

Enhanced induced magnetization in coupled magnetic trilayers in the presence of spin fluctuations

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Motivated by recent experiments, the effect of the interlayer exchange interaction J_{inter} on the magnetic properties of coupled Co/Cu/Ni trilayers is studied theoretically. Here the Ni film has a lower Curie temperature $T_{C,Ni}$ than the Co film in case of decoupled layers. We show that by taking into account magnetic fluctuations the interlayer coupling induces a strong magnetization for $T \geq T_{C,Ni}$ in the Ni film. For an increasing J_{inter} the resonancelike peak of the longitudinal Ni susceptibility is shifted to larger temperatures, whereas its maximum value decreases strongly. A decreasing Ni film thickness enhances the induced Ni magnetization for $T \geq T_{C,Ni}$. The measurements cannot be explained properly by a mean-field estimate, which yields a ten times smaller effect. Thus, the observed magnetic properties indicate the strong effect of two-dimensional magnetic fluctuations in these layered magnetic systems. The calculations are performed with the help of a Heisenberg Hamiltonian and a Green's-function approach. [S0163-1829(99)51446-X]

Recently, the element specific magnetization and the longitudinal susceptibility of magnetic epitaxial Co/Cu/Ni trilayers grown on Cu(001) have been measured.^{1,2} The two ferromagnetic Ni and Co films are coupled by the indirect exchange interaction J_{inter} across the nonmagnetic Cu layer, which exhibits an oscillatory behavior as a function of the thickness d_{Cu} of the spacer, and an overall decay like d_{Cu}^{-2} .²⁻⁴ The thicknesses d_{Ni} and d_{Co} of the Ni and Co films are chosen in such a way that for a vanishing interlayer coupling the Ni film has a lower Curie temperature than the Co film, i.e., $T_{C,Ni}(d_{Ni}) < T_{C,Co}(d_{Co})$. J_{inter} induces for $T > T_{C,Ni}$ a considerable magnetization in the Ni film, which has been measured to vanish $\sim 30-40$ K above $T_{C,Ni}$. In this work we show that the induced strong Ni magnetization can theoretically only be obtained properly by taking into account magnetic fluctuations in the Ni film. If these fluctuations are neglected in the calculations [for example within a mean-field theory (MFT) approach⁵], the resulting induced Ni magnetization $M_{Ni}(T)$ for $T \geq T_{C,Ni}$ is an order of magnitude smaller. Vice versa, the neglect of these fluctuations requires an unrealistic large value for J_{inter} to yield the observed shift ΔT of $M_{Ni}(T)$ to larger temperatures. Generally, spin fluctuations diminish the magnetization of a two-dimensional (2D) magnetic system more strongly than for bulk magnets.^{6,7} An external magnetic field suppresses the action of these fluctuations, resulting in a stronger increase of the magnetization in 2D than in bulk systems. Similarly, in case of a coupled trilayer the interlayer coupling reduces the fluctuation effect, since it acts as an external magnetic field. Consequently, J_{inter} has a pronounced effect on the Ni film magnetization. The magnetic behavior of such a system can be used to study the action of the strong 2D spin fluctuations.

To take into account the collective magnetic excitations (spin waves, magnons), we apply a many-body Green's-function approach, and use the so-called Tyablikov (or RPA) decoupling.⁸ Since within this method interactions between magnons are partly taken into account, the whole temperature range of interest up to the Curie temperature can be considered. A Heisenberg-type Hamiltonian on an fcc(001) thin film with $d = d_{Ni} + d_{Co}$ monolayers (ML) is assumed with localized magnetic moments $\boldsymbol{\mu}_i = \mu_i \mathbf{S}_i / S$ on lattice sites i :

$$\mathcal{H} = -\frac{1}{2} \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i \mathbf{B} \cdot \boldsymbol{\mu}_i + \frac{1}{2} \sum_{\substack{i,j \\ i \neq j}} \frac{1}{r^5} [\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j r^2 - 3(\mathbf{r} \cdot \boldsymbol{\mu}_i)(\mathbf{r} \cdot \boldsymbol{\mu}_j)]. \quad (1)$$

