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Oscillations of the Curie temperature and interlayer exchange coupling in magnetic trilayers

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The onset of long-range magnetic order in exchange-coupled epitaxial Co/Cu/Ni trilayers, 2–4 monolayers (ML) each, on Cu(001) is studied by element-specific x-ray magnetic circular dichroism between 30 and 300 K in ultrahigh vacuum. Oscillations of the enhancement of the ordering temperature of Ni (ΔT_{Ni}) by more than 40 K are measured as a function of interlayer exchange interaction by varying the Cu(001) spacer thickness. Below a Cu thickness of 2.3 ML antiferromagnetic coupling is measured. The period, phase, and amplitude of the ΔT_{Ni} oscillations are in excellent agreement with the theoretical prediction for the short- and long-period oscillations of the interlayer exchange coupling. [S0163-1829(99)50306-8]

The discovery of oscillatory exchange coupling between two ferromagnetic layers separated by a nonmagnetic spacer material has attracted a lot of interest over the last several years.[1] Theoretical approaches based on the Ruderman-Kittel-Kasuya-Yosida (RKKY) model or quantum well states have been able to describe the experimentally observed short- and long-period oscillations of the interlayer exchange coupling $J_{\mathrm{inter}}(d)$ as a function of the spacer thickness d.[2,3]

Most of the attention has focused on the understanding of giant magnetoresistance and on the dependence of the phase, amplitude, and periodicity of J_{inter} on the thickness and material of the spacer and ferromagnetic layers and the orientation of the substrate. Very little work has addressed the effect which the oscillatory exchange might have on the Curie temperature of the ferromagnetic layer. As already pointed out by Bayreuther's group,[4] simple mean-field theory would predict an oscillation of the Curie temperature which correlates with the exchange coupling. Evidence for this was found experimentally in polycrystalline Ni/Au multilayers.[4] In that experiment a set of different samples with nominally the same thickness of the magnetic layer and a variable spacer thickness was used. However, the reported uncertainty in the magnetic layer thickness could lead to T_C variations due to finite-size effects[5] similar in magnitude to the observed T_C oscillation amplitude.

On the contrary, epitaxial trilayers with two different ferromagnetic layers (e.g., Co and Ni) having separate ordering temperatures are ideal prototype systems for illustrating the relation between J_{inter} and T_C: We recently could demonstrate by means of the element specificity of the x-ray magnetic circular dichroism (XMCD) technique that the ordering temperature T^*_{Ni} of Ni in Co/Cu/Ni/Cu(001) trilayers is increased by ΔT_{Ni} compared to T_C of Ni in Cu/Ni/Cu(001) due to the presence of Co by up to 40 K. Despite the strong interlayer coupling that one expects for very thin spacers, the Co and Ni magnetizations still vanish at different temperatures.[6] Such a behavior could be interpreted as two different ordering temperatures of the exchange coupled layers, T_C of Co, the Curie temperature of the whole system, and T^*_{Ni}, the ordering temperature of the Ni layer.[6,7] Such an indirectly coupled system must be distinguished from directly coupled ferromagnets which have been discussed theoretically.[7] This has triggered our interest to investigate the following questions: (i) Is there an oscillation of ΔT_{Ni} with the spacer thickness d_{Cu} and can it be correlated with theoretical models of the oscillatory interlayer coupling? (ii) Is it possible to observe antiferromagnetic coupling for magnetic trilayers with Cu(001) spacer thickness in the range 2–4 monolayers (ML) and how does antiferromagnetic (AMF) versus ferromagnetic (FM) interlayer coupling shift the Curie temperatures of the ferromagnetic layer?

