Phase Shifts in the Oscillatory Interlayer Exchange Coupling across Cu Layers

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The dependence of the phase of the oscillatory exchange coupling between ferromagnetic (FM) layers across fcc Cu on the *composition of the ferromagnetic layer* has been studied in several epitaxial samples. In the case of (001) systems, phases remain constant for the long period oscillation, but for the short period oscillation they vary monotonically with the d-electron occupation number of the FM layers, at a rate of about 1.5 Å/electron. In the case of (110) systems the variation is nonmonotonic. A framework is presented with which the above observations can be understood.

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Since the discovery of oscillatory interlayer exchange coupling between transition metal ferromagnetic layers across nonmagnetic spacer layers [1], much progress has been made in the theoretical and experimental research of this phenomenon. Several different models have shown that the periods can be derived from the extremal Fermi surface spanning vectors of the nonmagnetic interlayer material (see [2,3], and references therein). The validity of these models regarding the oscillatory periods has been tested rather extensively [1]. However, their experimental determination does not reveal decisive information on the nature of the interactions, which give rise to the oscillatory coupling. New theoretical insight can be obtained by addressing the more sensitive issues of the *strength* and *phase* of the oscillating coupling. While recent theoretical predictions that the coupling strength may oscillate as a function of magnetic layer thickness [4,5] have now been confirmed in the Co/Cu/Co(001) system [6] and the Fe/Cr(100) system [7], the situation concerning the phase of the coupling is still unclear.

Different theoretical approaches have led to varying predictions of the phase. In analogy to the Friedel-Anderson-Caroli theory for the oscillating exchange interactions between magnetic impurities in a free electron metal, Bruno predicted that for systems containing 1 monolayer (ML) thick ferromagnetic layers with exchange-split s bands the phase of the coupling would depend as $2\pi n$ upon the $\mathbf{k}_{\|}$-resolved occupation numbers n of the virtual bound state (VBS) [8]. No extension was made to the case of exchange-split d states, or to thicker ferromagnetic (FM) layers. *Ab initio* electronic structure calculations by Lang *et al.* [9] of the interlayer exchange coupling across Cu(001) between Fe and Co monolayers yielded a shift in the peak positions of about 2.3 Å. However, no analysis or discussion on the physical origin was given. In contrast, with the aid of a second model proposed by Bruno, describing the coupling phenomenon as originating in spin-dependent reflection of electron waves at the interfaces [2,4], the essential electronic properties important for phase shift effects can, in principle, be identified although Bruno made no predictions regarding phases.

An experimental study of antiferromagnetic (AF) peak shifts for fcc (001) sputtered (Fe-Co-Ni)/Cu multilayers by Coehoorn and Duchateau [10] revealed a regular variation with the magnetic alloy concentration. Through extrapolation one could derive a shift of about 2.5–3 Å from Fe- to Co-based systems for the first AF peak, whereas the shift measured going from Co- to Ni-based systems was less than 1 Å. For sputtered (111) oriented Co-Ni/Cu systems Kubota, Ishio, and Miyazaki observed similarly small peak shifts [11]. It should be noted that variations in the degree of interface alloying or intermixing observed in these sputtered samples could in fact be masking the true trend [12]. In addition, for the (001) case the analysis has not made clear to what extent the peak shift is related to the separate phase shifts for the two oscillations. To extract reliable information concerning small phase shifts it is therefore desirable to avoid intermixing by preparing molecular-beam epitaxy (MBE) samples. As yet, however, no dedicated MBE studies have addressed the phase shift problem: one can only compare observations obtained for *different* samples, prepared under different conditions, by several groups ([13–16]), with the associated uncertainties in *absolute* interlayer thickness, even where AF-coupling peaks are found in the most carefully prepared wedge samples.

In this Letter we undertake to reduce these uncertainties by preparing and measuring *multiple* MBE-grown sandwiches simultaneously on the same single crystal substrate sharing *the same Cu wedge*. In this manner, we are able to unambiguously determine phase shifts with great accuracies. We will show that in the case of coupling across Cu(001) the phase of the short period oscillation varies monotonically with the number of d electrons in the FM layer, whereas the phase of long period oscillation remains constant. In contrast, in the case of (110) oriented systems the phase varies conspicuously nonmonotonically

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with the d band occupation of the magnetic layers. We will demonstrate that this strongly differing behavior can be understood from features in the band structures of the FM metals.

