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Magnetic moments and Curie temperatures of Ni and Co thin films and coupled trilayers

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We present x-ray magnetic circular dichroism (XMCD) measurements on single layers, bilayers, and trilayers consisting of Ni, Co, and Cu. Using XMCD sum rules, spin and orbital contributions to the total magnetic moment were determined for 2.1 ML Co and 4 ML Ni single layers on Cu(001). In accordance with theory for Co, both spin and orbital moments show an enhancement. In the case of Ni, both spin and orbital moments were reduced. The total magnetic moment of the 4 ML Ni film reduces by a factor of 2 as compared to the Ni bulk. The Curie temperature of both Ni and Co were found to reduce upon capping with Cu. In trilayers, the shape of the Ni magnetization curves were found to be influenced through exchange coupling due to the presence of Co. [S0163-1829(98)00433-0]

I. INTRODUCTION

The reduced symmetry and coordination number of low-dimensional magnetic systems leads to modifications in a variety of phenomena such as complex magnetic ordering, magnetocrystalline anisotropy, and localized electronic states. In the last decade many theoretical studies have been reported, especially involving $3d$ transition-metal (TM) films on magnetic and nonmagnetic substrates.[1–4] Also, as the understanding of the preparation of thick and thin films is increasing, many experimental studies dealing with the different phenomena mentioned above were reported (e.g., Ref. 5).

The x-ray magnetic circular dichroism (XMCD) has proven to be a useful technique to obtain element and site-specific information regarding magnetic moments, Curie temperature (T_C) etc., in a broad range of magnetic systems. Moreover, XMCD measurements using soft x rays (100–1500 eV) give access to $3d$ (for TM) and $4f$ (for rare-earth) states, which are responsible for most of the magnetic properties. Though many studies using XMCD (Refs. 6–9) were reported on thick, multilayer, and capped films on magnetic and nonmagnetic substrates, there are still few *in situ* studies involving single ultrathin films.[10,11] Recent fully relativistic linear muffin-tin orbital (LMTO) calculations[4] involving both spin-orbit coupling and orbital magnetism have shown that, as compared to the bulk values, the orbital moment of Fe, Co, and Ni deposited on Cu(001) is enhanced at the surface. With the exception of Ni on Cu(001), the spin moments also show the same trend. The enhanced orbital moment of Co/Cu(001) has also been confirmed experimentally.[10] However, theoretically predicted values of spin and orbital moments of Co and Ni uncovered ultrathin films on Cu(001) have not been examined separately. The next obvious step in this direction would be to extract information on the magnetic properties of bilayers and trilayers in the thickness range of ultrathin films. In the recent past, studies on bilayer, trilayer, or multilayer systems have shown very interesting magnetic properties, and hence attracted a great deal of attention.[12–32] For example, the exchange coupling across nonmagnetic layers periodically changes sign with increasing thickness of a nonmagnetic cap (in a bilayer) or spacer (in a trilayer or multilayer).[14,15,17,18,20,21,24–27,30,31] Also, interlayer exchange coupling has shown temperature dependence in some trilayer structures.[13,32] Ferromagnetic resonance (FMR) measurements on a series of Co/Ru/Co trilayer films have shown that the exchange field increases with decreasing temperature.[13] Similarly, FMR measurements at low temperatures on Fe/Pd/Fe and Fe/Cu/Fe trilayer structures have reported that the exchange coupling strength at 77 K is almost twice as large as the value at room temperature.[32]

The first part of the present paper describes the determination of the spin (M_S) and orbital (M_L) contributions to the total magnetic moments of ultrathin (2–4 ML) Co and Ni films on Cu(001) by using XMCD sum rules. The values of M_S and M_L are compared with the ones calculated by fully relativistic LMTO calculations. Therefore, in the second part, the shape of the magnetization curves $M_r(T)$ of Ni and Co measured individually by XMCD are reported for bilayers (viz. Ni and Co capped with Cu) and trilayers (viz. Ni capped with Cu and Co on top). The influence of an exchange coupling on the $M_r(T)$ curves and the Curie temperature is discussed in detail.

