Structural Dependence of the Oscillatory Exchange Interaction across Cu Layers

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A structure-dependent exchange coupling across wedge-shaped Cu layers in fcc (100) Co/Cu/Co and bcc (100) Fe/Cu/Fe epitaxial sandwiches has been determined from analysis of longitudinal magneto-optical Kerr effect hysteresis loops. In the fcc Co/Cu/Co system, the variation of the coupling with Cu thickness can be described as a superposition of two oscillatory terms, with periods of 2.6 and 8.0 monolayers (ML). The bcc Fe/Cu/Fe system shows oscillations with a period of 2 ML. These periodicities are related to the Fermi surface dimensions and topology of fcc and hypothetical bcc bulk Cu.

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Following the discovery by Parkin, Bhadra, and Roche [1] that in Fe/Cr, Co/Cr, and Co/Ru multilayer systems the exchange coupling of the ferromagnetic (F) transition-metal layers across the nonferromagnetic interlayers oscillates with the interlayer thickness, many more systems have been found to exhibit this phenomenon (see [2–4], for example). Initially, the periods of oscillation were found to be rather large (8–20 Å), but more recent work on molecular-beam-epitaxy- (MBE-) grown Fe/Cr/Fe [5], Fe/Mn/Fe [6], and Fe/Au/Fe [7] sandwich structures with extremely sharp interfaces has revealed additional oscillations with a period of 2 monolayers (ML). The great detail of these measurements on nearly perfect superlattice systems permits a close comparison with theoretical calculations, based on idealized structures.

One of the important issues of current interest is to what extent the period of the oscillations reflects the Fermi surface dimensions and topology of the bulk interlayer material. From the work of Wang, Levy, and Fry [8], it follows that, in general, the variation $J(t)$ of the exchange coupling parameter J with the interlayer thickness t depends on the whole band structure of the interlayer material, and not just on the Fermi surface. However, from an RKKY-like model, Bruno and Chappert [9] have argued that, in the case of a nearly free electron interlayer material, $J(t)$ will be a superposition of oscillating terms with periods which are determined by the appropriate extremal dimensions of the Fermi surface. Edwards *et al.* have arrived at a similar conclusion for the case of a single-band tight-binding structure [10]. The periods are equal to $2\pi/|\mathbf{q}_i|$, where the vectors $\mathbf{q}_i$ connect extremal points on the Fermi surface and are parallel to the growth direction of the layered system [9]. Because of the discreteness of the interlayer thickness t, only vectors $\mathbf{q}_i$ for which $|\mathbf{q}_i| \leq \pi/d$ are relevant ("aliasing effect" [11]). Here, d is the distance between lattice planes in the interlayer material. Therefore, the periods are always at least 2 ML, and are expected to depend on the crystallographic structure and growth direction of the interlayer, even in the case of spherical Fermi surfaces.

Of the noble metals Cu, Ag, and Au which are the most suitable candidates for an experimental verification of these theories, Cu has attracted the most attention. The reasons for this are the observation of antiferromagnetic (AF) coupling in MBE-grown Co/Cu multilayers [12] and sputtered Co/Cu [2] and Fe/Cu [13] multilayers and, in particular, the discovery of giant magnetoresistance (MR) effects in the sputtered samples. The MR displayed the same 12–15-Å long-period oscillation in both cases. These sputtered systems, probably with relatively rough or diffuse interfaces, did not show evidence of short-period oscillations. Short periods of the order of 2–3 ML have been predicted theoretically for fcc (100)-grown systems by Bruno and Chappert [9] and, from first-principles band-structure theory, by Herman, Sticht, and van Schilfgaarde [14]. However, even in epitaxial layers prepared on single-crystal substrates, the appearance of such shorter-period oscillations has so far avoided detection [12].

In this Letter, we report the experimental discovery of short-period oscillations, with different periods, in the exchange coupling across Cu interlayers in coherent epitaxial Co/Cu/Co and Fe/Cu/Fe (001) sandwiches. In both cases, the Cu interlayer was grown in the form of a wedge, but a crucial distinction was its structure, which was fcc in the case of the Co/Cu/Co sandwich but bcc in the Fe/Cu/Fe sample. The short-period oscillations in the Co/Cu/Co system were found to be superimposed on a longer-period oscillation. We discuss to what extent the measured oscillatory exchange coupling can be qualitatively and quantitatively related to the details of the Fermi surface of fcc and hypothetical bcc bulk Cu.

