Induced Spin Polarization in Cu Spacer Layers in Co/Cu Multilayers

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X-ray magnetic circular dichroism measurements reveal that Cu atoms in Co/Cu multilayers exhibit an induced magnetic spin moment in the d shell. The magnitude and sign of the moments and their variation with Cu thickness are in excellent agreement with moments calculated from a local spin density functional theory. The results indicate that the Cu d electrons play a large role in mediating the exchange coupling between successive Co layers.

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The discovery [1] of an oscillatory exchange coupling between successive 3d transition metal ferromagnetic layers, separated by a nonferromagnetic metal layer, has stimulated much work regarding its origin. To explain this coupling, the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction was adapted to a planar geometry [2]; the coupling is also consistent with a quantum-well model [3]. The latter model assumes a quantum confinement of electrons for certain energies and momenta which depends on the thickness of the spacer layers. Recently, quantum well states have been observed by inverse photoemission measurements of thin Cu and Ag layers grown on Co(100) and Fe(100) single crystals [4] and spin-polarized photoemission experiments [5-7] have shown their magnetic polarization. The precise nature of the coupling remains controversial [8], and the relationship between the magnetic polarization in the spacer layer and the exchange coupling is imperfectly understood. Previously, the spin polarization of the spacer layer has been studied for Fe/Cr/Fe [9] and Fe/Ru/Fe sandwiches [10] and Co/Pt and Co/Pd multilayers [11]. In all these systems the spacer is a transition metal, and the coupling to the d bands near the Fermi level strongly influences the spin polarization.

Here we present evidence for an induced magnetic moment in a noble metal spacer layer, i.e., Cu in Co/Cu multilayers. Both the sign and magnitude of the moment in the Cu d shell is directly measured by means of x-ray magnetic circular dichroism (XMCD). The Cu polarization is found to be concentrated near the Co interface. Its d component, which is parallel to that of Co, has a value of less than 0.05μB and is almost entirely of spin character (no detectable orbital component). The moment and its dependence on Cu thickness is found to be in excellent agreement with self-consistent local spin density functional (LSDA) calculations.

All samples were grown by dc magnetron sputtering. Reference samples of hcp Co and fcc Ni of 200 Å thickness were grown on Si(100) using a 100 Å Ru buffer layer and a 20 Å Ru capping layer to prevent oxidation. The Co/Cu multilayer samples were grown as structures, Si(100)/Ru(50 Å)/20(Co10 Å)/Cu(x Å))/Ru(15 Å) with x = 4, 5.5, and 13 Å. At these Cu layer thicknesses, the coupling between the Co layers is ferromagnetic [1]. We shall refer to these samples as Co10/Cu(x). We also studied a 200 Å Co90Cu10 alloy film, grown on Si(100) and a 100 Å Ru buffer layer, which was capped with 15 Å of Ru. All samples had an in-plane easy magnetization axis, saturation fields below 30 Oe, and remanent in-plane magnetization of nearly 100%.

The XMCD experiments were performed at the Stanford Synchrotron Radiation Laboratory (SSRL) on beamline 8-2, as discussed before [12]. The magnetization direction of the sample relative to the photon spin was changed by placing the sample in the gap between two coils and switching the magnetic field at each photon energy step. Spectra were measured by recording the photocurrent of the sample with a picoammeter, for both saturation and remanence magnetization at 2° grazing x-ray incidence. The spectra were normalized to the photon flux by division by the electron yield signal from a gold grid monitor [13]. For the Co L3,2 spectra a linear background was subtracted, with the slope determined by the pre-edge region. For the Cu L3,2 spectra the subtracted background was nonlinear and consisted of a polynomial fitted to the properly scaled extended fine structure (EXAFS) signal of Co metal. This procedure
removed a sizable EXAFS modulation of Co metal over the Cu L-edge region. Finally, the spectra were rescaled to unit step height far above the L edges, so that the measured dichroism signal corresponds to a per-atom basis [13].

Figure 1 shows experimental spectra for various Co/Cu multilayers, recorded for parallel (solid lines) and antiparallel (dashed lines) alignment of the photon spin and the sample magnetization directions, and their difference spectra. The spectra shown in Figs. 1(a) and 1(b) for Co(10)/Cu(5.5) are characteristic of the Co L_{2,3} spectra of all samples. The Cu L_{2,3} spectra and difference spectra shown in Figs. 1(c) and 1(d) also exhibit a dichroism effect, unambiguously demonstrating a magnetic polarization of Cu. The largest effect is observed for the Co_{90}Cu_{10} alloy. For the multilayers, the dichroism effect weakens with increasing Cu thickness. Since the dichroism effect has the same sign at the Co and Cu L_{2,3} edges, the observed Cu moments are parallel to those of Co.

