Orientational Dependence of the Oscillatory Exchange Interaction in Co/Cu/Co

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A strong oscillatory exchange coupling has been discovered in epitaxial (111) and (110) Co/Cu/Co sandwiches with wedge-shaped Cu layers, deposited by molecular-beam epitaxy (MBE) on single-crystal substrates. This finding proves the intrinsic nature of the strong antiferromagnetic coupling in (111)-textured sputtered Co/Cu multilayers. Combined with recent results on (100) Co/Cu/Co MBE-grown samples, it becomes evident that the nature of the oscillations in Co/Cu/Co depends markedly on growth direction, agreeing with predictions invoking the topology of the Fermi surface of the interlayer Cu.

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The discovery that the exchange coupling across nonmagnetic interlayers in a wide variety of magnetic multilayers oscillates as a function of the interlayer thickness [1-4] has presented a considerable challenge to solid state physicists. The initial long period of these oscillations (8-22 Å) was quickly ascribed to the *aliasing effect*, whereby rapid RKKY oscillations are only sampled at discrete thicknesses of the interlayer [5]. This argument subsequently led to a prediction of orientation-dependent oscillation periods in the coupling, even in the case of a nearly-free-electron metallic interlayer [6]. Further refinement by Bruno and Chappert [7] emphasized the role of the Fermi surface, extremal points of which, connected by vectors $\mathbf{q}$ parallel to the growth direction, generate oscillation periods equal to $2\pi/|\mathbf{q}|$. In this way, a multiplicity of oscillation periods may be encountered, the number and values of which are predicted to depend upon the growth direction of the samples. Strong support of this simple Fermi surface picture has been supplied by the latest series of coupling investigations on Co/Cu(100) [8], Fe/Au(100) [9], and Fe/Ag(100) [10] samples, where a superposition of oscillations with periods close to those predicted in [7] was established.

Oscillatory exchange coupling across Cu interlayers has attracted much attention from both fundamental and application viewpoints. On the one hand, the relatively simple nature of the Cu Fermi surface makes this a suitable candidate for the verification of exchange coupling theories; on the other hand, the discovery of sizable antiferromagnetic coupling and giant magnetoresistance (MR) effects in sputtered (111)-textured Co/Cu [1,2] and Fe/Cu [11] multilayers is of great interest to theorists and applications engineers alike. In an attempt to enhance these effects, Egelhoff and Kief [12] have used molecular-beam epitaxy (MBE) to grow a number of Co/Cu(111) and Fe/Cu(111) samples on single-crystalline Cu(111) substrates. However, in contrast to the situation in the sputtered samples, these supposedly high-quality MBE-prepared systems not only failed to give an enhanced antiferromagnetic (AF) coupling, but actually displayed no AF coupling at all.

In an attempt to explain this anomaly, it has recently been proposed [12] that the AF coupling in (111)-textured sputtered systems is attributable to a minority presence in the samples of (100) crystallites—to which a certain credence is lent by the samples' relatively broad rocking curves. However, in light of recent results on epitaxial MBE-grown (100) Co/Cu/Co [8], this proposal becomes fraught with problems. First, no detectable fraction of (100)-oriented crystallites has been observed by either x-ray diffraction or electron diffraction in strongly AF-coupled (111)-textured sputtered samples [13]. Second, the coupling oscillations in the two systems show different periods and phases. Finally, the AF coupling strength in the MBE-grown (100) system is lower than that observed in some sputtered (111) Co/Cu/Co samples, whereas a *minority* component is expected to possess a proportionally *greater* coupling strength to compensate for its reduced abundance.

In this Letter, we report the experimental discovery of strong oscillatory AF coupling in MBE-grown (111) Co/Cu/Co *and* (110) Co/Cu/Co samples with wedge-shaped Cu interlayers, both deposited on single-crystalline Cu substrates. In each sample, the coupling oscillates with a similar period and phase to those in the (111)-textured sputtered samples. The maximum coupling strength for the two systems is significantly greater than in MBE-grown (100) Co/Cu/Co [8]. Despite meticulous substrate preparation and growth, the observation of intermixed ferromagnetic (F) and AF coupling in the (111) MBE-grown samples suggests significant pinhole formation through, or local thinning of, the Cu interlayer. Since such effects are less likely to occur in sputtered layers (because of the smoothing effect of ion bombardment), this observation resolves the "MBE versus sputtered" anomaly associated with (111) Co/Cu/Co.