Quantum-mechanical spins with spin quantum number $S = 1$ are assumed. Due to the dipole interaction the magnetization $M_i(T) = \langle S_i^z \rangle$ is directed in plane, determining the quantization axis (z direction). The external magnetic field $\mathbf{B} = (0,0,B)$ is applied parallel to this axis. J_{ij} are the exchange couplings between nearest-neighbor spin pairs which are chosen in such a way that they yield the observed Curie temperatures for the separate (i.e., decoupled) layers. We put $J_{CoCo} = 398$ K per bond to obtain $T_{C,Co}(2) = 435$ K for a Co film with $d_{Co} = 2$ ML.¹ To account for the diminished interface magnetic state of the Ni film, we distinguish between exchange couplings in the interface layers and the film interior layers. With $J_{NiNi}^{interface} = 30$ K and $J_{NiNi}^{interior} = 172$ K per bond one obtains for a Ni film with $d_{Ni} = 5$ ML $T_{C,Ni}(5)$

$=267$ K.² These numbers for the exchange couplings are somewhat lower than the corresponding values obtained from the bulk Curie temperatures. In addition an interlayer exchange coupling J_{inter} across the nonmagnetic Cu spacer layer between Ni and Co spins in the interlayers next to Cu is assumed. Positive as well as negative values of J_{inter} can be considered, thus preferring parallel ($J_{inter}>0$) or antiparallel ($J_{inter}<0$) magnetized Ni and Co film. The last term in Eq. (1) is the magnetic dipole coupling between spins μ_i and μ_j separated by vectors $\mathbf{r}=\mathbf{r}_j-\mathbf{r}_i$, denoting $r=|\mathbf{r}|$. The slowly converging oscillating lattice sums are converted into rapidly converging ones with the help of Ewald summation.⁹ Layer-dependent magnetic moments μ_i are assumed.¹⁰ In particular we put $\mu_{Ni}^{interface}=0.46\mu_B$, $\mu_{Ni}^{interior}=0.61\mu_B$, and $\mu_{Co}=2.02\mu_B$ for all Co layers, where μ_B is the Bohr magneton. Lattice anisotropy terms are not considered here.

For the calculation of the layer-dependent magnetizations, $M_i(T)$, $i=1\dots d$, we consider the following two-times (commutator-) Green's functions, which are written in spectral representation as^{11,12}

$$G_{ij}^{+- (n)}(\omega, \mathbf{k}_{\parallel}) = \langle\langle S_i^+; (S_j^z)^n S_j^- \rangle\rangle_{\omega, \mathbf{k}_{\parallel}} = \langle\langle S_i^+; C_j^{(n)} \rangle\rangle_{\omega, \mathbf{k}_{\parallel}}. \quad (2)$$

Here the i, j refer to layer indices. Because we assume ferromagnetically ordered layers, the lateral periodicity has been used to apply a Fourier transformation into the 2D momentum space, \mathbf{k}_{\parallel} being the 2D wave vector. The Green's functions are determined by solving the familiar equation of motion. Higher order Green's functions are approximated by the Tyablikov (RPA) decoupling⁸ for the exchange and dipole interactions ($i \neq k$):

$$\langle\langle S_i^z S_k^+; C_j^{(n)} \rangle\rangle \approx \langle S_i^z \rangle \langle\langle S_k^+; C_j^{(n)} \rangle\rangle = M_i(T) G_{kj}^{+- (n)}, \quad (3)$$

i.e., spin operators S_i^z are replaced by their expectation values $M_i(T)$. Different integers $n \leq 2S-1$ have to be considered in order to calculate different spin quantum numbers S .¹¹ The equations of motion lead to a set of d coupled linear equations for the Green's functions. With the help of the spectral theorem the respective expectation values (or correlation functions) $\langle\langle (S_j^z)^n S_j^- S_i^+ \rangle\rangle$ are determined. The magnetization $M_i(T)$ is obtained from the usual relations between spin operators. By comparison with a recent quantum Monte Carlo calculation of a Heisenberg monolayer in an external magnetic field¹³ it was shown that the applied Green's-function method yields a satisfactory result for the magnetization.^{14,15}

In Fig. 1 we present results for the Ni magnetization $M_{Ni}(T)$ as a function of the temperature T calculated with different interlayer couplings J_{inter} . We consider a Co/Cu/Ni trilayer with $d_{Ni}=5$ Ni, $d_{Cu}=3$ Cu, and $d_{Co}=3$ Co monolayers, respectively. For comparison experimental results for the same system are also shown.² The layer-dependent magnetizations $M_i(T)$, $i=1\dots d$, are determined from an iterative procedure. Presented are the Ni magnetizations $M_{Ni}(T)$ averaged over all Ni layers. We use the inflection point T_{infl} of $M_{Ni}(T)$ as a measure of the corresponding temperature shift $\Delta T(J_{inter})=T_{infl}-T_{C, Ni}$ of the Ni magnetization with respect to the decoupled case. One observes that already a small value of the interlayer coupling