The L_3 and L_2 dichroism intensities, \( \Delta A_{L_3} \) and \( \Delta A_{L_2} \), i.e., the areas of the difference spectra shown in Figs. 1(b) and 1(d), are related to the orbital moment \( M_L = -(L_2 \mu_B / h) \) of the d valence shell according to [14]

\[
\Delta A_{L_3} + 2 \Delta A_{L_2} = \Delta A_{\text{orb}} = B \frac{A_i}{n} M_L ,
\]

where \( A_i \) represents the polarization-averaged summed intensities of the L_3 and L_2 resonances (“white lines”), \( n \) is the number of holes in the d shell in the ground state, and \( B \) is a constant. If the magnetic dipole term is negligible, a sum rule can be derived for the spin moment \( M_S = -2(S_2 \mu_B / h) \) of the d shell as well [15]:

\[
\Delta A_{L_3} - 2 \Delta A_{L_2} = \Delta A_{\text{spin}} = C \frac{A_i}{n} M_S .
\]

The proportionality constants \( B \) and \( C \) depend on the angular momenta of the p core and d valence shells involved in the dipole transition [14, 15]. Use of the above equations requires the determination of \( A_i \) from the data and knowledge of \( n \) which, in practice, cause large uncertainties in the derived values for \( M_L \) and \( M_S \) [12]. A more accurate procedure is to use the fact [16] that \( A_i \) is proportional to \( n \). The two quantities are linked by Fermi’s golden rule according to \( A_i = |M_{pd}|^2 n \), where \( M_{pd} \) is the radial dipole transition matrix element. Atomic calculations show that \( M_{pd} \) is constant to within a few percent for the 3d elements Co through Cu [17] so that the proportionality constants \( B^* = B A_i / n \) and \( C^* = C A_i / n \) are transferable for atoms with similar atomic numbers.

We have tested the sum-rule and transferability concept by measuring high quality XMCD spectra of Co and Ni metal. Using the calculated values \( M_S = 1.64 \mu_B \) and \( M_L = 0.14 \mu_B \) [18] and the measured XMCD intensities \( \Delta A_{L_3} \) and \( \Delta A_{L_2} \) for Co metal as a standard, we determine the proportionality constants \( B^* = -21.0 \mu_B \) and \( C^* = -5.6 \mu_B \). From the XMCD intensities for Ni metal, we then derive the values \( M_S = 0.62 \mu_B \) and \( M_L = 0.07 \mu_B \).

<table>
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<tr>
<th>Sample</th>
<th>Atom</th>
<th>( M_S^* ) (( \mu_B ))</th>
<th>( M_L^* ) (( \mu_B ))</th>
<th>( M_S ) (( \mu_B ))</th>
<th>( M_L ) (( \mu_B ))</th>
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<td>Co</td>
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<td>0.14</td>
<td>1.64</td>
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<tr>
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<td>Ni</td>
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<td>0.157</td>
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<td></td>
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<tr>
<td></td>
<td>Cu</td>
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<td></td>
<td>1.64</td>
<td>0.07</td>
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* Determined from Eqs. (1) and (2) relative to the listed theoretical values for Co metal.

b From Ref. [18].
$M_L = 0.06 \mu_B$ which agree well with the calculated values [18] $M_S = 0.64 \mu_B$ and $M_L = 0.07 \mu_B$. The dichroism results for Co and Ni metal are summarized in Table I. We conclude that the transferability concept works to an accuracy of about 15% between Co and Ni and thus we presume that we can extend it to Cu.

The results for the spin and orbital moments for Cu derived from the data are listed in Table I, and the spin moments are plotted in Fig. 2(a) as a function of the Cu layer thickness. In particular, we note the small size of the measured Cu moments, indicating the extreme sensitivity of the XMCD technique. It is interesting that the Cu dichroism signals at the $L_2$ edge around 934 eV and at the $L_2$ edge around 954 eV are of nearly equal magnitude. This indicates a nearly vanishing orbital $d$-band moment according to Eq. (1).

