Magnetic Layer Thickness Dependence of the Interlayer Exchange Coupling in (001) Co/Cu/Co

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A dependence of the strength of the antiferromagnetic coupling across Cu on the *Co layer thickness* has been observed in an epitaxial fcc (001) triple wedge sample containing two Co wedges and one Cu wedge. Our result is consistent with an oscillation period of 6–7 Å Co — a value that agrees with the period of 3.5 monolayers of Co derived from the extremal wave vector that spans the ellipsoidal hole pocket centered at the X point of the fcc Co spin-down Fermi surface. This shows that the interlayer exchange coupling does not just involve an interaction localized at the interfaces, but is a property of the sandwich as a whole.

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Since the discovery of antiferromagnetic coupling between ferromagnetic layers across nonmagnetic spacer layers [1], its oscillatory nature [2], and its universality [3], much progress has been made in the theoretical and experimental research of this indirect exchange coupling. Several different models have shown that a multiplicity of oscillations may be encountered with periods that can be derived from the extremal Fermi surface spanning vectors of the nonmagnetic interlayer material (see [4,5] and references therein). The validity of the Fermi surface picture has been tested rather extensively through a number of experiments including investigations on Fe/Au(001) [6], Fe/Ag(001) [7], Co/Cu(001) [8,9], Co/Cu(110) [10], Co/Cu(111) [11], and samples with Cu-based alloy interlayers [12,13].

Although the multiplicity and values of the periods are fairly well understood for the monovalent noble metal interlayers, the role of the *magnetic* layer has not been clarified. Dedicated experiments to investigate the latter are scarce. Qui *et al.* [14] have studied the coupling behavior in three (001) Co/Cu/Co sandwiches with different Co layer thicknesses, and concluded that the coupling was a pure interface effect, i.e., independent of the Co layer thickness. However, recent theoretical predictions by Bruno [15] and Barnaś [16] have shown that the magnetic coupling may *oscillate* with the ferromagnetic layer thickness.

In this Letter we report the first experimental evidence for interlayer coupling oscillations as a function of the Co layer thickness in a coherent epitaxial fcc (001) Co/Cu/Co sandwich grown by molecular-beam epitaxy (MBE) on a single-crystal Cu(001) substrate. The Co and Cu layers were deposited in the form of wedges oriented perpendicularly with respect to each other. This allows for independent investigation of the Cu *and* Co thickness dependence of the coupling across Cu(001) in a *single sample*. We will show that the strength of the first and second antiferromagnetic (AF) peaks, such as observed in an experiment of varying Cu thickness, oscillates as a function of the Co layer thickness with a period of 6–7 Å. The behavior is discussed in relation to recent theories and to results obtained from self-consistent augmented-spherical-wave (ASW) band-structure calculations.

The overlayers were deposited on a single-crystalline Cu(001) substrate in a multichamber MBE system (VG Semicon V80M). The two samples that have been prepared (shown schematically in Fig. 1) are composed as follows:

Cu(001)/x Å Co /15 Å Ni/
Co wedge (0 Å – 20 Å; 2.3 Å/mm)/
Cu wedge (0 Å – 8 * y Å; y Å/mm)/
Co wedge (0 Å – 20 Å; 2.3 Å/mm)/
15 Å Ni/x Å Co/10 Å Cu/30 Å Au.

in which $x = 50$ and $y = 5.0$ for sample I and $x = 30$ and $y = 3.1$ for sample II. The reason for the additional x Å Co and 15 Å Ni layers which seem, *a priori*, unnecessary for the present experiment, will be discussed later on. The substrate temperature was 50 °C during the Cu wedge deposition and 20 °C for all other depositions. The thicknesses were determined using a quartz crystal monitor. The slopes and starting points of the wedges and the thicknesses of the other layers were subsequently confirmed after deposition using combined *in situ* Auger electron spectroscopy (AES) and scanning electron microscopy (SEM). The accuracy of the determination of the slopes was better than 10% and the starting points of the two Co wedges were found to coincide within experimental accuracy (0.1 – 0.2 mm). Further details concerning the substrate preparation can be found in [8].