II. EXPERIMENTAL AND DATA ANALYSIS

All films were grown on a Cu(001) substrate in ultrahigh vacuum conditions. Films were characterized by means of low-energy electron diffraction. The thickness was calibrated by using a quartz microbalance and cross checked using the Ni, Co, and Cu $L_{2,3}$-edges jump ratios as discussed elsewhere.[9] The $L_{2,3}$-edge spectra were recorded in the partial electron yield mode using circularly polarized light at the SX 700 monochromator beamlines at BESSY, the synchrotron facility in Berlin. The XMCD spectra were taken by keeping the helicity of the incident light fixed and reversing the direction of the remanent magnetization by means of a pulse driven electromagnet. The spectra were first corrected for the saturation effects.[9] The degree of circular polarization ($P_C = 0.73$) was determined by using a thick film of Co($\approx$50 ML) on Cu(001), fixing $M_S = 1.55\mu_B$,[7] and applying directly the sum rules. The $P_C = 0.73$ agrees well with the one (0.7 ± 0.1) determined by optical considerations in this energy range. The number of d holes (n_h) was taken as 2.43 for this purpose.[33] The applicability of sum rules for such a thick film is well proven.[7]

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To determine the absolute values of M_L and M_S, a step function with a step height ratio $L_3:L_2$ of 2:1 was fitted. For both edges the inflection point was taken at the resonance maxima. The $L_{2,3}$ absorption spectra are sensitive to both s and d electrons, which means that the $4s$ magnetism certainly affects the XMCD spectra, but the calculation of the sum rules, which include many-body effects, treat only the $2p \rightarrow 3d$ transitions. Therefore, the background XMCD signal between the L_3 and L_2 edges was subtracted to avoid the contribution from the $4s$ electrons whose spin is oppositely polarized to the $3d$ electrons,[2,4–6] and to prevent some overlapping between the L_3 and L_2 contributions. Thereafter, sum rules[34,35] were used to determine values of orbital (M_L) and spin (M_S) moments. The M_L is directly determined from the integrated area, whereas for the determination of M_S, the contribution of the spin magnetic dipole operator (T_Z) is subtracted. This is important, as the contribution from T_Z is negligible only for atoms in cubic symmetry (i.e., ''bulk''). In the case of atoms in noncubic environments such as surfaces, interfaces,[1,3] and low-dimensional systems, T_Z can be very large (0.01 and -0.03 for bulk surfaces of Ni and Co, respectively). In our recent study, we have shown that at low coverages ($\leqslant 4$ ML), the number of d holes (n_h) vary with the thickness of the film.[36] Therefore, the change in the d-hole count was taken into consideration while using a particular thickness.

III. RESULTS AND DISCUSSION

A. Determination of spin and orbital moments: XMCD sum rules versus theory

Figures 1 and 2 show the normalized absorption spectra and the normalized XMCD spectra of 2.1 ML Co and 4.0 ML Ni face-centered-tetragonal (fct) (Ref. 37) thin films on Cu (001), respectively. The remanent magnetization (M_r) was found to be oriented parallel to the surface for both films. These data were taken at 40 K for an incident photon beam parallel to the [100] axis. From the normalized absorption spectra of Co and Ni (upper panels Figs. 1 and 2), the ratio of the spin averaged signal, ie., taking the mean of the absorption signals when the remanent magnetization was almost parallel (dotted) and antiparallel (solid) at 20° grazing incidence to the photon spin, turns out to be 1.6. This number (1.6) is quite close to the ratio of the number of d holes ($n_h^{\mathrm{Co}}/n_h^{\mathrm{Ni}} \approx 2.35/1.25 \approx 1.8$) used for 2.1 ML of Co and 4.0 ML of Ni on Cu(001) in the present analysis. The lower panels of Figs. 1 and 2 show the XMCD difference of the two absorption spectra. As evident from the figures, the difference spectra exhibit a flat base line, confirming the reliability of the data normalization. The M_L and M_S deduced from the spectra using the sum rules are presented in Table I. The value of M_S was determined after subtracting the spin magnetic dipole contribution (T_Z) (Ref. 1) and adding the $4s$, $4p$ contributions to the spin moment (the contribution to the orbital moment can be neglected).[4] This correction (both T_Z and addition of $4s$, $4p$ contributions) in M_S was found to be -1.8% for Co bulk, $+5.5\%$ for 2.1 ML Co on Cu(001) and -10% for 4 ML Ni on Cu(001).[7] Here, it has been assumed that for ultrathin films these contributions are localized mostly at the surface. Therefore theoretically predicted values of bulk surfaces were used for 2.1 ML Co and 4 ML Ni on Cu(001).