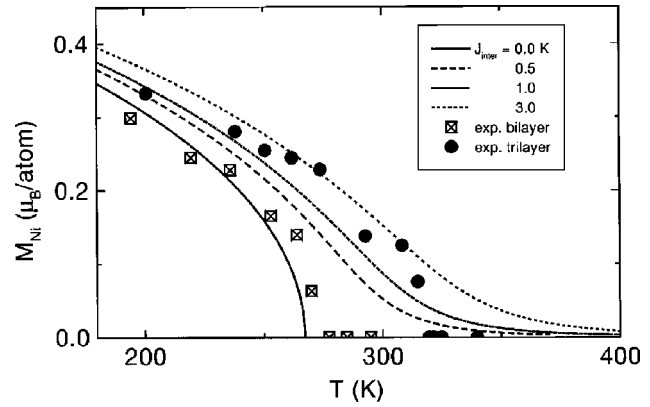


FIG. 1. Ni magnetization $M_{Ni}(T)$ for a Co/Cu/Ni trilayer as a function of the temperature T calculated by the Green's-function approach. Different interlayer couplings J_{inter} (in K/bond) are assumed as indicated. An epitaxial trilayer with $d_{Ni}=5$ Ni and $d_{Co}=3$ Co monolayers (ML) is assumed. The values for the exchange couplings within the Ni and Co films and the corresponding magnetic moments are given in the text. $T_{C, Ni}=267$ K is the Curie temperature of the single Ni film. In addition experimental results for the Ni magnetization are displayed for a single 4.8 ML Ni film capped with Cu ('bilayer'), as well as for a Co/Cu/Ni trilayer with $d_{Ni}=4.8$ ML, $d_{Co}=2.8$ ML, and $d_{Cu}=2.8$ ML (Ref. 2).

J_{inter} produces a comparably large $\Delta T(J_{inter})$. For example, $J_{inter}=1$ K results in $\Delta T \approx 30$ K. Such a value for J_{inter} compares well with various results measured formerly with different methods.^{4,16} Corresponding results have been determined by us also from a MFT approach. For the same value of J_{inter} the calculated $\Delta T(J_{inter})$ obtained from this approximation is about ten times smaller than the value resulting from the Green's-function method.

We stress that this strong difference is a result of the 2D character of the magnetic trilayer system. The action of an external magnetic field for $T \geq T_C$ is much more pronounced for a 2D magnet than for a corresponding bulk system.⁷ For the coupled trilayer system under consideration the interlayer coupling J_{inter} acts similar as an external magnetic field. Therefore, for temperatures $T \geq T_{C, Ni}$ close to the Curie temperature of the single Ni film already a small J_{inter} is sufficient to induce a marked Ni magnetization and the corresponding temperature shift ΔT . In contrast, within a MFT approach the exchange coupling alone results in a finite remanent magnetization for a 2D magnet, and does not need the support of the dipole coupling or an external magnetic field. In this case a small interlayer coupling adds simply to the strong Ni-Ni exchange coupling and results in a correspondingly small value of ΔT .

We have tested the assumption that the interlayer coupling acts similar as an external magnetic field. The results of $M_{Ni}(T)$ for a single (i.e., decoupled) Ni film with $d_{Ni}=5$ ML, with an external magnetic field acting exclusively on the topmost Ni layer with a strength $B=J_{inter}/\mu_{Ni}$, are practically the same as for the corresponding coupled trilayer system.

Furthermore, we have calculated the induced Ni magnetization at $T \geq T_{C, Ni}$ for different thicknesses d_{Ni} of the Ni film. Results for $\Delta T(d_{Ni})$ are shown in Fig. 2 for two different values of J_{inter} and for $1 \leq d_{Ni} \leq 6$ ML. The other coupling parameters are the same. The resulting *absolute* value