The structure of the layers has been investigated with low energy electron diffraction (LEED) and yielded results consistent with earlier observations [8,17]. The Cu wedge displays identical lattice constants to those of the Cu substrate, and maintains an fcc structure. The Co displays an identical surface net to the Cu, but grows with a face centered tetragonal (fct) structure. The per-

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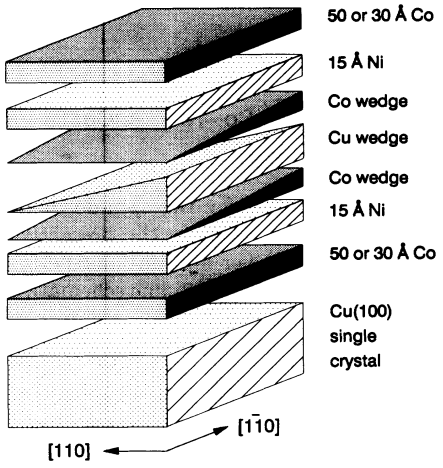

FIG. 1. Stacking sequence and relative wedge orientations for the samples. Mutually perpendicular Co and Cu wedges have been used to allow for independent investigation of the Cu and Co layer thickness dependence of the interlayer exchange coupling in one single sample. The uniform Ni (15 Å) and Co layers (50 Å for sample I and 30 Å for sample II) have been employed so as to eliminate problems related to small MOKE signals at low Co wedge thicknesses. For clarity the cap layers have been omitted.

pendicular Co-Co spacing (1.70 ± 0.02 Å) appears to be reduced by around 4% with respect to the bulk value (1.78 Å).

The antiferromagnetic coupling behavior was investigated at room temperature by measuring magnetic hysteresis loops at various positions on the sample via the longitudinal magneto-optical Kerr effect (MOKE). The additional Co and Ni layers were employed to enable such measurements at the very low Co wedge thicknesses, i.e., to enhance the MOKE signals that otherwise would have been too small because of the combined effect of their scaling with magnetic thickness and the additional reduction in signal associated with a lowering of the Curie temperature with decreasing Co thickness [17]. We have added a uniform Ni layer adjacent to each Co wedge and not a uniform Co layer because the latter would yield an offset in the Co thickness. This is unwanted since the largest Co thickness dependent effects are to be expected at low Co thicknesses [15,16]. Of course the Ni layers may affect the coupling but, as we will discuss later on, this concerns only the amplitude and the phase but not the period of an oscillation with varying Co thickness. Finally, the uniform Co layers that have been added at the outer part of the sandwich merely serve to maintain an optimal accuracy in the determination of the coupling strength: Co(001) layers exhibit a relatively large fourfold in-plane anisotropy, leading to an abrubt well-defined spin-flip transition from antiparallel to saturation [18]. The rather low fourfold in-plane anisotropy of Ni(001), on the other hand, would have led to a gradually increasing magnetization from which it would have been considerably more difficult to accurately determine the coupling strength. Although the samples may seem complicated because of these additional layers it is clear that to enable accurate room temperature MOKE experiments at ultralow Co thicknesses, these layers are crucial.

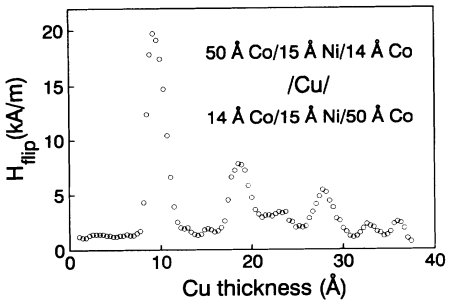

FIG. 2. The flip field as a function of the Cu thickness as obtained on sample I from highly localized MOKE hysteresis loop experiments taken along the Cu wedge at a Co wedge thickness of 14 Å. The coupling strengths corresponding to the peaks at 9.4 Å Cu and 18.6 Å Cu amount to -0.24 and -0.09 mJ/m^2, respectively.