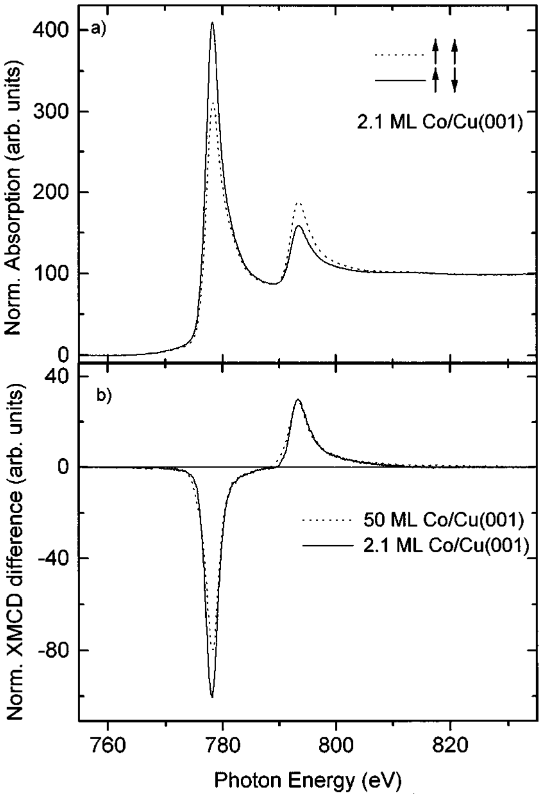

FIG. 1. (a) The normalized absorption spectrum for 2.1 ML Co/Cu(001). The remanent magnetization is parallel (dashed line) or antiparallel (full line) to the photon spin. All data are normalized to the same signal-to-background ratio, which is set to 100 arbitrary units. (b) The difference of the data of (a) as well as for 50 ML Co/Cu(001).

We start with 2.1 ML Co on Cu(001). It is clear from Table I that the theoretically calculated value of M_L for bulk Co is in good agreement ($\leqslant 15\%$) with experiments. If we compare the experimentally determined values for 2.1 ML of Co on Cu(001) with the one for the Co bulk, we find that both M_S and M_L are enhanced compared to the corresponding bulk values. An enhancement of M_L is much more pronounced (by a factor of 2) than the one of M_S, confirming the outcome of our earlier study.[10] However, previously,[10] only the ratio M_L/M_S was determined to avoid the complications arising through some parameters, e.g., n_h. Our recent work[36] enabled us to use much more realistic values of n_h and, hence, making the separate determination of M_L and M_S possible. The reasons for an enhancement in M_L for thin films were discussed in detail in earlier studies.[4,10] The experimentally obtained values of M_L, M_S, and M_L/M_S lie between the theoretical values for 2 and 1 ML Co on Cu(001), in fact closer to 1 ML on Cu(001). An inaccuracy in the determination of n_h, P_C and the normalization factors cannot be held responsible for this observation, as M_L/M_S, which is independent of these parameters, also shows the same trend. The possible explanation for this observation is the hybridization at the Co/Cu interface. This reduces the effective thickness of the film, contributing to a ferromag-

