

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

M. G. Samant,¹ J. Stöhr,¹ S. S. P. Parkin,¹ G. A. Held,² B. D. Hermsmeier,³ and F. Herman¹

¹IBM Research Division, Almaden Research Center, 650 Harry Road, San Jose, California 95120

²IBM Research Division, Thomas J. Watson Research Center, P.O. Box 218, Yorktown Heights, New York 10598

³IBM San Jose, 5600 Cottle Road, San Jose, California 95153

M. van Schilfhaarde

SRI International, 333 Ravenswood Avenue, Menlo Park, California 94025

L.-C. Duda, D. C. Mancini, and N. Wassdahl

Department of Physics, Uppsala University, S-751 21 Uppsala, Sweden

R. Nakajima

Department of Materials Science, Stanford University, Stanford, California 94305

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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 $0.05\mu_B$ 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{Co(10 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 Co(10)/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 20° 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 $L_{3,2}$ spectra a linear background was subtracted, with the slope determined by the pre-edge region. For the Cu $L_{3,2}$ 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 Co₉₀Cu₁₀ 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, ΔA_{L_3} and ΔA_{L_2} , i.e., the areas of the difference spectra shown in Figs. 1(b) and 1(d), are related to the orbital moment $M_L = -\langle L_z \rangle \mu_B / \hbar$ of the d valence shell according to [14]

$$\Delta A_{L_3} + \Delta A_{L_2} = \Delta A_{\text{orb}} = B \frac{A_t}{n} M_L, \quad (1)$$

where A_t 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 $M_S = -2\langle S_z \rangle \mu_B / \hbar$ of the d shell as well [15]:

$$\Delta A_{L_3} - 2\Delta A_{L_2} = \Delta A_{\text{spin}} = C \frac{A_t}{n} M_S. \quad (2)$$

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_t 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_t is proportional to n . The two quantities are linked by Fermi's golden rule according to $A_t = |M_{pd}|^2 n$, 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 $B^* = BA_t/n$ and $C^* = CA_t/n$ 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 ΔA_{L_3} and ΔA_{L_2} for Co metal as a standard, we determine the proportionality constants $B^* = -21.0 \mu_B^{-1}$ and $C^* = -5.6 \mu_B^{-1}$. From the XMCD intensities for Ni metal, we then derive the values $M_S = 0.62 \mu_B$ and

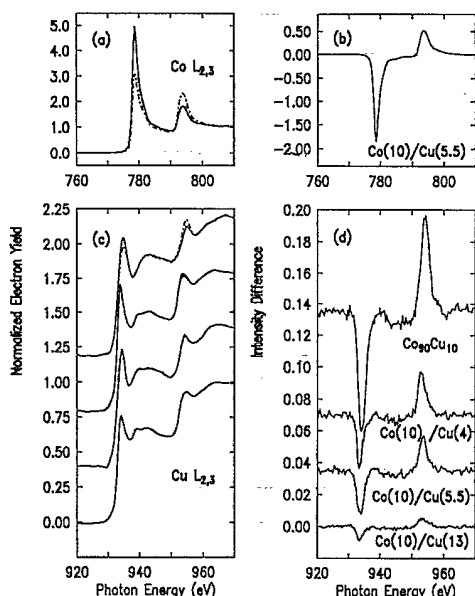


FIG. 1. (a) Co $L_{2,3}$ x-ray absorption spectra for Co(10)/Cu(5.5 Å) recorded with parallel (solid line) and antiparallel (dashed line) alignment of photon spin and sample magnetization directions and (b) the difference spectrum. (c) Cu $L_{2,3}$ spectra and (d) difference spectra for Co₉₀Cu₁₀ and Co/Cu multilayers. Spectra in (c) and (d), respectively, are plotted on the same vertical scale but are offset for clarity.

TABLE I. Experimental and calculated d -shell spin and orbital moments.

Sample	Atom	Experiment		Theory	
		M_S^a (μ_B)	M_L^a (μ_B)	M_S (μ_B)	M_L (μ_B)
Co	Co	1.64	0.14	1.64 ^b	0.14 ^b
				1.65	
Ni	Ni	0.62	0.06	0.64 ^b	0.07 ^b
				0.62	
Co ₉₀ Cu ₁₀	Co	1.50	0.157		
	Cu	0.13	0.005		
Co/Cu(4)	Co	1.71	0.126		
	Cu	0.054	0.0008		
3Co/2Cu	Co			1.64	
	Cu			0.046	
Co/Cu(5.5)	Co	1.71	0.16		
	Cu	0.046	0.0013		
3Co/3Cu	Co			1.64	
	Cu			0.031	
Co/Cu(13)	Co	1.66	0.24		
	Cu	0.013	0.0008		
3Co/7Cu	Co			1.64	
	Cu			0.014	

^a 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_3 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/ y Cu), with $y = 2, 3, 4, 7,$ and 8 . The (3Co/ y Cu) 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 $Co_{90}Cu_{10}$ 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

