and LEED studies,¹³ but the Fe/Mo/Fe sandwiches exhibited shortperiod oscillations. Therefore, the present results suggest that the observation of short-period oscillations in the Co/Cu/Co system will be more demanding to interfacial perfection than was the case for Fe/Mo/Fe.

To estimate the AF-coupling strength, we used the conventional assumption that the coupling only occurs at the interfaces. As discussed at the beginning of this paper, this implies that the coupling strength per unit area of interface, J_{AF} , is independent of the thickness d_{Co} of the magnetic layer. In this way, the coupling strength easily can be derived from the value of H_S . The idea is that at the switching field the magnetic energy of the anti-parallel-alignment configuration is the same as that

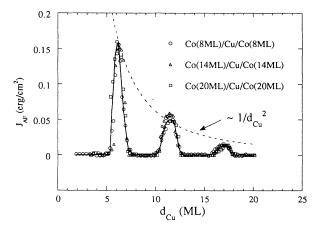


FIG. 5. J_{AF} ($\sim M_S H_S d_{Co}$) vs Cu spacer thickness for the three series of samples from the figure. The coincidence of the three curves indicates that the AF coupling occurs at the Co/Cu interfaces and is independent of d_{Co} . The dashed line represents $1/d_{Cu}^2$ relation in the coupling strength, which is predicted by RKKY interaction calculation.

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for parallel alignment of the two Co films. Then we define

$$J_{\rm AF} \approx M_S H_S d_{\rm Co}$$
,

where $M_S = 1.47 \times 10^3$ emu/cm³ is the saturation magnetization. We see from this equation that if J_{AF} is independent of Co thickness, H_S should be inversely proportional to d_{Co} . This trend is illustrated in Fig. 5 where we replotted the data from Fig. 4 as J_{AF} vs Cu spacerlayer thickness. Indeed, the three curves from Fig. 4 fall onto a single curve in Fig. 5. This result demonstrates a major conclusion of our work, that is that the coupling occurs at the Co/Cu interfaces and, thus, is independent

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of $d_{\rm Co}$. In addition, the $1/d_{\rm Cu}^2$ relation in the AFcoupling strength as predicted by RKKY interaction calculation, where $d_{\rm Cu}$ is the thickness of the intervening Cu layer, can be clearly seen in Fig. 5 (dashed lines).

In summary, we investigated Co/Cu/Co sandwiches grown by MBE on Cu(100) with wedge-shaped Cu spacer layers. We find that the interlayer exchange coupling oscillates between AF and ferromagnetic with a periodicity of ~ 5.5 ML's of Cu. This periodicity and the coupling strength are independent of the Co-layer thickness.

The work was supported by U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under Contract No. W-31-109-ENG-38.

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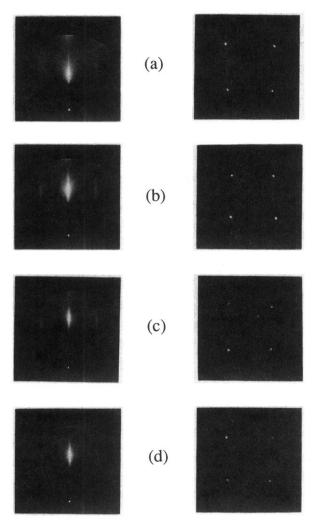


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