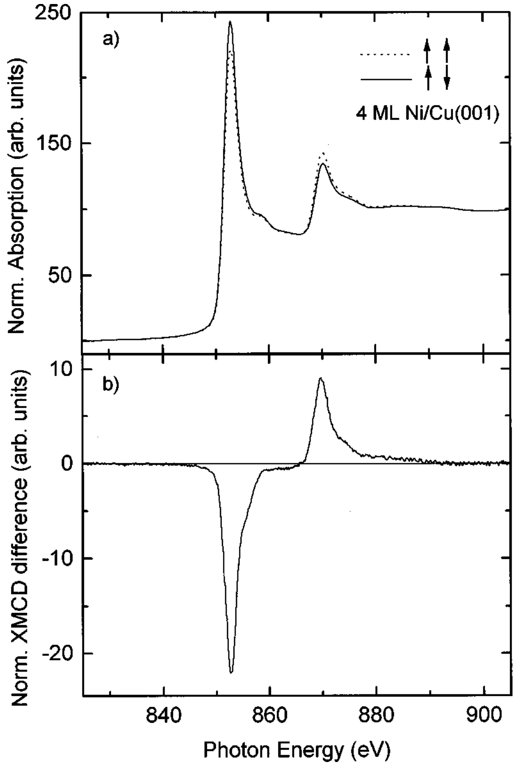

FIG. 2. (a) The normalized absorption spectrum for 4.0 ML Ni/Cu(001). The remanent magnetization is parallel (dashed line) or antiparallel (full line) to the photon spin. All data are normalized to the same signal-to-background ratio, which is set to 100 arbitrary units. (b) The difference of the data of (a).

netic response, and therefore one expects the values to be closer to the theoretically calculated ones of 1 ML Co on Cu(001).

Now, we turn our attention to Ni on Cu(001). In Table I, magnetic moments determined by applying XMCD sum rules for 4 and 4.3 ML (spectra not shown) single layers on Cu(001) are presented. For the 4.3 ML Ni film, spectra were recorded at 160 K. The M_L and M_S given in Table I were obtained by extrapolating the data to $T=0$ K by using the temperature dependence of the XMCD difference of a 4 ML Ni film on Cu(001).[38] The reproducibility of the values of M_L and M_S for both Ni films shows the reliability of our data analysis. The calculated values for 1 ML Ni on Cu(001) and the Ni bulk are also listed. The experimentally determined ratio M_L/M_S for both films is enhanced by 45% and decreased by 25%, as compared to the calculated values for the Ni bulk and 1 ML Ni on Cu(001), respectively. Examining the values of M_L and M_S separately reveals that both are reduced in comparison to the bulk values. The total magnetic moment (M^{tot}) of a 4 ML Ni film on Cu(001) is reduced by more than a factor of 2 in comparison to the Ni bulk. Such a large reduction in the total magnetic moment cannot be due to the existence of some magnetic dead layers, since a clear ferromagnetic response has been found down to 1.6 ML Ni on Cu(001).[38] Recently, XMCD and polarized neutron reflection studies on Cu/Ni/Cu/Si(001) structures also reported the $M^{\text{tot}}<0.3\mu_B$ for $\sim$30 ML-thick Ni films, a reduction by more than a factor of 2 as compared to the bulk Ni value. It was attributed to strain in the Ni lattice.[39] A reduction of the spin moment for 1 ML Ni on Cu(001) as compared to the bulk has been predicted by theory due to Ni-*d*–Cu-*d* hybridization.[4] In the present analysis we do see an appreciable reduction in M_S as compared to the bulk, hence confirming the theoretical prediction and clearly showing that, even at 4 ML thickness, Ni does not behave bulklike. However, contrary to the theory, M_L was also found to be reduced. Experimental values of M_S and M_L presented in the table cannot be compared directly to the theoretical values of 1 ML Ni on Cu(001), as thicknesses of the films are different in the two cases. However, our preliminary XMCD analysis of even thinner (<4 ML) Ni films shows a further reduction in M^{tot}. A reduction in M^{tot} of Ni ultrathin films (4–6 ML) as compared to the bulk has also been observed by FMR measurements.[40]

B. Magnetic response of Ni in bilayers and trilayers

Figure 3 shows $M_r(T)$ curves of 4.3 ML Ni and 1.9 ML

TABLE I. Experimentally determined and calculated values of M_S, M_L and their ratio (M_L/M_S) for Ni and Co films on Cu(001). In the experimental section the values of M_S were corrected for magnetic dipole (T_Z) and s, p contributions.

