Two Susceptibility Maxima and Element Specific Magnetizations in Indirectly Coupled Ferromagnetic Layers

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The ac susceptibility χ of ultrathin Co and Ni layers coupled indirectly across a Cu spacer is measured as a function of temperature. Depending on the strength of the interlayer exchange interaction a single χ maximum (singularity) or two χ maxima (one singularity, one resonantlike signal) appear near the respective Curie temperatures of bare layers. Using the element specificity of x-ray magnetic circular dichroism two separate temperature-dependent magnetization curves for Co and Ni are recorded, vanishing at different temperatures. These results support theoretical predictions for the onset of long-range order in model superlattices. [S0031-9007(98)07090-2]

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The magnetic properties of metallic multilayers have shown a rich variety of new physical phenomena [1]. Most of the studies are focused on interlayer coupling, giant magnetoresistance, and quantum well effects. However, very little work has been directed to a question of fundamental importance, that is, how the interlayer coupling effects the basic magnetic observables, these are the Curie temperature T_C, the sublayer magnetizations M_i, and the susceptibility χ of the ferromagnetic layers. We choose a trilayer prototype system as shown schematically in Fig. 1 consisting of two ferromagnetic sublayers separated by a nonmagnetic spacer mediating the interlayer coupling to study $\chi(T)$, $M_i(T)$, and the behavior of T_C. The strength of the interlayer coupling $J_{\rm inter}$ can be tuned by the spacer thickness, whereas the intrinsic magnetic properties, M_i and T_C for example, are determined by the thickness d and the type of element. Co and Ni were selected and their thicknesses were chosen to be in a convenient temperature range for a precise determination of T_C [2–4].

Before discussing the results let us ask the following question: Suppose the intrinsic T_C's of the two uncoupled magnetic sublayers, that is $T_C^{\rm high}$ (for Co) and $T_C^{\rm low}$ (for Ni) are different, will the trilayer show two separate T_C's or will the system undergo one phase transition? Surprisingly enough, this fundamental question has been addressed very little in recent literature. Within Ginzburg-Landau theory, Wang and Mills [5] studied the onset of long-range order in a model superlattice consisting of two components, which are both either ferromagnets or antiferromagnets presenting two different ordering temperatures in the uncoupled case ($T_C^{\rm high}$ and $T_C^{\rm low}$). The magnetic susceptibility is calculated and presents two maxima: One "true singularity" [5] at the thermodynamic phase transition near the higher ordering temperature $T_C^{\rm high}$ and a resonantlike anomaly near $T_C^{\rm low}$. The order parameters, that is the magnetization of the two constituents, is also calculated and is nonzero in the temperature range between the χ maxima. They also expect this effect to be present in indirectly coupled ferromagnetic layers and stress the importance of susceptibility measurements to clarify the temperature-dependent behavior, but no experiment is available to date. Measurements of the thermal expansion coefficient [6] and the specific heat [7] of antiferromagnetic multilayers consisting of two components are in good agreement with the theoretical description also given by other authors [8,9]. Recent measurements [10] of the total magnetization (nonelement resolved) of thick Co/Ni multilayers show a weak kink at a specific temperature, interpreted as the ordering temperature of the Ni sublayers. The surface magnetization of an ultrathin film of Fe with a Ta spacer on a ferromagnetic substrate was investigated, and an effect on the ordering temperature of the film has been seen with a varying spacer thickness [11]. These results on ferromagnets are based on the remanent magnetization averaged over the sample volume or detecting the remanent magnetization of the surface only. Since $M(T)$ is a vanishing quantity at T_C and is effected by residual fields, an unambiguous determination of T_C from kinks in $M(T)$ is difficult. The susceptibility χ is the preferable quantity to be investigated. The first element specific χ measurements were performed for a directly coupled Co/Ni/Cu(001) double layer showing one T_C [12].

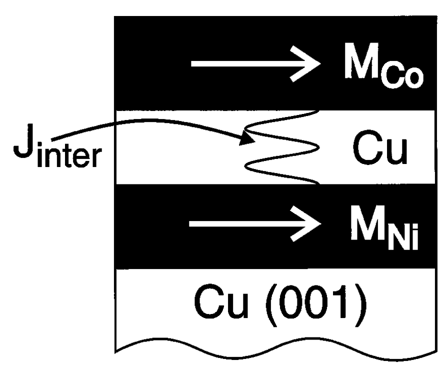

FIG. 1. The investigated system schematically: An epitaxial trilayer on Cu(001) substrate consisting of in-plane magnetized $M_{\rm Ni}$ and $M_{\rm Co}$ separated by a Cu spacer layer mediating the interlayer coupling $J_{\rm inter}$.