Figure 2 shows the typical Cu thickness dependence of the flip field as observed for samples I and II when performing a positional scan along the Cu wedge at a fixed Co thickness. The flip field is proportional to the strength of the interlayer coupling and is defined in the same manner as in [8]. The coupling behavior in Fig. 2 displays a superposition of a long and a short period oscillation. This is in accordance with earlier observations [8,9] and with the prediction for Cu(001) interlayers [19]. The presence of the short period in both samples indicates that these samples are of high structural quality.

The dependence of the coupling strength on the Co thickness has been investigated for constant Cu thickness precisely at the first (≈ 9 Å Cu) and second (≈ 19 Å Cu) peaks in the AF coupling. In principle, it should be sufficient to perform one positional scan along the Co wedge for each position on the Cu wedge where a peak in the AF coupling occurred. However, to avoid experimental artifacts (e.g., a slight misorientation in scan direction) in connection with the extremely narrow peaks (Fig. 2), we have taken multiple parallel scans along the Co wedge that were densely spaced (≤ 0.2 Å), around the maximum of the first and second AF peaks, in such a way that the AF peak value at each Co thickness is determined with the highest achievable accuracy. The results for the maxima (involving about 2500 hysteresis loop measurements) are shown in Fig. 3. Here the Co thickness dependence of the strength of the first and second AF coupling peaks for sample I (a) and sample II (b) are reported. We remark that the latter sample has been studied in more detail with respect to the Co thickness resolution.

Figure 3 shows that at very small Co thicknesses, below 3 Å, the coupling strength rapidly decreases with decreas-

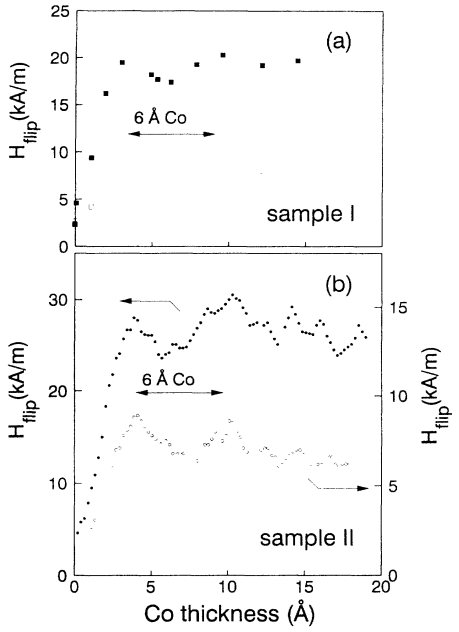

FIG. 3. The strength of the interlayer exchange coupling in the first and second antiferromagnetic peaks as a function of the Co thickness for sample I (a) and sample II (b).

ing Co thickness. At higher Co thickness an oscillatory-like behavior with an apparent period of 6–7 Å is observed in all experimental scans. The decrease at small Co thicknesses is attributed to Co-Ni alloy formation or the presence of Ni patches at the interface with Cu. It is known that these reduce the coupling [20]. However, this effect cannot explain the oscillatory changes in the strength of the coupling at the larger Co thicknesses. These oscillations are therefore interpreted as reflecting the intrinsic Co thickness dependence of the coupling.

The present experiment essentially probes the electron interference effects in the Co layer adjacent to the Cu. Following Bruno [15] and Barnaś [16], we describe the potential of the sample as a sequence of potential steps in the growth direction and with translation invariance in the in-plane directions, and consider the effect of multiple reflections of free-electron-like waves. This situation is shown schematically in Fig. 4 for one-half of the experimental system and for the spin-down direction. The potential steps for the spin-up direction are considerably smaller. The strength and phase of the coupling are determined by the spin dependence of the reflectivity of electrons at the Cu/Co interface at $z = 0$. This reflectivity, in turn, is determined by interference effects due to the partial reflection and transmission at each subsequent potential step at $z > 0$. As a result, the coupling will oscillate with the thickness of any particular layer, with a period which, in the limit of large spacer and magnetic layer thickness, is determined by the Fermi wave vector in that magnetic layer. In the present case this is the wedged Co layer sandwiched between the Cu and the Ni.