Experimental	M_S (μ_B)	M_L (μ_B)	M_{tot} (μ_B)	M_L/M_S
Co(2.1 ML)/Cu(001)	1.77±0.1	0.24±0.05	2.02±0.15	0.136±0.03
Ni(4.0 ML)/Cu(001)	0.24±0.1	0.035±0.01	0.275±0.1	0.145±0.03
Ni(4.3 ML)/Cu(001)	0.25±0.1	0.036±0.01	0.29±0.1	0.144±0.03
Co(bulk)/Cu(001)	1.55[a]	0.142±0.01	1.692±0.01	0.092±0.01
Theory				
Co(1.0 ML)/Cu(001)[b]	1.85	0.261	2.11	0.141
Co(2.0 ML)/Cu(001)[c]	1.73	0.188	1.918	0.107
Co(bulk)[b]	1.63	0.123	1.753	0.075
Ni(1.0 ML)/Cu(001)[b]	0.45	0.087	0.54	0.193
Ni(bulk)[b]	0.58	0.058	0.638	0.100

[a] Reference 7.
[b] Reference 4.
[c] Reference 41.

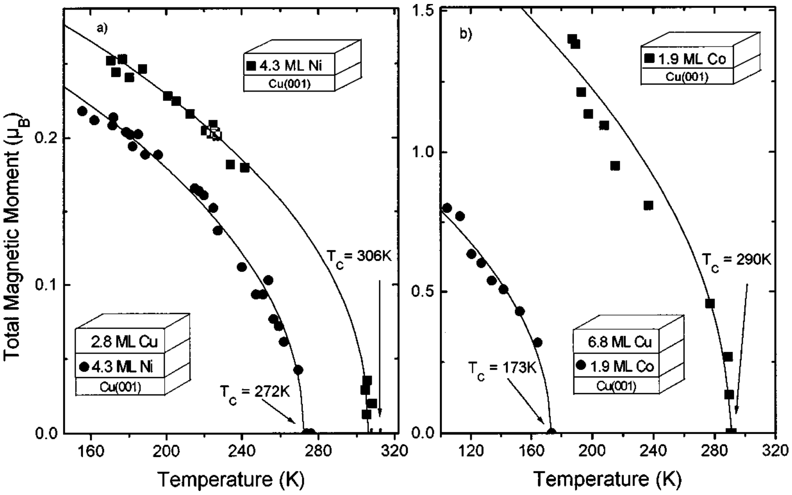

FIG. 3. (a) The $M_r(T)$ curves for 4.3 ML Ni/Cu(001) and (b) 1.9 ML Co/Cu(001) before and after capping with Cu. The temperature where the XMCD difference vanishes can be used to determine the critical temperature T_C. A clear reduction in T_C can be seen on capping. A power-law function At^x, displayed in both (a) and (b), was used to improve the guide to the eye lines.

Co films capped with 2.8 ML and 6.8 ML Cu cap layers. For a better quantitative comparison, in contrast to previous publications, the y axis has been calibrated in units of μ_B. For this purpose all experimental data in the $M_r(T)$ curves were rescaled for 100% circular polarization. The absolute calibration for μ_B was done by applying the sum rules. All curves were fitted with a power law of the type At^x, where $t = (T_C - T)/T_C$. This appears to be a reasonable analytical function in analogy to critical scaling. It can be seen from Fig. 3 that in the presence of Cu cap layers, the T_C of both Ni (T_C^{Ni}) and Co (T_C^{Co}) are lowered by 34 K and 117 K, respectively. The values of the prefactor A and the critical exponent x will be discussed in a separate paper. Here, we use the power law only as a better replacement for the guide to the eye lines. The reason for the lowering of T_C has already been discussed and attributed to increasing overlap of spin-up and spin-down states and hybridization of Co and Ni d states with Cu d and sp states.[28] In an earlier investigation,[42] on capping the Ni(111) with Cu, a strong reduction in the magnetic hyperfine field at the interface was analyzed on the basis that the T_C stays the same. However, the present study clearly demonstrates reduction of T_C on capping.

Figure 4 shows $M_r(T)$ curves of both Ni and Co for two different trilayers. One of them [shown in Fig. 4(a)] is obtained by evaporating, consecutively, 2.0 ML Co on the bilayer shown in Fig. 3(a). The element and site specificity of the XMCD technique allows us to treat Ni and Co independently in a trilayer. Once again, all curves were fitted by using a power law as described above. From Fig. 4 it is clear that (i) in both trilayers, we obtain two different $M_r(T)$ curves for Ni and Co, showing the vanishing of the magnetization (within the noise level) at different temperatures, (ii) if one compares Figs. 3(a) and 4(a), it is clear that for Ni, as compared to the bilayer, the temperature at which magnetization vanishes is enhanced by approximately 40 K. The second observation is also true for the other trilayer shown in Fig. 4(b), though the corresponding data for the bilayer are not shown.