The overlayers were deposited on either single crystalline Cu(001) or Fe(001) whisker substrates in a multichamber molecular-beam epitaxy system (VG Semicon V80M). Before deposition, the Cu(001) crystal was first chemomechanically polished for 8–12 h in an aqueous mixture of H_2O_2 and Syton, and then subjected to several cycles of 1-h sputtering and 1-h annealing at 700 °C; the Fe whisker was prepared as described in [5] (Purcell *et al.*). The completed samples were composed as follows: Fe(001)/Cu wedge (0–40 Å; 1.83 ML/mm)/Fe(80 Å)/Au(20 Å); Cu(001)/Co(60 Å)/Cu wedge (0–40 Å; 2.5 ML/mm)/Co(60 Å)/Cu(7 Å)/Au(20 Å). The substrate temperature was 50 °C during both Cu depositions,

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and 20 °C for all other depositions. In the case of Co/Cu/Co, the Cu thickness was determined using a quartz crystal calibrated from Cu on Cu(001) reflection high-energy electron diffraction (RHEED) oscillations; for Fe/Cu/Fe, the Cu thickness was derived directly from oscillations in the RHEED intensity during monolayer-by-monolayer growth on the Fe(001) substrate. More precise details concerning the growth technique can be found in [5] (Purcell *et al.*).

The structure of the Co/Cu/Co sample is consistent with the observations of de Miguel *et al.* [15]. The perpendicular and parallel lattice spacings have been determined kinematically by measuring the energies of the primary Bragg LEED reflections along the [00] rod and by comparison of LEED patterns at constant electron energy, respectively. Across the entire sample, the Cu wedge displays identical lattice constants to those of the Cu(001) substrate, and maintains an fcc structure. The Co, which was deposited at 20 °C to avoid interdiffusion [15], displays an identical surface net to the Cu, and also grows with an fcc structure. The perpendicular Co-Co spacing was observed to reduce from 1.78 to 1.70 Å.

Our observations both during and after the growth of the Cu wedge on the Fe substrate are in agreement with the RHEED measurements of Heinrich *et al.* [16]. The presence of RHEED intensity oscillations during our growth indicates that the Cu grows in a layer-by-layer mode, and RHEED patterns suggest that the Cu initially grows in a bcc structure with an in-plane lattice very similar to that of the Fe substrate. The resolution of our RHEED system precludes confirmation of the 1% Cu in-plane relaxation reported in [16]. Since RHEED is generally insensitive to the vertical lattice spacing, it was unable to resolve the question of possible tetragonality in this case. We therefore extended our investigation to include LEED analysis of both the in-plane and perpendicular lattice. These measurements confirmed the RHEED observation of an almost unaltered surface net and, furthermore, revealed that the perpendicular lattice constant remains identical to that of the Fe(001) substrate: Thinner Cu layers (< 10 ML) are clearly of bcc symmetry. Heinrich [16] has observed that thicker Cu layers not only showed somewhat more diffuse RHEED patterns, but also additional weaker RHEED streaks, suggesting a Cu superlattice formation. The same observation was made in this present work. However, a simultaneous LEED study revealed that, where the RHEED superstructure becomes distinct (10–20 ML), the LEED patterns show no obvious change, remaining indicative of bcc Cu but becoming slightly weaker as the Cu layer increases in thickness. These findings suggest that the extreme surface sensitivity of the RHEED may be probing a weaker surface reconstruction of an otherwise unaltered bcc Cu(001) film, as seen by the less-surface-sensitive LEED. The Fe overlayer also grew in the bcc structure.

The AF coupling strengths were determined from analysis of hysteresis loops measured as a function of Cu thickness via the longitudinal magneto-optical Kerr effect (MOKE). In Figs. 1(a),1(b), and 1(c),1(d) we show two pairs of such loops for the Co/Cu/Co and Fe/Cu/Fe samples, respectively. Two distinct loop shapes are observed: Figs. 1(a),1(c) show a single transition in the signal around zero field (F or weak AF coupling), while Figs. 1(b),1(d) show transitions at higher fields (corresponding to a magnetization switch of the AF-coupled magnetic layers from antiparallel to parallel orientation, as applied field increases). A detailed interpretation of the loop shapes can be found in [17]. For the Co/Cu/Co sample (equal magnetic layer thickness), $J \cong -dM_s H_{\text{flip}}$, where d and M_s are the magnetic overlayer thickness and magnetization, respectively, and the switching field H_{flip} is defined in Fig. 1(b); in the case of Fe/Cu/Fe (unequal magnetic layer thickness), $J \cong -dM_s(H_1+H_2)/2$, where the critical fields H_1 and H_2 are depicted in Fig. 1(d).