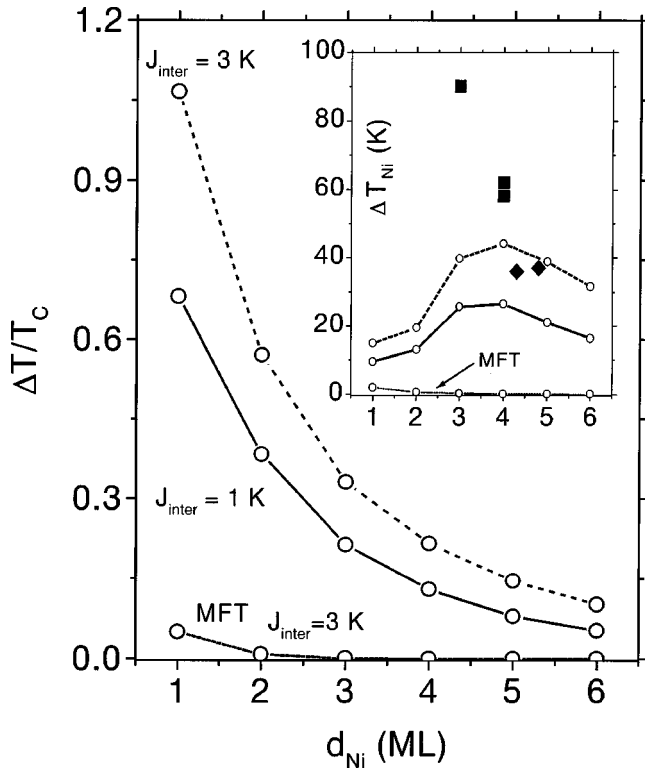


FIG. 2. The temperature difference $\Delta T(d_{\text{Ni}})$ between the inflection point of the Ni magnetization $M_{\text{Ni}}(T)$ in the coupled Co/Cu/Ni trilayer and the Curie temperature $T_{C,\text{Ni}}(d_{\text{Ni}})$ of the single Ni film as a function of the Ni film thickness d_{Ni} . Two different interlayer couplings $J_{\text{inter}}=1$ K and 3 K are assumed, and the other system parameters are the same as for Fig. 1. The figure displays the relative temperature difference ΔT scaled by $T_{C,\text{Ni}}(d_{\text{Ni}})$. In the inset the absolute temperature shift is given. For comparison we show the respective results for ΔT calculated by an MFT approach, assuming $J_{\text{inter}}=3$ K. Experimental results are also given for two different Cu thicknesses: \blacklozenge : $d_{\text{Cu}}=2.8$ ML; and \blacksquare : $d_{\text{Cu}}=2.0$ ML (the latter corresponds to a larger $J_{\text{inter}}>3$ K).

$\Delta T(d_{\text{Ni}})$ exhibits a maximum at about $d_{\text{Ni}}=4$ ML, see inset of Fig. 2. For comparison the corresponding results calculated by the MFT approach are also shown. On the other hand, the relative temperature shift $\Delta T/T_{C,\text{Ni}}(d_{\text{Ni}})$, scaled by the Curie temperature $T_{C,\text{Ni}}(d_{\text{Ni}})$ of the single Ni film, increases by reducing the thickness of the Ni film (see Fig. 2). This indicates the increasing importance of the action of the magnetic fluctuations for a decreasing film thickness. Experimental results are also displayed for two different Cu spacer thicknesses.

In addition we investigate the longitudinal susceptibility $\chi_{\text{Ni}}(T)$ of the Ni film. For this purpose $\chi_i(T)$ for the i th magnetic layer, $i=1 \dots d$, is calculated from the difference of the magnetizations $M_i(T,B)$ with $B=0$ and $B=2$ G,

$$\chi_i^{zz}(T) \equiv \chi_i(T) = \frac{\partial M_i(T,B)}{\partial B} \approx \frac{\Delta M_i(T,B)}{\Delta B}. \quad (4)$$

In Fig. 3 the Ni susceptibility $\chi_{\text{Ni}}(T)$ averaged over all Ni layers is displayed, corresponding to the results of Fig. 1. A resonancelike peak of $\chi_{\text{Ni}}(T)$ is obtained for $T \geq T_{C,\text{Ni}}$ as has been reported previously.⁵ With increasing interlayer cou-