The latter question goes beyond the specific interest for the relation between J_{inter} and T_C. Theoretical works for noble-metal spacers based on the RKKY model have predicted two oscillations of the interlayer coupling with the spacer thickness reflecting the topological properties of the spacer Fermi surface:[2]

$$J_{\mathrm{inter}}(d)=\frac{1}{d^2}\{A_1\sin(2\pi d/\Lambda_1+\Phi_1)+A_2\sin(2\pi d/\Lambda_2+\Phi_2)\}. \tag{1}$$

The earlier multilayer literature has reported only the longer-period oscillations of J_{inter}.[8] The short-period oscillations were recorded later on in high-quality epitaxial trilayers with Cr (Ref. 9) or Cu(001) (Ref. 10) spacers. Moreover, it is only for few trilayers that short-period oscillations and AFM coupling have been observed for spacers thinner than 3–4 ML, e.g., for Fe/Au/Fe.[11] For the case of Cu(001) spacers a systematic spin-polarized scanning electron microscopy (spin-SEM) study on M/Cu/Co trilayers (M=Fe,Co,Ni) (Ref. 12) has revealed both short- and long-period oscillations in good agreement with theoretical values of $\Lambda_1=2.56\,\mathrm{ML}$ (1 ML $=0.18$ nm) and $\Lambda_2=5.88\,\mathrm{ML}$, respectively.[2] The phases and the amplitude ratio A_1/A_2 have been found to depend critically on sample quality and ferromagnetic layer thickness. For the Ni/Cu/Co/Cu(001) trilayer an amplitude ratio of $A_1/A_2=1.3\pm0.5$ was observed.[12] However, no oscillation or AFM coupling was ever found for Cu(001) spacers thinner than 5 ML. This had been attributed to the existence of pinholes[13] since the growth of Co on Cu(001) (Refs. 14 and 15) is known to be problematic due to the segregation of Cu to the top for the above system.

In this work we provide experimental evidence for an AFM interlayer coupling in trilayers with Cu(001) spacers thinner than 5 ML. We measure the enhancement of the element-specific ordering temperature ΔT_{Ni} which is found

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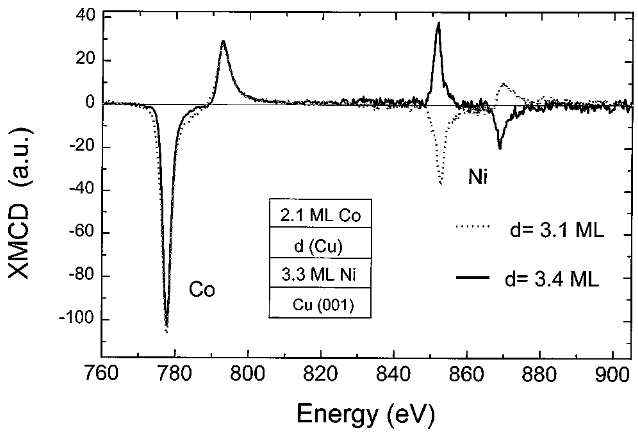

FIG. 1. XMCD spectra for Co/Cu/Ni/Cu(001) trilayers with different Cu spacer thickness d. Note the change in the sign of the Ni spectrum (above 840 eV) clearly showing AFM coupling for $d=3.4$ ML.

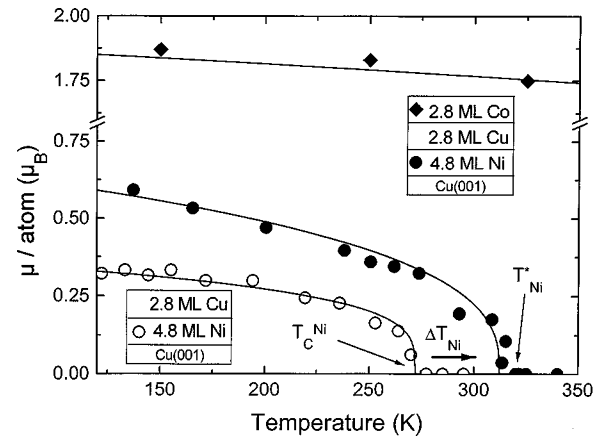

FIG. 2. $M_r(T)$ for Ni and Co vanishing at different temperatures. The lines are guides to the eyes. The y axis is calibrated in μ_B as it has been explained in Ref. 16.

to oscillate with double the periodicity of $J_{\mathrm{inter}}(d)$ as given by Eq. (1). This is to our knowledge the first unambiguous experimental proof that the Curie temperature of a ferromagnetic layer is enhanced for both ferro- and antiferromagnetic coupling. Furthermore, we will show that the oscillatory ΔT_{Ni} is in excellent agreement with Eq. (1) by scaling J_{inter} to ΔT_{Ni} and using the theoretically predicted phases ($\Phi_1=0.5\pi$, $\Phi_2=\pi$) and periodicities ($\Lambda_1=2.56$ ML, $\Lambda_2=5.88$ ML) (Ref. 2) and the experimental amplitude ratio of 1.3 (Ref. 12) without any other fitting parameter.