In Fig. 2, we plot H_{flip} as a function of Cu thickness in the Co/Cu/Co sample. The graph depicts five peaks in the AF coupling: a strong AF peak at 6.6 ML (12 Å) surrounded by two F (or weak AF) regions, followed by a series of four strong AF peaks regularly spaced at intervals of 2.6 ML. The maximum coupling strength (at ~6.6 ML Cu) corresponds to $J \cong 0.4$ mJ/m^2.

Figure 3 depicts H_1 and H_2 as a function of Cu thickness in Fe/Cu/Fe. In this sandwich, all measurements below a Cu thickness of 10 ML (14.3 Å) show F (or weak AF) coupling of the Fe overlayer to the Fe(001) substrate, in close agreement with the observations in [16]; above 10 ML, the coupling becomes strongly AF. The critical fields H_1 and H_2 oscillate as a function of the bcc Cu thickness, showing four strong oscillations in magnitude with a period of 2 ML Cu. The maximum critical fields (at ~12 ML Cu) correspond to $J \cong 0.1$ mJ/m^2, which is much weaker than for the Co/Cu/Co sample.

Comparison of Figs. 2 and 3 shows that the variation of

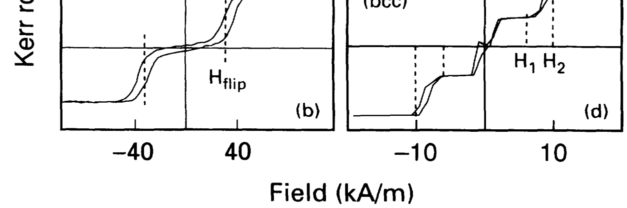

FIG. 1. Longitudinal MOKE hysteresis loops measured on (a),(b) Co/Cu/Co and (c),(d) Fe/Cu/Fe samples. The Cu thicknesses are given for each loop.

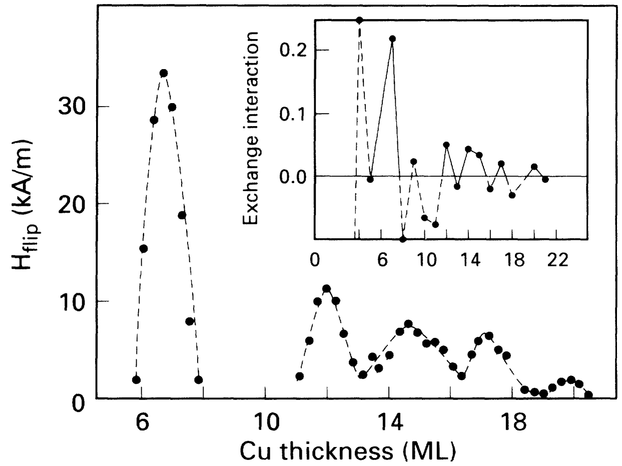

FIG. 2. The critical field H_{flip} defined in Fig. 1(b) for the Co/Cu/Co sample, plotted as a function of the thickness of the Cu spacer. Inset: Calculation according to [9], with $\Lambda_1=8.0$, $\Lambda_2=2.59$, $\psi_1=14.2$, $\psi_2=14.5$, and a relative amplitude of the short-period to long-period terms of 1.25.

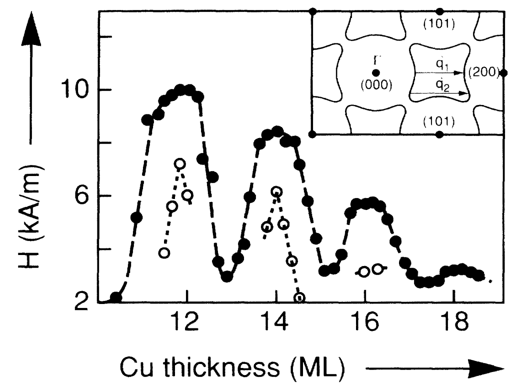

FIG. 3. The switching fields H_1 (○) and H_2 (●) defined in Fig. 1(d) for the Fe/Cu/Fe sample, plotted as a function of the thickness of the Cu spacer. Inset: The bcc Cu Fermi surface, calculated using the ASW technique.

the exchange coupling with the interlayer thickness is entirely different for the two samples. The most apparent explanation of this difference is the different structure of the Cu interlayers. No theoretical indications have been given that the different nature of the magnetic atoms (Fe or Co) by itself would result in different periods (although the amplitude and phase of the oscillations are likely to be affected by this difference [9,18]).