The overlayers were deposited on single-crystalline Cu substrates in a multichamber molecular-beam-epitaxy system (VG Semicon V80M). The completed samples were composed as follows: Cu(111)/Co(40 Å)/Cu wedge (0-44 Å; 3.0 ML/mm)/Co(40 Å)/Cu(7 Å)/Au(20 Å); Cu(111)/Co(40 Å)/Cu wedge (0-35 Å; 1.6 ML/mm)/

Co(40 Å)/Cu(7 Å)/Au(20 Å). The Cu wedge shape was formed by slowly withdrawing an eclipsing shutter located between the single crystals and Cu source. The substrate temperature was 50 °C during both Cu wedge depositions, and 20 °C for all other depositions. The Cu thickness was determined using a quartz crystal monitor calibrated both from comparison with chemically analyzed reference samples and from reflection high-energy electron diffraction (RHEED) intensity oscillations observed during earlier deposition of Cu on Cu(100). Thicknesses thus determined were subsequently confirmed after deposition using combined *in situ* Auger electron spectroscopy (AES) and scanning electron microscopy (SEM). The other film thicknesses were also determined using AES. More precise details concerning the substrate preparation and growth technique can be found in [8].

The structure of the samples showed similar characteristics to those of the (100) Co/Cu/Co systems investigated earlier [8,14]. The perpendicular and parallel lattice spacings were determined by measuring the energies of the primary Bragg low-energy electron diffraction (LEED) reflections along the [00] rod, and by comparison of LEED patterns at constant electron energy, respectively. Across the entire sample, the Cu wedges displayed identical lattice constants to those of the Cu single-crystalline substrates, and maintained an fcc structure. The Co, which was deposited at 20 °C to avoid interdiffusion [14], displayed a nearly identical fcc surface net to that of the Cu. For the (110) sample, a certain amount of streaking occurred in the LEED patterns upon deposition of Co on the Cu substrate, being particularly obvious in the [100] directions, where the surface is most open. A further evaporation of Cu reduced the width of these streaks considerably, with subsequent Co evaporation resulting in the reappearance of streaks and slight degradation of the pattern. Similarly, although the LEED pattern of Co on Cu(111) was seen to be slightly diffuse (though no streaks were observed), the diffraction spots sharpened up considerably upon subsequent Cu deposition. The perpendicular Co-Co spacing was observed to be 1.23 Å for the (110) sample and 2.02 Å in the (111) case—slightly less than the respective Cu-Cu spacings of 1.27 and 2.09 Å.

The AF coupling strengths were determined from analysis of loops measured at room temperature along the wedges (i.e., as a function of Cu thickness) via the longitudinal magneto-optical Kerr effect (MOKE). The external magnetic field was applied along the [111] (easy) axis for the (110) sample, and in an arbitrary direction for the (111) system. Figures 1(a)–1(c) and 1(d)–1(f) depict sets of loops for the (110) and (111) samples, respectively. A clear transition between AF and F (or weak AF) coupling is revealed as the Cu thickness changes. For AF-coupled loops [Figs. 1(a) and 1(e), for example], the Kerr signal rises from a minimum at low

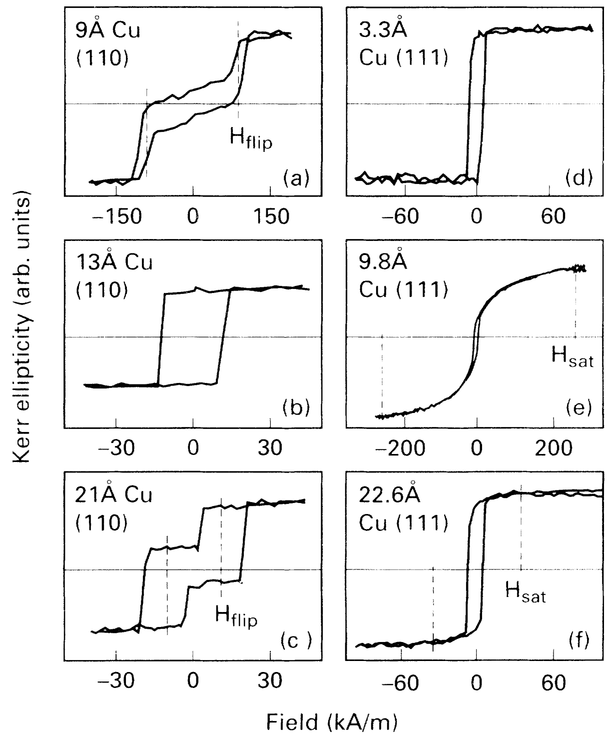

FIG. 1. MOKE hysteresis loops for MBE-grown (110) and (111) Co/Cu/Co. The Cu thickness is stated in each individual case.