To better understand the experimental results, we carried out local spin density calculations for [111] oriented magnetic multilayers, since the sputtered films were largely oriented along [111]. The calculations were done for superlattices of three Co layers and a variable number of Cu layers, denoted (3Co/yCu), with $y = 2, 3, 4, 7,$ and $8$. The (3Co/yCu) sandwich was repeated in the unit cell, to ensure a lattice vector normal to the interface. Stacking between the Cu-Cu planes was taken in the usual $ABC$ pattern appropriate for [111] fcc. Stacking between Co-Co and Co-Cu planes was $AB$ appropriate to hcp, and arranged so that the entire cell preserved inversion symmetry. The lattice constant was set to that of bulk Cu, 3.615 Å. The Co-Co interplanar spacing was compressed by 5%; the Co-Cu spacing was taken to be the average of the Co-Co and Cu-Cu planes. In all cases, coupling was taken to be ferromagnetic. The basis included orbitals to $l = 2$ and the atomic spheres approximation with the “combined correction” term [19] and the Barth-Hedin exchange functional were employed [20]. Integrations over the Brillouin zone were made with the linear tetrahedron method, using a mesh of $24 \times 24 \times 8$ points. This was sufficient to converge the magnetic moments to a precision better than 0.001$\mu_B$.

Theoretical results for the layer-by-layer $d$-band spin moments in a multilayer consisting of three Co layers and seven Cu layers are shown in Fig. 2(b). The Co moments are close to those in bulk Co and are also constant with Cu layer thickness, as summarized in Table I. The Cu moments are largest in the Cu interface layer ($0.046 \mu_B$) where they are parallel to the Co moment, then oscillate to a small negative value ($-2.8 \times 10^{-4} \mu_B$) and become positive again in the center layers ($\sim 1.6 \times 10^{-3} \mu_B$). Since the dichroism measurements average over all Cu atoms, we have listed in Table I the average Cu moments calculated for structures with thicknesses close to the experimental ones. A comparison of the calculated and experimentally determined average moments, plotted in Fig. 2(a) as a function of Cu layer thickness, reveals a striking agreement. In the figure, we also show, as solid and dash-dotted lines, curves fitted to the experimental and calculated data which vary as $1/d_{Cu}$, indicating a dominant interfacial nature of the Cu spin polarization, with a value of $\sim 0.05 \mu_B$ [21].

The enhanced Cu $d$ moment near the interface is the result of a considerable hybridization of the Cu and Co $d$ orbitals near the interface. Our multilayer calculations show an increase in the number of $d$ holes per Cu atom as its separation from the Co layers decreases. Moreover, the Cu $L_{3,2}$ spectra in Fig. 1(c) clearly show an increase in the “white line” intensity with decreasing Cu thickness, again indicating an increased number of $d$ holes for Cu atoms near the interface. The above conclusions for the multilayers are supported by the experimental results for the Co$_{20}$Cu$_{10}$ alloy which show a still larger spin moment and white line intensity. In this case Cu atoms are surrounded by even more Co neighbors.

Unfortunately, the relatively large moment on the Cu interface atoms in the Co/Cu multilayers makes it virtually impossible to deduce experimentally whether the interior Cu atoms are spin polarized [21]. The excellent agreement between experiment and theory reported here for the absolute size of the Cu interface moment, however, lends credence to the theoretical determination of $d$
spin moments on the interior Cu atoms. They are found to oscillate in sign and are at most 3% of the Cu interface moments in magnitude. As shown in Fig. 2(c), the sum of the Cu s and p spin moments at the interface is calculated to be opposite in sign to the Cu d moments and about 30% smaller. The sign and small size of the Cu p moment has, in fact, been verified by Cu K-edge XMCD measurements [22]. Our calculation also shows that the sp moment decays even more rapidly than the d moment so that in the interior of the film the sp moment is only 25% of the d moment. These results strongly suggest that the exchange magnetic coupling of the Co layers is largely mediated by the Cu d electrons. The importance of Cu d electrons in the exchange coupling has also been suggested by spin-polarized photoemission measurements for Cu on Co(100) [6]. We conclude that models of the exchange coupling that ignore the Cu d bands may be misleading.

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[17] The radial dipole matrix elements for transitions 2p3d4s4s → 2p3d3f4s4s in atomic Co and 2p3d4s4s → 2p3d3f04s4s in atomic Ni differ by only about 3% [P. S. Bagus (private communication)].
[21] The measured 1/dCu falloff of the average Cu moment with layer thickness would also follow from an RKKY-like amplitude falloff of the layer-by-layer Cu moment. Our results do not contradict the spin polarization observed in Cu deposited on Co(100) up to at least 10 monolayer coverage by spin-polarized angle-resolved photoemission [6, 7]. Such measurements are very sensitive to the existence of spin polarization because they sample only a restricted region of the Brillouin zone.
[22] A. Fontaine and G. Schütz (private communication).