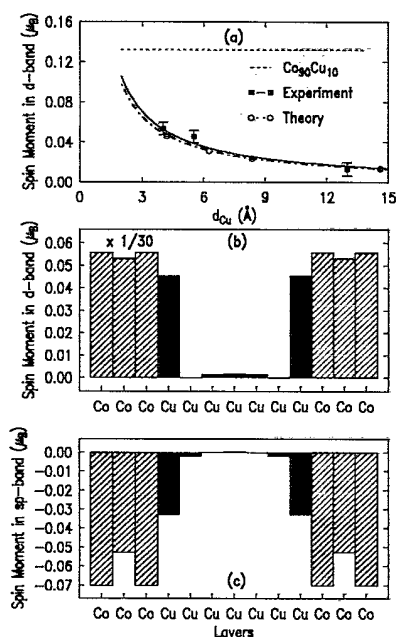


FIG. 2. (a) Experimentally determined average spin moment of the Cu d shell for $Co_{90}Cu_{10}$ (dashed line) and experimental (full squares) and calculated (open circles) average Cu spin moments for various Co/Cu multilayers, as a function of Cu thickness. The solid and dash-dotted lines are fits with a $1/d_{Cu}$ function to the experimental and theoretical data, respectively. (b) Calculated layer-by-layer d -shell spin moments for Co (reduced by a factor of 30) and Cu in a multilayer consisting of three Co layers and seven Cu layers. (c) Same as (b) for the summed s and p spin moments. Note that Co and Cu moments are plotted on the same scale.

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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- [1] S. S. P. Parkin, N. More, and K. P. Roche, *Phys. Rev. Lett.* **64**, 2304 (1990); S. S. P. Parkin, R. Bhadra, and K. P. Roche, *Phys. Rev. Lett.* **66**, 2152 (1991).
- [2] P. Bruno and C. Chappert, *Phys. Rev. B* **67**, 1602 (1991); **46**, 261 (1992).
- [3] D. M. Edwards and J. Mathon, *J. Magn. Magn. Mater.* **93**, 85 (1991); B. A. Jones and C. B. Hanna, *Phys. Rev. Lett.* **71**, 4253 (1993).
- [4] J. E. Ortega and F. J. Himpsel, *Phys. Rev. Lett.* **69**, 844 (1992); J. E. Ortega, F. J. Himpsel, G. J. Mankey and R. F. Willis, *Phys. Rev. B* **47**, 1540 (1993).
- [5] N. B. Brookes, Y. Chang, and P. D. Johnson, *Phys. Rev. Lett.* **67**, 354 (1991).
- [6] K. Garrison, Y. Chang, and P. D. Johnson, *Phys. Rev. Lett.* **71**, 2801 (1993).
- [7] C. Carbone, E. Vescovo, O. Rader, W. Gudat, and W. Eberhardt, *Phys. Rev. Lett.* **71**, 2805 (1993).
- [8] M. van Schilfgaarde and W. A. Harrison, *Phys. Rev. Lett.* **71**, 3870 (1993).
- [9] T. G. Walker, A. W. Pang, H. Hopster, and S. F. Alvarado, *Phys. Rev. Lett.* **69**, 1121 (1992); J. Unguris, R. J. Celotta, and D. T. Pierce, *Phys. Rev. Lett.* **69**, 1125 (1992); Y. U. Idzerda, L. H. Tjeng, H.-J. Lin, C. J. Gutierrez, G. Meigs, and C. T. Chen, *Phys. Rev. B* **48**, 4144 (1993).
- [10] K. Totland, P. Fuchs, J. C. Gröbli, and M. Landolt, *Phys. Rev. Lett.* **70**, 2487 (1993).
- [11] R. Wienke, G. Schütz, and H. Ebert, *J. Appl. Phys.* **69**, 6147 (1991); D. Weller, Y. Wu, J. Stöhr, B. D. Hermsmeier, and M. G. Samant (unpublished).
- [12] Y. Wu, J. Stöhr, B. D. Hermsmeier, M. G. Samant, and D. Weller, *Phys. Rev. Lett.* **69**, 2307 (1992).
- [13] J. Stöhr, *NEXAFS Spectroscopy*, Springer Series in Surface Sciences Vol. 25 (Springer, Heidelberg, 1992).
- [14] B. T. Thole, P. Carra, F. Sette, and G. van der Laan, *Phys. Rev. Lett.* **68**, 1943 (1992); R. Wu, D. Wang, and A. J. Freeman, *Phys. Rev. Lett.* **71**, 3581 (1993).
- [15] P. Carra, B. T. Thole, M. Altarelli, and X. Wang, *Phys. Rev. Lett.* **70**, 694 (1993).
- [16] A. F. Starace *Phys. Rev. B* **5**, 1773 (1972).
- [17] The radial dipole matrix elements for transitions $2p^6 3d^8 4s^1 \rightarrow 2p^5 3d^9 4s^1$ in atomic Co and $2p^6 3d^9 4s^1 \rightarrow 2p^5 3d^{10} 4s^1$ in atomic Ni differ by only about 3% [P. S. Bagus (private communication)].
- [18] P. Söderlind, O. Eriksson, B. Johansson, R. C. Albers, and A. M. Boring, *Phys. Rev. B* **45**, 12911 (1992); O. Eriksson (private communication).
- [19] O. K. Andersen, O. Jepsen, and D. Glözel, in *Highlights of Condensed Matter Theory*, edited by F. Bassani *et al.* (North-Holland, Amsterdam, 1985).
- [20] U. von Barth and L. Hedin, *J. Phys. C* **5**, 1629 (1972).
- [21] The measured $1/d_{Cu}$ 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 any spin polarization because they sample only a restricted region of the Brillouin zone.
- [22] A. Fontaine and G. Schütz (private communication).