Following the theoretical approach of Bruno and Chappert [9], we have attempted to fit the experimental coupling data for the Co/Cu/Co system in terms of a superposition of a short-period (Λ_1) and a long-period (Λ_2) oscillation. The decay in J was assumed to be quadratic with increasing Cu thickness, and the phases of both oscillations (ψ_1,ψ_2) were regarded as free parameters. The corresponding extremal vectors $\mathbf{q}$ are depicted in Fig. 1(b) of Ref. [9]. Our results are rendered in Fig. 2 (inset), in which the fit parameters are also stated. The form of the fit is in good agreement with the experimental results: A strong AF peak at 6.6 ML is followed by a predominantly F region and subsequently by a series of four alternately sharp and rounded AF peaks of decaying intensity. It was not necessary to add a roughness parameter to the theory to obtain this fit, perhaps highlighting the extremely high growth quality of our sample. However, a certain amount of roughness is of course present, even in the best samples. This roughness may slightly smear out certain predicted features, such as the AF peaks at ~9 ML (weak) and ~4 ML (strong).

The experimental periods are $\Lambda_1=2.60\pm0.05$ ML and $\Lambda_2=8.0\pm0.5$ ML. From de Haas–van Alphen data on the Fermi surface of Cu, Bruno and Chappert have predicted periods of 2.56 and 5.88 ML, whereas we have found that their theoretical approach, when applied to the extremal $\mathbf{q}$ vectors which follow from first-principles self-consistent augmented-spherical-wave (ASW) band-structure calculations, leads to periods of 2.6 and 6.4 ML, respectively. The predicted short-period oscillations are in excellent agreement with experiment. However, there is a discrepancy with respect to the long period. We note that the observed long period is essentially the same as in epitaxial fcc(100) Fe/Cu structures, for which a value of $=7.5\pm0.5$ ML has been found [4].

Another important result concerns the phase of the long-period oscillation. Compared to epitaxial fcc(100) Fe/Cu samples [4], the long-period oscillation in our Co/Cu/Co sample displays a shift of 0.6 ± 0.4 ML towards lower Cu thickness, corresponding to a phase difference $\Delta\phi=(0.2\pm0.1)\pi$. We regard this estimate of $\Delta\phi$ to be more reliable than the value $\Delta\phi\simeq\pi$ previously derived from a comparison of structurally dissimilar Co/Cu and Fe/Cu multilayers [13].

To extend the analysis to the Fe/Cu/Fe system, we have calculated the Fermi surface of hypothetical bcc Cu using the ASW method. The relevant cross section of the Fermi surface is shown in Fig. 3 (inset). The most obvious difference from the fcc case is that the hole orbits around the N point (bcc) have a considerably squarer character than the "dog bone" orbits around the X point (fcc). The extremal $\mathbf{q}$ vectors for the (100) direction, $\mathbf{q}_1$ and $\mathbf{q}_2$, correspond to periods of 2.56 and 2.22 ML, respectively. The consequence of a period of 2.22 ML would be that four oscillations of precisely 2 ML would be found, followed by a phase shift. Such a series of oscillations is indeed found in our measurements (Fig. 3). The thickness interval in which the oscillatory exchange coupling was observed was too small to analyze the experimental results more quantitatively, as for the Co/Cu/Co case. In particular, no assessment of the possible contribution of longer-period oscillations, such as the predicted one with a period of 2.56 ML, can be given. It remains unclear why the Fe/Cu/Fe system is F (or weak AF) up to 10 ML: No long period can be anticipated from the Fermi surface. Furthermore, the first-principles band-structure calculations of the coupling by Herman, Sticht, and van Schilfgaarde [14], which also predict 2 ML oscillations, indicate that the system should become AF for Cu thicknesses above 5 ML. The anomaly suggests the possible presence of pinholes in the Cu interlayer.

In conclusion, we have observed that the exchange interaction across wedge-shaped Cu interlayers in highly epitaxial fcc(100) Co/Cu/Co and bcc(100) Fe/Cu/Fe samples depends in markedly different ways on the Cu thickness. Both systems displayed rapid oscillations, with periods of 2.6 and 2.0 ML, respectively, which were shown to be in good agreement with those expected from the Fermi surface dimensions using the theoretical approach by Bruno and Chappert. An additional 8-ML period was observed in fcc Co/Cu/Co, which was shown to be at variance with the theoretically predicted value by about 30%.

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