Two sample concepts were developed to extract reliable phase shift information. The first sample (type I) was designed around a triple wedge, composed as follows [6]: Cu(001)/50 Å Co/15 Å Ni/Co wedge/Cu wedge/Co wedge/15 Å Ni/50 Å Co/10 Å Cu/30 Å Au. The Co wedges were grown at right angles to the Cu wedge. This sample was designed to probe the dependence of the phase on the thickness of the ferromagnetic layer. In the second sample concept a single Cu wedge was prepared which separated ferromagnetic layers that were deposited in the form of (three) parallel stripes running along the direction of the Cu wedge with each stripe having a different composition. The ferromagnetic layers (stripes) in samples (IIa, IIc) consisted of Co, Ni, and $Co_{50}Ni_{50}$, whereas sample (IIb) incorporated Co, Ni, and $Fe_{60}Ni_{40}$ layers. The samples were composed as follows:

Samples IIa, IIb: Cu(001)/x Å Co/FM/Cu wedge/FM/x Å Co/10 Å Cu/20 Å Au;

Sample IIc: Cu(110)/25 Å Co/FM/Cu wedge/FM/25 Å Co/10 Å Cu/20 Å Au,

with x = 40 Å and FM = 6 Å Ni, 12 Å $Co_{50}Ni_{50}$, or absent in the case of samples IIa and IIc, and with x = 35 Å and FM = 6 Å Ni, 15 Å $Fe_{60}Ni_{40}$, or absent in the case of sample IIb. Typical wedge slopes are 3 Å/mm. Portions of all samples, viz., where the FM layer is absent in the above notation, therefore comprised the simple Co/Cu wedge/Co system which was necessary for calibration purposes, not only to establish a reference point for defining phase shifts, but also [through control of the coupling strength and presence of short periods in the (001) oscillatory coupling] for ensuring the constant high quality of the sample. The overlayers were deposited on single crystalline copper substrates in a multichamber MBE system. The substrate temperature was 50 °C during the Cu wedge deposition and 20 °C during all other depositions. The accuracy of the wedge slopes was better than 10%, as controlled during deposition by a quartz monitor, and confirmed after deposition using combined *in situ* Auger electron spectroscopy (AES) and scanning electron microscopy (SEM). Further details concerning the substrate and sample preparation and the structural characterization can be found in [17]. No evidence was found in the AES for differing interdiffusion in either (100) or (110) FM/Cu interfaces.

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). Reliable determination of the phase shift requires a careful experimental procedure. As an example, Fig. 1 illustrates how the peak shift between (001) oriented Co/Cu/Co and Ni/Cu/Ni (sample type I) was derived. In order to avoid experimental artifacts, related to a possible minor azimuthal misalignment of the sample, multiple scans were performed. The figure shows the Cu thicknesses at which the first and second maxima in the AF coupling were found, recorded at twelve different positions across the triple wedged sample. Once the baseline had been established, peak shifts could be defined by measuring the offset observed when moving from the Co to the Ni interface layers: in this case the AF peaks obtained for Ni were shifted 1.2–1.3 Å towards thicker Cu than in the case of Co.

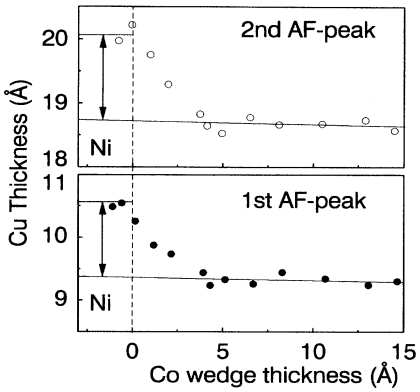

FIG. 1. Peak positions of the first and second AF peaks as obtained from Cu scans (at 300 K) of sample I as a function of the Co wedge thickness.

Another important aspect illustrated in Fig. 1 is the Co thickness dependence of the peak shifts. It can be seen that beyond an FM layer thickness of 4 Å the peak positions remain constant. Presumably, at this point, the interface consists entirely of Co [18]. This observation is consistent with recent theoretical results obtained by Nordström *et al.*, who from *ab initio* electronic structure calculations predicted a negligible phase difference of oscillatory coupling for (001) Co/Cu systems with 1 and 5 ML Co thicknesses [19]. This layer thickness independence suggests that the approach adopted for the type II samples, where only a *constant* FM layer thickness is considered, will not yield erroneous results. Figure 1 also shows that the peak shift is almost identical for both AF peaks, which indicates that the oscillatory period is unaffected by the nature of the FM layer. Although in agreement with models relating the oscillation period solely to the Fermi surface dimensions [2,3], this is in contradiction with the recently proposed well-depth-related period models, according to which the change in potential step height from Co to Ni should induce a period variation [20].