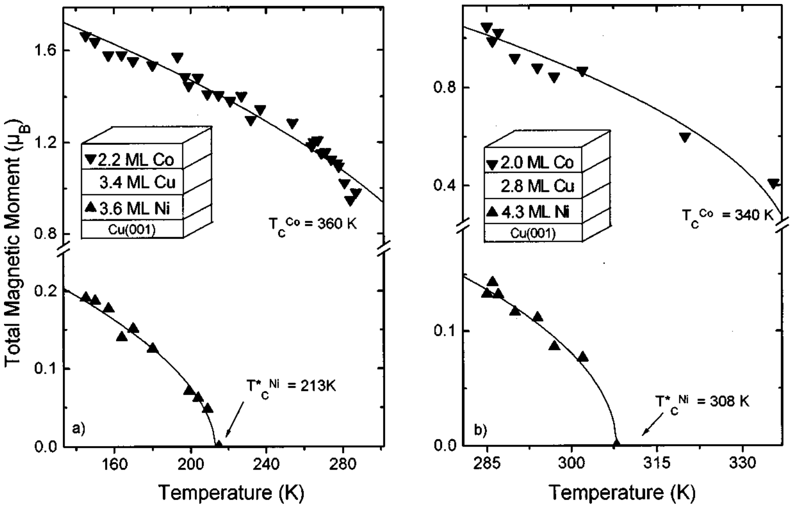

FIG. 4. The $M_r(T)$ curves of Ni and Co for the two trilayers. The trilayer shown in (a) is obtained by evaporating consecutively 2.0 ML Co on the bilayer shown in Fig. 3(a). In both cases, two magnetization curves vanishing at different temperatures were obtained. An asterisk (*) has been used for the temperature at which the magnetization of Ni vanishes to distinguish it from the true phase transition. A power-law function At^x, displayed in both (a) and (b), was used to improve the guide to the eye lines.

The first observation compels one to think if in the present trilayers, consisting of two ferromagnetic elements (Ni and Co) separated by a nonmagnetic spacer, i.e., Cu, one realizes two distinct phase transitions. To find an explanation, we first refer to another experimental study and consequent theoretical work. Experimental data on FeF_2 (m layers)/CoF_2 (n layers) (Ref. 22) superlattices display either two or one magnetic transitions depending on the composition m/n. To explain this experimental observation, it has been suggested theoretically[29] that increasing the thickness of each material will acquire response characteristics of its bulk form and therefore the superlattice will behave as a collection of two bulk materials rather than a single new material. In this case, the material having a higher T_C will show a *true* phase transition. However, in a narrow region of temperature near the T_C of second material (i.e., the one with a lower T_C) the structure exhibits a strongly enhanced response to an external field resulting in *near singularities* in the thermodynamical properties. But, one does not realize two phase transitions, only the material having the lower T_C tends to order spontaneously near its T_C. Another theoretical study[23] suggested that in addition to the relative thicknesses of the components constituting a superlattice, weak or strong interface coupling leads to two or one transitions. With this background we try to understand our experimental results. In both trilayers, Ni films are thicker than Co films. However, the presence of Cu as a spacer limits the influence of Co (having a higher magnetic moment) on Ni, and the exchange coupling due to the presence of Co affects only a few layers of the Ni film. This interlayer exchange is only an interface effect.[20] Therefore, one does see the tendency of Ni to order near a particular temperature, e.g., around 308 K in the case of the trilayer shown in Fig. 4(a). However, as discussed earlier, this is not the true magnetic phase transition and, hence, is shown as $T_C^{*\mathrm{Ni}}$ in the figures. The value of $T_C^{*\mathrm{Ni}}$ depends on the spacer (Cu) thickness as it changes the nature of coupling.[14] The true T_C is the high-temperature transition corresponding to T_C^{Co} at approximately 340–360 K [that corresponds to T_C^{Co} of 2.0–2.2 ML Co single layer on Cu(001)] in the two trilayers. At this temperature, Ni is in a paramagnetic disordered phase and does not contribute to the ferromagnetic response. We have mentioned in our earlier paper[28] that for a Ni/Co bilayer, i.e., in the absence of the Cu spacer, one indeed gets only one $M_r(T)$ curve.