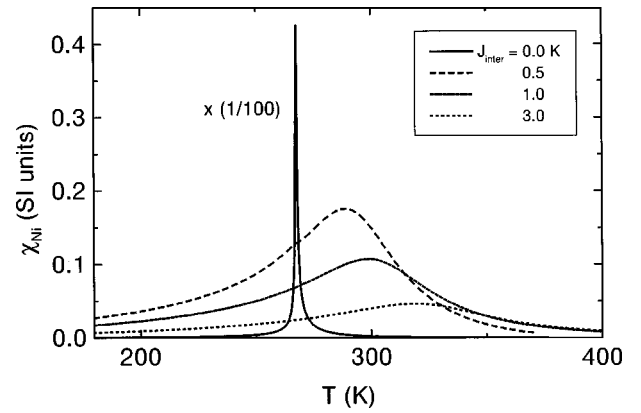


FIG. 3. Ni film susceptibility $\chi_{\text{Ni}}(T)$ as a function of the temperature for different interlayer couplings as indicated. The susceptibility field amplitude is 2 G. The same Co/Cu/Ni trilayer system as described in Fig. 1 is assumed. Note that the susceptibility curve for $J_{\text{inter}}=0$ is scaled by the factor 1/100.

pling J_{inter} , the susceptibility peak is shifted to higher temperatures. Also, the maximum value of $\chi_{\text{Ni}}(T)$ reduces strongly and its corresponding width increases markedly.⁵ For a strong J_{inter} the Ni susceptibility is so small that it may be hardly measurable. A singularity of $\chi_{\text{Ni}}(T)$ will occur at $T=T_{C,\text{Co}}$ since there the induced Ni magnetization, although small, vanishes in accordance with the vanishing Co magnetization. Thus, $T_{C,\text{Co}}$ corresponds to the true phase transition temperature of the coupled magnetic Co/Cu/Ni trilayer system.⁵

In Refs. 1 and 2 it was found for the Co/Cu/Ni trilayer system that the observed Ni remanent magnetization vanishes above a temperature T_{Ni}^* , where $T_{C,\text{Ni}} < T_{\text{Ni}}^* < T_{C,\text{Co}}$. On the other hand, either no susceptibility signal or only a small peak in $\chi_{\text{Ni}}(T)$ could be measured at T_{Ni}^* . This might be due, e.g., to the occurrence of a multidomain state or to a magnetic reorientation in the Ni film. A true phase transition in the thermodynamic sense is reminiscent to a nonanalytical behavior of the free energy, resulting in singularities of, e.g., the correlation length or the susceptibility, as found at $T_{C,\text{Co}}$. Regardless of the particular behavior of the magnetic properties at T_{Ni}^* , we emphasize that the observed strong induced Ni magnetization and the shift of the Ni susceptibility for $T \geq T_{C,\text{Ni}}$ due to the interlayer coupling J_{inter} is caused by the presence of magnetic fluctuations. To compare the measured and calculated temperature shift ΔT we have assumed $T_{\text{infl}} \approx T_{\text{Ni}}^*$.

We have investigated asymmetric trilayers which exhibit different Curie temperatures for the decoupled Ni and Co films, e.g., for single magnetic layers or for a trilayer with a thick nonmagnetic spacer. The interlayer coupling J_{inter} influences mainly the magnetization of the Ni film, which has the lower ordering temperature, whereas $T_{C,\text{Co}}$ stays practically constant. On the other hand, a symmetric system, e.g. a Ni/Cu/Ni trilayer with equal Ni film thicknesses or a periodic multilayer system, has a single Curie temperature T_C . Here J_{inter} will enhance T_C considerably by amounts similar as discussed for the asymmetric trilayer. Indeed this has been observed for a Ni/Au multilayer system.⁴ In principle, by an appropriate combination of tri- and multilayers, for instance by varying the materials and the thicknesses of the magnetic

and nonmagnetic layers, one might tune the magnetic properties according to possible applications.

In summary, we have calculated the action of the interlayer exchange coupling in a magnetic Co/Cu/Ni trilayer system by means of a many-body Green's-function approach for a Heisenberg Hamiltonian. J_{inter} induces a considerable magnetization in the Ni film for $T \gtrsim T_{C,Ni}$, and shifts the Ni susceptibility peak to larger temperatures. Also the width of the Ni susceptibility increases, whereas its maximum value decreases strongly. We have shown that for reasonable values of J_{inter} the observed strong induced Ni magnetization can be obtained only if the magnetic fluctuations in these 2D systems are taken into account properly. Corresponding re-

sults as calculated by a MFT approach, which neglect these fluctuations, yield a ten times smaller increase, and cannot explain the observed magnetic behavior for the Co/Cu/Ni trilayer system. The influence of the magnetic fluctuations becomes stronger for smaller Ni film thicknesses (see Fig. 2), indicating the 2D character of the important correlations. Note that we have investigated the effect of J_{inter} on the magnetic properties solely by considering thermal fluctuations. Other possible influences such as magnetic noncollinearities are not discussed here.

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