The results obtained for the various (001) oriented systems investigated are presented in Fig. 2, together with the Co/Cu reference curves. In the latter short period oscillations were present at larger Cu thicknesses (see also Fig. 2 of Ref. [6]), whereas for the other cases the fact that the AF peaks remain narrow suggests that they also contain the short period. The observed peak shifts relative

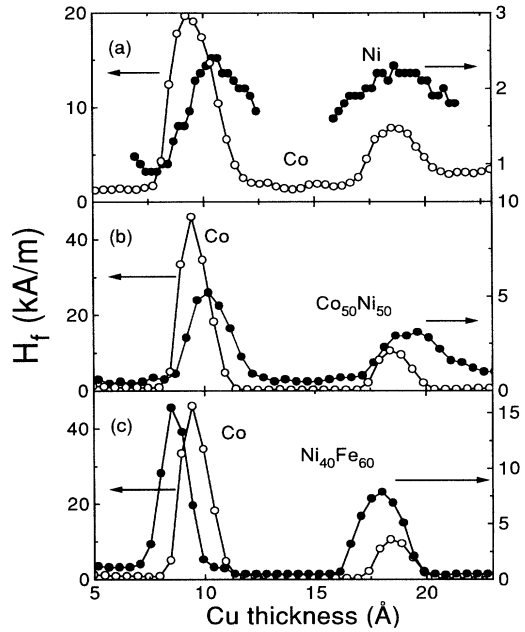

FIG. 2. Cu thickness dependence of the flip field (at 300 K) for various ferromagnetic materials at the interface with a Cu(001) interlayer.

to Co are +1.3 Å (Ni), +0.8 Å ($Co_{50}Ni_{50}$), and −0.5 Å ($Fe_{60}Ni_{40}$). These results reveal a monotonic, not quite linear, shift of the AF peak position with the number of $3d$ electrons. Fitting a superposition of a long and a short period oscillation to the oscillatory behavior observed for the different compositions showed that the peak shifts are primarily related to the short period: Leaving the phase of the long period oscillation unaltered and varying the phase of the short period oscillation is sufficient to account for both the peak shifts *and* the modifications in relative intensities of the first two AF peaks [21]. The peak shifts should therefore be interpreted in terms of a phase shift of the short (4.6 Å) period, with an average rate of about 0.65π/electron.

These results are quite dissimilar from the large phase shifts (2π/electron) predicted by the VBS model. Even greater disagreement was found for the (110) oriented systems. For this orientation, the analysis of peak shifts in terms of phase shifts is facilitated by the presence (experimentally) of only a single long oscillatory period [16]. Measured variations in the flip fields with Cu thickness around the first AF peak are shown in Fig. 3(b). For Co/Cu/Co a broad peak, centered at 8.5 Å with a coupling strength $J = 0.65$ mJ/m^2, was measured, in accordance with previous observations [16]. For the $Co_{50}Ni_{50}$ alloy the peak is phase shifted towards larger Cu thicknesses by 1 Å. However, surprisingly, the peak obtained for Ni/Cu/Ni is phase shifted in the *opposite* direction (towards smaller Cu thicknesses) by 1.7 Å, in sharp contrast with the monotonic behavior observed in the case of (001) oriented samples. This behavior clearly falls beyond the monotonic dependence predicted

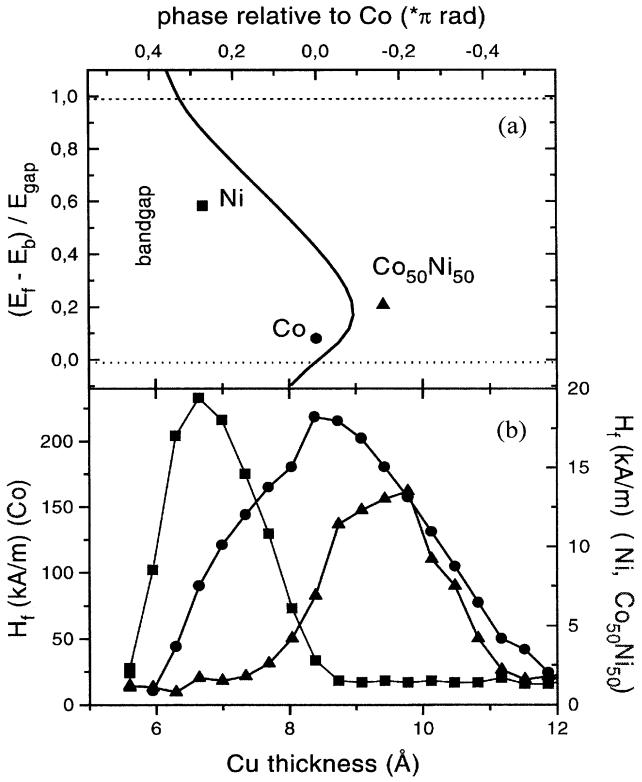

FIG. 3. (a) Calculation of the phase of the coupling as a function of the relative position of the Fermi level with respect to the bottom of the band gap. (b) Flip field as a function of the Cu thickness around the first AF peak for the Co/Cu (circles, left axis), $Co_{50}Ni_{50}$/Cu (triangles, right axis), and Ni/Cu(110) (squares, right axis) systems.

on the basis of the VBS picture and demands further consideration.