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In our experimental work we study the onset of long-range order in a prototype system: Two ultrathin ferromagnetic films, namely Co and Ni, which are separated by a nonmagnetic Cu spacer layer. This offers the opportunity to tune the exchange interaction between both magnetic constituents without changing the thickness of the magnetic layers. Because of the presence of the interlayer exchange interaction $J_{\rm inter}$ this material is expected to undergo a single phase transition, but the spatial variation of the order parameters in the Co and Ni layers may differ from the directly coupled case due to the separation by nonmagnetic Cu. We measure the magnetic susceptibility that is the response function of the whole system. As was predicted in Ref. [5] the susceptibility shows one signal near $T_C^{\rm high}$ and a weaker maximum at a temperature $T^* > T_C^{\rm low}$. In addition, the order parameters of the Ni and Co sublayers are recorded separately by virtue of the element specificity of x-ray magnetic circular dichroism (XMCD). The temperature dependent magnetization of Ni and Co sublayers are vanishing at two different temperatures. Thus it is here for the first time that all the fundamental quantities calculated in Ref. [5], that is $\chi(T)$ and element-resolved $M_i(T)$, are experimentally investigated, and a new insight is gained into the problem of long-range order in indirectly coupled layers.

Depending on the interlayer interaction, three coupling regimes must be distinguished in ultrathin trilayers with increasing spacer thickness: (i) Two ultrathin ferromagnetic films coupled directly without a spacer show one common phase transition [12,13], similar to NiCo alloy. (ii) Two films separated by an ultrathin spacer layer. Such samples are investigated in the present work. In this case the sublayer magnetizations vanish at different temperatures: The higher one is at $T_C^{\rm high}$ and the lower one at T^* appears slightly higher than $T_C^{\rm low}$ due to the additional coupling energy of the interlayer exchange. (iii) A thick spacer layer yields completely decoupled sublayer magnetizations with their intrinsic Curie temperatures as in a powder of Co and Ni with sufficiently large grains.

The investigated system may be phenomenologically described by the following: In the mean-field model the interaction responsible for the T_C of a ferromagnet is the exchange energy $E_{\rm intra}$ for two neighboring spins $\mathbf{S}_i$, $\mathbf{S}_j$:

$$E_{\rm intra} = -2J_{\rm intra}\mathbf{S}_i \cdot \mathbf{S}_j \, . \tag{1}$$

In a trilayer (Fig. 1) the interlayer exchange energy $E_{\rm inter}$ is given by

$$E_{\rm inter} = -J_{\rm inter}\frac{\mathbf{M}_{\rm Ni}\cdot\mathbf{M}_{\rm Co}}{M_{\rm Ni}M_{\rm Co}} \, . \tag{2}$$

In a first approximation we can calculate T^* by

$$T^* = \frac{1}{k_B}\left(\frac{2}{3}\,J_{\rm intra}S(S+1)z + J_{\rm inter}\frac{\mathbf{M}_{\rm Ni}\cdot\mathbf{M}_{\rm Co}}{M_{\rm Ni}M_{\rm Co}}\right), \tag{3}$$

with z the number of nearest neighbors and different $J_{\rm intra}$ for the respective sublayer.

A series of Co/Cu/Ni/Cu(001) ultrathin single crystalline films was investigated. All parts of the experiments were performed *in situ* under UHV conditions ($p \leq 2 \times 10^{-10}$ mbar). The sublayers show a layerwise growth monitored with oscillating medium energy electron diffraction (MEED) intensity. Edge jump analysis of the x-ray absorption signal [14] is in favor of the argument that the amount of pinholes (so the corresponding ferromagnetic "bridges") between the magnetic layers can be considered negligible. To avoid interdiffusion the measurements were performed mostly below room temperature. We chose $d_{\rm Co} \approx 2$ ML and $d_{\rm Ni} \approx 4$ ML. The Cu spacer was varied between 2 and 25 ML. The hybridization of the Cu s-d bands with Ni or Co has strong effects on T_C which was published before [13]. Therefore one has to compare the magnetic properties of the capped magnetic films with the ones of the trilayer. All films are magnetized in the plane and the magnetization data are taken at remanence.