To see the influence of the exchange coupling on the shape of the $M_r(T)$ curve more clearly, and also to understand the second observation, i.e., enhancement in T_C^*, we measured the $M_r(T)$ curves for another trilayer to a much lower temperature (30 K). The $M_r(T)$ curves for Ni in a bilayer and in a trilayer are shown in Fig. 5. Once again, all observations (i) and (ii) described above can be seen in Fig. 5. The T_C^{Ni} in the case of the bilayer was 275 K. On putting Co on top, the temperature at which magnetization vanishes for Ni ($T_C^{*\mathrm{Ni}}$) was found to be 312 K. Figures 4 and 5 suggest that the shape of the $M_r(T)$ curve of Ni not only changes under the influence of exchange coupling, but also depends on the Co and spacer (Cu) thicknesses. Also, the data down to 30 K show that the difference between the $M_r(T)$ curves of bilayers and trilayers are not the same at different temperatures. It has been found experimentally that the coupling strength can be significantly different between room temperature and low temperature.[13,32] For the Fe/Ta/Fe system, it has been shown[12] that the transferred field has a profound influence on the temperature dependence of the magnetization curve in the two-dimensional overlayer film. Also, a zero-field Mössbauer spectroscopy of Fe(110)/Ag(111) multilayer structures reported modifications of the spin-wave spectrum showing the temperature dependence due to coupling.[43] In the light of these experimental results, we can qualitatively understand the $M_r(T)$ curves shown in Fig. 5. The increase in the transition temperature from T_C^{Ni} to $T_C^{*\mathrm{Ni}}$ can be understood from the fact that the transition that occurs in the presence of exchange coupling has a finite effect even at $T > T_C$. This results in the observed shift of transition to the nonmagnetic state.

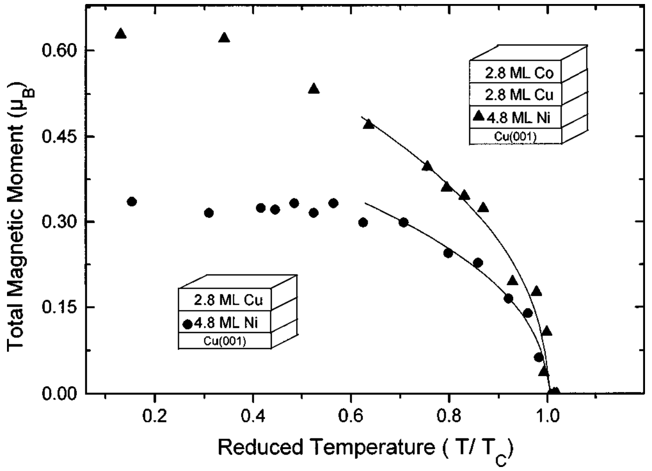

FIG. 5. The $M_r(T)$ curves of Ni in the bilayer and trilayer as a function of reduced temperature. The data were taken down to 30 K.

IV. CONCLUSIONS

The present XMCD experiments show a clear enhancement in the orbital and spin moments of Co thin films (2 ML) on Cu (001), in accordance with the theoretical predictions. For a 4 ML Ni film, compared to the bulk, both spin and orbital moments were found to be reduced. The total magnetic moment of the Ni film was found to decrease almost by a factor of 2 as compared to the Ni bulk. The agreement between experiments and theory may be improved if hybridization with the substrate can be taken into consideration in the sum rules. In bilayers, capping with Cu was found to reduce the T_C of both Ni and Co. We discussed the measurement of two element-specific magnetization curves for Ni and Co and have shown the influence of exchange coupling on the shape of Ni $M_r(T)$ in trilayers. The exchange coupling was found to be temperature dependent and its presence was found to be responsible for a shift of the $M_r(T)$ curves near the T_C of Ni.

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