The remainder of this Letter will be devoted to the mechanism underlying these remarkable phase shift observations. The basis is formed by Bruno's "electron optics" model [2,4]. An important, as yet unrecognized, consequence of the model is that the phase of the coupling is directly determined by the phase of the complex reflection coefficient of electron waves at the Cu/FM interface [see, for instance, Eq. (14) in Ref. [2]]. If applied to free-electron-like electronic structures, phase shifts disappear in the limit of large FM and spacer thicknesses [22]. Non-free-electron-like features in the band structure of the FM metal drastically change this picture. The crucial feature in the real band structures of fcc $Co_{1-x}Ni_x$ alloys is the presence at certain points in the zone of an energy band gap at the Fermi level E_F. Clearly, at the band gap the complex reflection coefficient becomes unity in amplitude and acquires an imaginary component depending on the position of the electron state under consideration relative to the bottom of the gap. It will hence be clear that the phase of the coupling may depend sensitively on the position of E_F relative to the band gap and is therefore also expected to depend on the composition of the FM layer.

To infer whether all experimentally observed effects are consistent with this picture we have (i) performed self-consistent *ab initio* augmented spherical wave (ASW) band structure calculations for fcc Co, Ni, and (within the virtual crystal approximation) $Co_{50}Ni_{50}$ and (ii) used this information to calculate phase shifts within Bruno's model for a situation that simulates our experimental situation. The ASW results show that the spin down band structures display gaps at E_F along the lines relevant for the short period in the oscillatory coupling across Cu(100) and for the oscillatory coupling across Cu(110). No gap is present at E_F in the spin down band structure along the line, which is relevant to the long period oscillation across Cu(100).

To simulate the effect of a band gap at E_F on the phase of the coupling, the wave vector dependent complex reflection coefficients of the Cu/FM interface were calculated using an approach previously used in 1D-LEED theory [23]. The multiple scattering processes occurring within the FM layer at the *lattice* planes parallel to the interfaces (which are responsible for the band gaps) were accounted for by introducing a single transmission parameter for the scattering of the spin down electrons at each FM lattice plane. The results of a model calculation with $t_{Cu} = 8.5$ Å and infinitely thick FM layers are summarized in Fig. 3(a). Here, the vertical axis corresponds to the position of the Fermi level relative to the bottom (E_b) of the band gap (E_g). In the calculation the transmission parameter was chosen to match the band gap found (neglecting symmetry arguments) for Co from the band structure calculation for the (110) case. The potentials were chosen to be such that the Fermi levels for Co(Ni) are, realistically, close to the lower (upper) band edges. No reflection at the interfaces or at lattice planes was assumed for spin up electrons. It can be seen that close to the $Co_{50}Ni_{50}$ alloy composition an extremum in phase is obtained. This trend, which is insensitive to details of the calculations, is in good qualitative agreement with the nonmonotonic phase shift measured for the (110) systems. We stress that the actual band structures are considerably more complicated than assumed in our model calculations. A quantitative explanation will require a much more complicated calculation if based on more realistic band structures.

The FM spin down band structure that is relevant for the (001) short period revealed a wider, 1.9 eV, gap, with the Fermi level for Co closer to the center of the gap $[(E_F - E_b)/E_g \approx 0.4]$. As suggested by Fig. 3(a), phase changes from Co to Ni $[(E_F - E_b)/E_g \approx 0.9]$ would then be expected to be monotonic rather than showing a maximum. This is, indeed, the behavior observed experimentally. Along ΓX, the line which is relevant to the long period oscillation along Cu(001), no band gaps are present at or near E_F. The band structure accommodates a highly dispersive nearly free electron band, the precise position of which had been used earlier to successfully predict oscillations in the coupling strength with the Co layer thickness [4,6]. Within the framework introduced above these conditions are not expected to lead to significant phase shifts. This is again in agreement with the experimentally observed absence of a phase shift for the long period.

In summary, the careful experimental approach adopted here, in combination with the atomically sharp interfaces produced via MBE sample preparation onto single crystalline substrates, has enabled the unambiguous determination of the phase shift in the oscillatory exchange coupling. Within Bruno's "electron optics" model for interlayer exchange coupling we have shown that band gaps at crucial (extremal) points of the ferromagnets' spin-dependent Fermi surface control the phase of the coupling.

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[21] We used fit parameters (defined as in [22]) of $\Lambda_1 = 2.50$ ML, $\Lambda_2 = 7.00$ ML, $r = 1.73$, $\psi_2 = 1.75$ for all compositions. For the short period phase of Co, for instance, we find $\psi_1 = 1.48$ as giving the correct peak intensity ratio of 3.5 for the first to second AF peak [see Fig. 2(a)], whereas for Ni we find $\psi_1 = -0.63$ giving a ratio of 1.4 similar to the experiment in Fig. 2(a).

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