fields (intrinsic antiparallel magnetic alignment of the Co layers) to a saturation value at higher fields (forced parallel magnetic alignment of the Co layers). The exact form of the loops is determined by the in-plane (magnetocrystalline) anisotropy of the Co layers. In the (110) case, this is rather large: A MOKE anisotropy study performed on the sample at a constant Cu thickness of 15 Å and for a number of azimuthal angles revealed an associated maximum anisotropy field $H_K = 200$ kA/m. As a result, magnetization reversal begins with a gentle rotation at low fields and is terminated by a relatively sharp switch at the flip field H_{flip} [Fig. 1(a)]. In contrast, the in-plane anisotropy in the (111) sample is minimal ($H_K \approx 0$), and the magnetization switch in that system is accordingly much more gradual: Uniform rotation of spins begins at low fields and continues until complete reversal is achieved at the saturation field H_{sat} [Fig. 1(e)]. The AF coupling strengths in the two different cases then follow (approximately) from the relations $J_{110} \approx -dM_s H_{\text{flip}}$ (for $H_{\text{flip}} \ll H_K$) and $J_{111} \approx -\frac{1}{2} dM_s H_{\text{sat}}$ (for $H_{\text{sat}} \gg H_K$), where d and M_s are the relevant magnetic layer thicknesses and saturation magnetizations, respectively [15].

In Fig. 2, we plot H_{flip} as a function of Cu thickness for the (110) sample. The graph depicts three peaks in the AF coupling: a primary peak at 8.5 Å (6.5 ML) Cu, followed by two weaker peaks (with faint indications of a

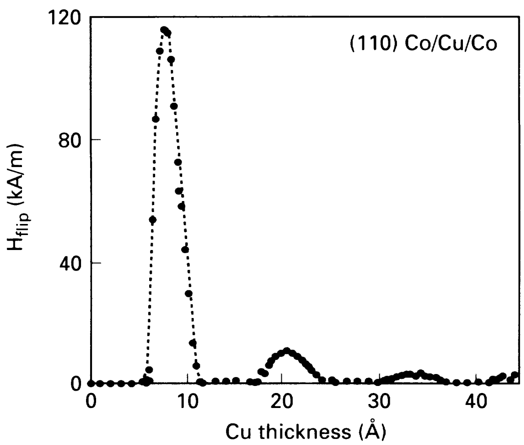

FIG. 2. The flip-field H_{flip} defined in Fig. 1(a) for the (110) sample, plotted as a function of Cu thickness.

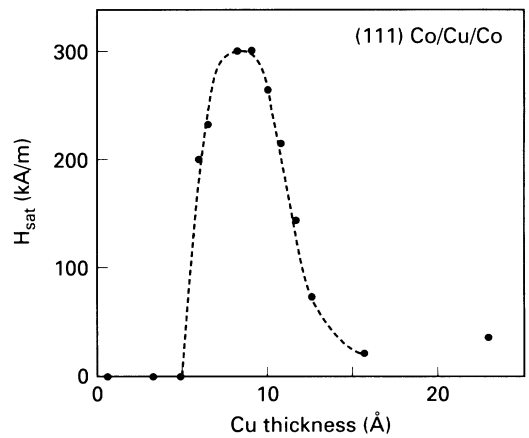

FIG. 3. The saturation field H_{sat} defined in Fig. 1(e) for the (111) sample, plotted as a function of Cu thickness.

third) at intervals of 12.5 Å. The maximum flip field (at 8.5 Å Cu) yields an AF coupling strength $J_{110} \approx -0.7$ mJ/m^2 [16]. Shorter periods of 2.7, 3.2, and 4.2 Å are also predicted to appear in the (110) system [7], but these were not found in the first sample. A second sandwich, with a shallower Cu wedge, was therefore prepared and carefully scanned, but this too failed to reveal any evidence of additional shorter-period oscillations. Extremely high interface quality would be required to detect such periods.