First, we present the *in situ* UHV mutual inductance ac-susceptibility ($\chi_{\rm ac}$) measurements [4] for the upper limit of a thick spacer layer:

$$\chi_{\rm ac}(T) = \frac{\Delta M(T)}{\Delta H} \, . \tag{4}$$

We are in the $\omega \to 0$ and $\Delta H \to 0$ limit with 213 Hz and an amplitude of $\hat{H} = 26$ A/m. Figure 2(a) shows the temperature dependent $\chi_{\rm ac}(T)$ of a 25 ML Cu/5.0 ML Ni/Cu(001) sample. The peak at 248 K with $\chi_{\rm max} = 380$ determines the $T_C^{\rm low}$. After adding 2.1 ML Co on top, the data of Fig. 2(b) were recorded. Two separate signals in $\chi_{\rm ac}(T)$ are observed [15]. The upper peak at 435 K with $\chi_{\rm max} = 10\,000$ is the same as we record for a bare Co film of the same thickness and corresponds to the Curie-Weiss singularity at the Curie temperature. The lower cusp at 250 K is the one of the Ni layer and is reduced compared to the uncoupled case shown in Fig. 2(a). Comparing $\chi_{\rm max}$ of Ni in the uncoupled [Fig. 2(a)] and the coupled case in the trilayer [Fig. 2(b), solid line], the value is 4 times reduced. This observation confirms the presence of interlayer coupling between Ni and Co and can be interpreted in terms of an exchange field $H_{\rm exch}$. The χ signal of Ni in the trilayer is caused by spin fluctuations. But in contrast to the uncoupled Ni film $H_{\rm exch}$ is present and damps the spin fluctuations leading to a reduced $\chi_{\rm max}$. To verify the influence of $H_{\rm exch}$ on $\chi_{\rm max}$ of Ni an additional experiment was performed. With a larger $\hat{H} = 147$ A/m an increased $\chi_{\rm max}$ is recorded [dashed line Fig. 2(b)]. The usual behavior would be a decrease of $\chi_{\rm max}$, since the data are normalized to the external amplitude [see Eq. (4)]. The opposite result seen in Fig. 2(b) implies that the larger amplitude cancels partly the effect of the exchange field. Since an increase of only $\approx$100 A/m in the amplitude is able to cause differences in the χ signal, we conclude that the amplitude is comparable to $H_{\rm exch}$. That is, the sample is in the weak coupling regime. The decrease of $\chi_{\rm max}$ is in good agreement with the theoretical scheme of Ref. [5]: The $\chi_{\rm max}$ at $T_C^{\rm high}$ corresponds to the phase transition while $\chi_{\rm max}$ at T^* to a susceptibility resonance. Another interesting conclusion, which is unrelated to the

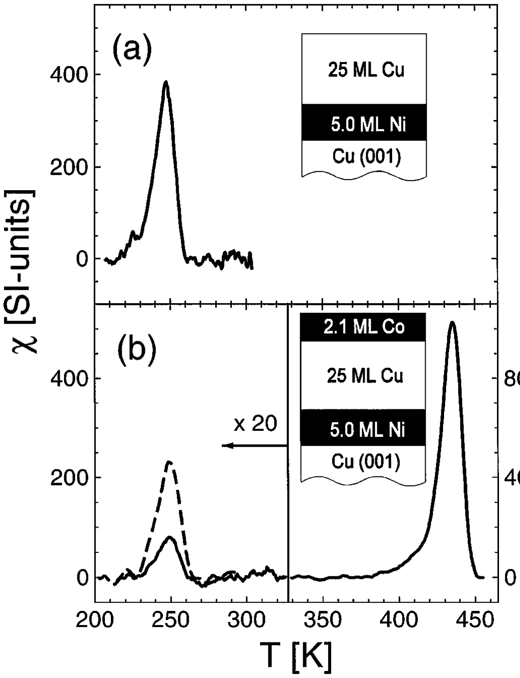

FIG. 2. (a) $\chi_{\rm ac}(T)$ data in SI units for Cu/Ni/Cu(001) and (b) for Co/Cu/Ni/Cu(001) trilayer. The left axis corresponds to the Ni signal at 250 K, the right one to the Co signal, respectively. For the dashed line, see the text.