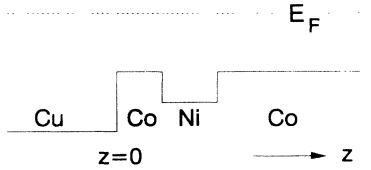

FIG. 4. Potential steps for the spin-down direction that occur at the several interfaces in the presently investigated sample. Only the interlayer and one magnetic (Co/Ni/Co) layer are represented. The broken line schematically indicates the position of the Fermi level.

Evidently the role of the Ni layer is simply to introduce an extra potential step to effectively reduce the Co thickness. The period of the oscillation with this thickness is thus solely a property of Co. The combination of the precise heights of the potential steps and the thicknesses of the Ni layer and the outer Co layer merely set the effective reflection amplitude at the Co-wedge/Ni interface and thus only affect the amplitude and phase of the oscillation.

Bruno has recently proposed that the period for the Co/Cu system is mainly determined by the spin-down electronic structure of Co [15]. The good agreement between the period of 3.5 ML Co (6.2 Å) that he derived from the Fermi wave vector of the spin-down electrons of Co and the presently observed experimental period, provides support for his proposal. To evaluate possible complicating effects resulting, for example, from the experimentally observed tetragonal disortion of the Co, we have carried out *ab initio* self-consistent band-structure calculations using the ASW method. In undistorted fcc Co the spin-down Fermi surface contains an ellipsoidal hole pocket centered at the X point, determined by the free-electron-like bands. Its spanning vector parallel to the growth direction yields a period of 6.1 Å — a value which agrees well with Bruno's result. However, the analogous oscillation period for fct Co, with a perpendicular compression of 4% and a lateral expansion of 2% in both directions, is reduced by 23% to 4.7 Å. This shows that the aforementioned agreement might be fortuitous. Other reasons for keeping some reserve with respect to the theory given in [15] are that (i) inspection of the spin-down Fermi surface of fcc Co has revealed ten different extremal spanning vectors for (001) growth [21] and (ii) for the low Co thicknesses here, the asymptotic thickness limit, where the Fermi-level wave vector determines the period, might not have been approached sufficiently close. Clearly a considerable research investment in theory is required to resolve these questions. The fact that no clear third peak is observed might be related to such complications and could be explained, for instance, by a beating effect resulting from an interference with a second oscillation.

In conclusion, we have shown a clear dependence of the strength of the coupling across Cu(001) on the Co

thickness suggesting an oscillatory behavior with a period of 6–7 Å Co. This period is in good agreement with a recent prediction [15] where the period stems from the extremal wave vector spanning the ellipsoidal hole pocket centered at the X point of the spin-down Fermi surface of fcc Co. From ASW band-structure calculations we have shown that more realistic theoretical descriptions should discuss the effects of a tetragonal distortion of the Co layers and the possible presence of additional oscillations. Although, at present, the theory is still in an embryonic stage, the results strongly suggest that the present type of exchange coupling experiments provides spin selective information on the Fermi surfaces of ferromagnetic materials.

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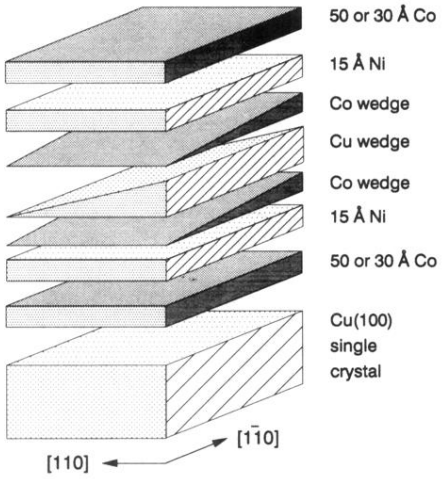
[21] Conservation of the parallel component of the wave vector under reflection implies that not all ten extremal wave vectors are relevant. For example, if we are essentially measuring strength changes of the long period (corresponding to the center of the 2D Brillouin zone), only those between points on the Γ-X line will be relevant [P. Bruno (private communication)].



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