The Co/Cu/Ni trilayers were grown on a Cu(001) single crystal under ultrahigh vacuum (UHV) conditions, as it is described elsewhere.[6,16] A precise thickness determination has been achieved by precalibrated quartz crystal microbalance, MEED oscillations, and by using the Ni, Co, and Cu $L_{2,3}$-edges jump ratios.[17] Initially, Ni layers in the range from 3 to 4.8 ML with an accuracy of 0.1 ML were deposited onto Cu(001) providing a pseudomorphically crystalline perfect buffer[18,19] for the subsequent deposition of Cu and Co. Moreover, from our results on the interlayer coupling, we conclude that first depositing Ni on the Cu(001) substrate gives trilayers with fewer problems than depositing Co first.

The trilayers were measured by means of the XMCD technique.[20] This technique has the advantage of measuring the magnetization with element specificity. Separate signals for Ni and Co are recorded. $L_{2,3}$-edge spectra were measured in the partial electron yield mode using circularly polarized light at the SX 700 monochromator beamlines at BESSY, the synchrotron facility at 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 temperature range of these measurements varied between 30 and 300 K.

Figure 1 depicts typical XMCD spectra for a trilayer at $T=63$ K with a Cu spacer of $d=3.1$ ML and $d=3.4$ ML. The lower-energy spectrum corresponds to Co, the higher-energy spectrum to Ni. It is evident that signals at the $L_{2,3}$-edges of Ni change sign. Contrary to a spacer with $d=3.1$ ML with parallel alignment, antiparallel alignment is observed for $d=3.4$ ML. It is the first time, to our knowledge, that an AFM interlayer coupling is recorded in trilayers with such thin Cu(001) spacers. The sign of the interlayer coupling remains the same at all temperatures in agreement with theoretical predictions and previous experiments.[21,22]

One should note that in weakly coupled layers with large uniaxial anisotropies[22] a parallel alignment of the magnetizations may be present in spite of an AFM interlayer coupling, making ambiguous the determination of the coupling sign from the remanent-magnetization configuration. However, this is not the case here since (i) the spacers are only 2–4 ML thick and J_{inter} attains very large values (see, e.g., Ref. 2), and (ii) the in-plane fourfold anisotropy is usually quite small to compete with J_{inter} in the magnetization reversal processes.

Another important finding is the AFM coupling for different samples (e.g., at $d_{\mathrm{Cu}}=2.2$ and 3.4 ML) which shows the high quality of our trilayers. No evidence for pinholes in these structures is detected for our trilayers since Co and Ni enter the paramagnetic phase in the trilayers at separate temperatures. Therefore a direct exchange coupling between them (through pinholes) is not likely. Finally, the unambiguous proof for ideal-like spacer layers is the observation of an antiferromagnetic interlayer coupling in Fig. 1.

In Fig. 2 the element-specific remanent-magnetization data as a function of temperature are shown for Ni before (open) and after (solid circles) the evaporation of the Co layer (diamonds) in the trilayer Co (2.8 ML)/Cu (2.8 ML)/Ni (4.8 ML)/Cu(001). The Curie temperature of Ni is lower than the bulk value due to finite-size effects, and it is in good agreement with our previous results for ultrathin Ni/Cu(001) films.[5] The interlayer coupling in this sample is of FM character. From Fig. 2 the following may be clearly observed: (i) Separate magnetization curves for Co and Ni in the trilayer are recorded which vanish at different temperatures. (ii) The temperature T^*_{Ni} where the remanent magnetization of Ni vanishes is different (higher) in the trilayer than in the Cu/Ni bilayer. According to simple mean-field theory, the Curie temperature should increase in proportion to the extra energy added to the magnetic system due to the interlayer coupling, irrespective of its anti- or ferromagnetic character. In our case, we deal with asymmetric trilayers where the two magnetic layers have separate bare Curie temperatures. When they are brought to proximity they interact through the Cu spacer. In a strict thermodynamic sense the system should exhibit only one true phase transition at the temperature