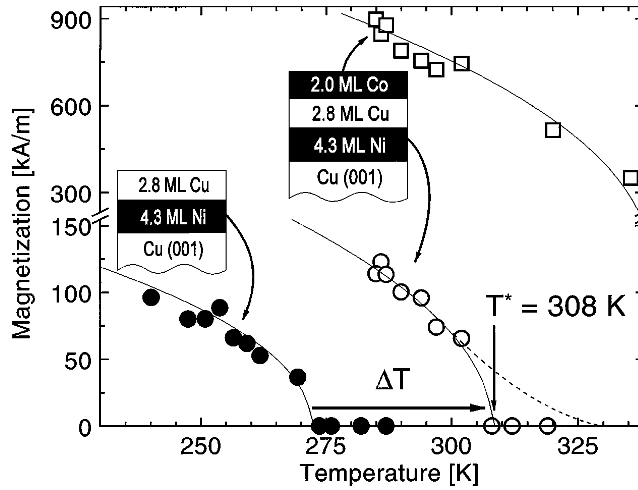

FIG. 3. Temperature-dependent element-specific magnetization measured by XMCD: Solid circles show $M_{\rm Ni}(T)$ of Cu/Ni/Cu(001), open symbols for the Co/Cu/Ni/Cu(001) trilayer. $M_{\rm Ni}(T)$ (squares) and $M_{\rm Co}(T)$ (circles). The solid lines are analytical functions fitted to the data. The dashed line is one possible behavior of an induced magnetization. The magnetic moment of bulk Ni ($0.6\mu_B$) corresponds to 515 kA/m.

onset of long-range order, can be seen in the χ signal of the uncoupled Co and Ni films. The $\chi_{\rm max}$ of Co [Fig. 2(b)] is ≈25 times larger than the one of the bare Ni film in Fig. 2(a). Since the susceptibility is a quadratic function of the magnetic moment per atom ($\mu_{\rm Co}, \mu_{\rm Ni}$), one finds a ratio $\mu_{\rm Co}/\mu_{\rm Ni} \approx 5$. This is larger than the corresponding ratio of the bulk moments ($\mu_{\rm Co}/\mu_{\rm Ni} \approx 2.8$ [16]), but agrees well with a reduced magnetic moment $\mu_{\rm Ni} = 0.35\mu_B$ measured in 4 ML Ni/Cu(001) [17]. This yields $\mu_{\rm Co}/\mu_{\rm Ni} = 5.1$ assuming that $\mu_{\rm Co}$ is bulklike.

For strongly coupled films, that is with thinner Cu spacer, one single maximum in $\chi_{\rm ac}(T)$ is observed. $H_{\rm exch}$ reduces $\chi_{\rm max}$ at the lower temperature drastically and due to the limited sensitivity of the $\chi_{\rm ac}$ setup no $\chi_{\rm max}$ at T^* is recorded in that case. However, the behavior of the order parameter $M_{\rm Ni}$ can be measured with XMCD. In Fig. 3 we compare $M_{\rm Ni}(T)$ in a Co/Cu/Ni/Cu(001) trilayer and Cu/Ni/Cu(001) bilayer. The remanent magnetization $M_{\rm Ni}(T)$ of the 4.3 ML Ni layer magnetization capped with 2.8 ML Cu is shown by the solid circles in Fig. 3. The signal vanishes at $T_C^{\rm low} = 272$ K. Subsequently, 2.0 ML Co was evaporated on top. $M_{\rm Co}(T)$ is given by the open squares with $T_C^{\rm high} \approx 340$ K. The solid lines are analytical functions for consistent fitting of the experimental data. The behavior of the Ni magnetization in the trilayer (open circles) is the interesting result: The temperature where $M_{\rm Ni}$ vanishes is shifted by $\Delta T = 36$ K to $T^* = 308$ K. Above 308 K we do not detect a XMCD signal of Ni. Since $J_{\rm inter}$ was not acting in the capped Ni sample, the positive ΔT arises from the increased coupling energy in the trilayer. According to Eq. (3) one finds $k_B\Delta T = J_{\rm inter}$ and $J_{\rm inter} \approx 3$ meV. This is a reasonable value for magnetic layers separated by very thin spacers; see, for example, Fig. 3 of Ref. [18]. It is still much smaller than typical values for $J_{\rm intra} \approx 100$ meV. The stronger increase in ΔT compared to the weakly coupled case in Fig. 2 is also consistent with the calculations of [5]. However, the functional behavior of $M_{\rm Ni}(T)$ does not change as expected. In the presence of an exchange field $H_{\rm exch}$ $M_{\rm Ni}(T)$ should qualitatively follow a temperature dependence as indicated by the dashed line in Fig. 3. Within our experimental accuracy we clearly rule out such a dependence, but a very small nonzero $M_{\rm Ni}(T) < 9$ kA/m may still be present above T^*, as it is theoretically predicted [5]. In the experiment, however, T^* looks like a critical temperature above which long range order disappears in the Ni layer.