An equivalent graph for the (111) sandwich is rendered in Fig. 3, where the saturation field H_{sat} is plotted as a function of Cu thickness. A strong AF peak is again seen at 8.5 Å Cu, with faint suggestions of a second (much weaker) peak at about 20 Å [see Fig. 1(f)]. This would suggest a similar oscillatory period to that in the (110) system (11–12 Å). The saturation field in the primary peak corresponds to an AF coupling strength $J_{111} \approx -1.1$ mJ/m^2, which is even higher than the (110) value. It is therefore clear that strong oscillatory AF coupling can exist in MBE-grown (111) Co/Cu/Co samples.

Detailed analysis of the form of the AF loops in Figs. 1(a) and 1(e) reveals a nonzero Kerr signal at low fields and a steplike switch in the signal at the origin. These effects are believed to be caused by the presence of intermixed regions of F coupling within the predominantly AF-coupled region illuminated by the probing Kerr light beam (width ~100 μm). Such intermixing is possibly caused by the high mobility of Cu on Co during MBE growth, and the tendency of Cu to migrate on top of Co to reduce surface tension [17]. This can lead to local thinning of the Cu film (and possibly pinholes). The effect is more evident in the (111) sample than in the (110) system, and was not observed in the (100) samples [8], despite the equal care with which all systems were prepared and grown; a second (confirmatory) set of samples of identical constitution showed precisely the same phenomenon. The extent of the intermixing, even in (111) and (110) samples of this excellence, suggests that AF coupling will be completely masked in less perfectly grown (epitaxial) systems. This offers a convincing clarification of the absence of AF coupling from epitaxial samples investigated earlier [12].

The compilation of experimental coupling data for the

TABLE I. Measured coupling data for the Co/Cu/Co system, comprising the position (t_1) and FWHM (Δt_1) of the first AF peak, the maximum AF coupling strength (J), and the long (Λ_1) and short (Λ_2) oscillatory periods. Also included are predicted periods derived from [7], where de Haas-van Alphen (dHvA) data are employed, and from the same theoretical approach applied to *ab initio* ASW calculations. For the (110) system, three short periods are predicted (see text).

Sample	Ref.	t_1 (Å)	Δt_1 (Å)	$-J$ (mJ/m^2)	Λ_1 (Λ_2) (Å) Measured	Predicted (dHvA)	Predicted (ASW)
(111) MBE		8.5	5.5	1.1	11–12	9.4	9.0
(111) sputtered	[1,2]	8–9	4	0.3, 0.5	11–12		
(100) MBE	[8]	12	2.5	0.4	14 (4.6)	10.6 (4.6)	11.6 (4.6)
(100) sputtered	[13]	10–11	3	0.3	13		
(110) MBE		8.5	2.9	0.7	12.5	12.2 (3×)	11.2 (3×)

Co/Cu/Co systems in Table I clearly reveals the orientational dependence of the (longest) oscillatory period (Λ_1), the position (t_1) and FWHM (Δt_1) of the primary AF peak, and the maximum AF coupling strength (J). It is seen from the table that both J_{111} and J_{110} are significantly higher than the maximum coupling strength in the sputtered (111) systems and the MBE-grown and sputtered (100) systems studied earlier [8,13]. However, corresponding MBE-grown and sputtered systems display closely agreeing values of t_1, Δt_1, and Λ_1. The table also gives the theoretical periods, both as predicted in [7] from de Haas–van Alphen measurements of the Fermi surface and as rendered by the same theory applied to self-consistent *ab initio* augmented spherical wave (ASW) calculations of the Fermi surface. The measured periods for the [100] and [111] directions are somewhat greater than those predicted in [7], and this is also the case for the [110] direction if comparison is made with the ASW results. This finding may stimulate further investigations, in particular regarding the question of whether asymptotic behavior (i.e., period and phase independent of the interlayer thickness) has already been effectively reached for Cu thicknesses near the first AF peak.

In conclusion, we have shown that the oscillatory exchange interaction between MBE-grown Co layers separated by Cu interlayers is strongly orientation dependent, in quantitative agreement with the Fermi surface topology picture of Bruno and Chappert [7]. The discovery of intrinsically strong oscillatory coupling in the MBE-grown (111) Co/Cu/Co system resolves an anomaly regarding the coupling in (111)-textured sputtered Co/Cu multilayers.

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