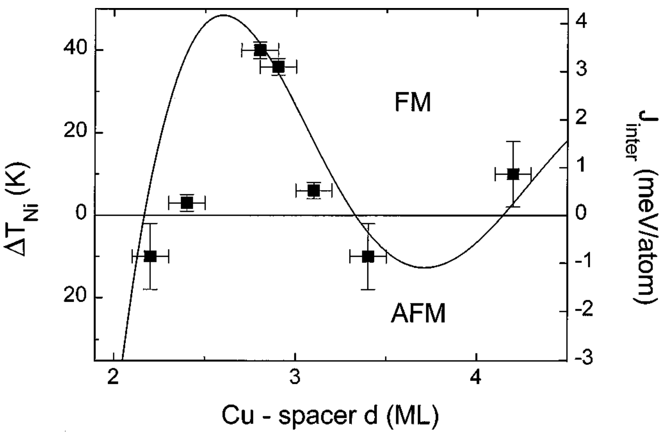

FIG. 3. Shift of the ordering temperature of Ni ΔT_{Ni} as a function of the Cu spacer thickness (2–4 ML) for various Co/Cu/Ni/Cu(001) trilayers. The right y axis is in coupling units as explained in the text.

where the higher T_C element (here it is Co) is placed.[7] However, Fig. 2 reveals that Ni orders at a temperature T^*_{Ni} where its $M_r(T)$ curve attains sizable values.[6]

To check the relation of the shift ΔT_{Ni} to the strength of the interlayer coupling we plot ΔT_{Ni} as a function of spacer thickness d in Fig. 3 (solid symbols). The shift of T^*_{Ni} is always to higher temperatures for stronger exchange, irrespective of the sign of the coupling. To account for the different signs which we unambiguously know from the XMCD spectra, we plot the positive shift for FM coupled layers upward on the y axis and the positive shift for AFM coupled layers downward. The maximum variation with d_{Cu} is about 40 K. In the simplest mean-field model, ΔT_{Ni} is due to J_{inter}.[4,6] In this picture one can derive the magnitude of J_{inter} from the simple relation

$$z^* J_{inter} = k_B \Delta T_{Ni}. \tag{2}$$

Its sign is determined by the XMCD spectra directly. The values of J_{inter} are given in meV/ atom on the right axis of Fig. 3. They are reasonable for magnetic layers separated by very thin spacers.[6] This implies that the ''effective coordination number'' z^* equals 1, which is reasonable because one assumes that the magnetizations as a whole contribute to the interlayer coupling.[6] The solid line in Fig. 3 is calculated according to Eq. (1) with the theoretical parameters $\Lambda_1 = 2.56$ ML, $\Lambda_2 = 5.88$ ML, $\Phi_1 = \pi/2$, and $\Phi_2 = \pi$.[2] The experimental amplitude ratio[12] $A_1/A_2 = 1.3$ is used. Note that no adjustable parameter was used in this calculation.

In conclusion, epitaxial Co (2–2.8 ML)/Cu (2.2–4.5 ML)/Ni (3–4.8 ML)/Cu(001) were characterized via the XMCD technique between 30 and 300 K. XMCD spectra determine directly the sign of the interlayer exchange coupling (antiferromagnetic or ferromagnetic). In our study an antiferromagnetic interlayer coupling is experimentally verified for Cu(001) spacers thinner than 5 ML. The remanent magnetization curves of Ni and Co vanish at different temperatures. The enhancement of the ordering temperature ΔT_{Ni} is a measure for J_{inter} and it oscillates with double its periodicity. The variation of J_{inter} perfectly agrees with the theoretically predicted short- and long-period oscillations.

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