The results are summarized in Table I. The specific temperatures, that is $T_C^{\rm low}$, T^*, the difference of the two ΔT and $T_C^{\rm high}$ of various bi- and trilayers are given. The two directly coupled Co/Ni bilayers in the first two lines are taken from Refs. [12,13] and have one common T_C. Six sets of trilayer data are listed with increasing Cu spacer layer thickness. The data of Fig. 3 are given in line 5 and the ones of Fig. 2 in the last line, respectively. The monotonic decrease of ΔT with increasing $d_{\rm Cu}$ is evident and confirms that ΔT is monotonically related to the interlayer coupling strength $J_{\rm inter}$. For a few cases a lower limit for ΔT or $T_C^{\rm high}$ could be given only, since $T_C^{\rm high}$ is above the temperature, where interlayer diffusion sets in. Note that for one data set (a) Co is at the bottom and Ni is the top layer. The positive ΔT is present for that case as well and does not depend on the sequence of Ni and Co layers.

TABLE I. Characteristic temperatures for various bi- and trilayers: The first column lists T_C^{low} of uncapped (first two rows) and capped layers. An increase in T_C^{low} in the trilayer is shown as T^* and $\Delta T = T^* - T_C^{\text{low}}$. The last column corresponds to T_C^{high}. Monolayer thicknesses are given in parentheses.

	T_C^{low}/K	T^*/K	ΔT/K	T_C^{high}/K
Co(1.3)/Ni(5.3)/Cu(001), Ref. [12]	345		90	435
Co(1.7)/Ni(4.1)/Cu(001), Ref. [13]	242		>208	>450
Co(2.2)/Cu(2.0)/Ni(4.0)/Cu(001)	197	259	62	>270
Co(1.8)/Cu(2.0)/Ni(4.0)/Cu(001)	200	258	58	262
Co(2.0)/Cu(2.8)/Ni(4.3)/Cu(001)	272	308	36	340
Co(2.9)/Cu(2.8)/Ni(4.8)/Cu(001)	275	312	37	>450
Ni(4.2)/Cu(6.0)/Co(2.1)/Cu(001)[a]	198	225	27	300
Co(2.1)/Cu(25)/Ni(5.0)/Cu(001)	248	250	...	435

[a]For this sample Co and Ni are interchanged. Values in the first two columns are for Co and the last column for Ni.

From an experimental point of view, the two transitions (Table I) seen in the susceptibility [Fig. 2(b)] and the element resolved magnetization (Fig. 3) may look like two Curie temperatures. But in terms of thermodynamics there is only one true phase transition at the higher temperature. Interestingly, the temperature dependence of M_{Ni} (Fig. 3, open circles) in the strongly coupled case does not show any evidence for the presence of an exchange field. It looks the same as in the bare case (solid circles) with a shifted temperature of vanishing magnetization.

In summary, the onset of long-range order in exchange coupled epitaxial monolayers is investigated by temperature dependent susceptibility and element resolved magnetization measurements. Our experimental results support the theoretical analysis of Wang and Mills [5]. We find two signals in the temperature-dependent susceptibility of weakly coupled films, corresponding to the "true singularity" at the phase transition of the whole layer stack and a resonantlike anomaly at the lower transition. In the strongly coupled case only one susceptibility signal at the phase transition is observed. The order parameters of the two ferromagnets, which we measure element specific, vanish at different temperatures. The lower transition temperature in the trilayer is shifted to higher temperatures due to the interlayer exchange coupling.

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