Magnetic properties of a Co/Cu/Ni trilayer on the Cu(100) surface

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Magnetic moments, total energies, and exchange interactions have been calculated for $Co_3/Cu_N/Ni_5$ trilayer and Cu_N/Ni_5 bilayer on the Cu(100) substrate by means of the interface Green's-function technique within the basis set of linear muffin-tin orbitals. We find that Co and Ni layers in the trilayer are exchange coupled, and observe that the energy difference between the ferromagnetic and the paramagnetic samples is substantially larger than the energy difference between the ferromagnetic sample and the sample where only Ni layers are paramagnetic, while Co layers have nonvanishing magnetic moments. These results are in agreement with recent experiments, where it was observed that the magnetization of Co and Ni layers vanishes at different temperatures, and the difference oscillates as a function of thickness of nonmagnetic spacer layer. Magnetic moment profiles, as well as layer-resolved effective exchange parameters, were calculated and discussed.

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I. INTRODUCTION

Progress in growing of high-quality multilayers by means of molecular-beam-epitaxy method has made it possible to design structures with tailor-made magnetic properties. Many studies have been devoted to experimental and theoretical investigation of the Co, Ni, and Co/Cu/Ni films on the Cu(100) surface[1–8] which in this regard have become model systems for understanding of physical phenomena in magnetic materials with reduced dimensionality, including the interlayer exchange coupling[9–12] and the giant magnetoresistance effects.[13] Also, some work has been devoted to an investigation of the Curie temperature T_C of magnetic layers. In Refs. 14,15 the T_C of Co and Ni layers on the Cu(100) surface were studied, respectively. Srivastava *et al.*[16] and Ney *et al.*[17] for the first time drew attention to the investigation of T_C of Co/Cu/Ni trilayer on the Cu(100) surface. The most important issue here was that two magnetic subsystems have quite different T_C in the bulk, $T_C^{\mathrm{Co}}=1380\,\mathrm{K}$ and $T_C^{\mathrm{Ni}}=627\,\mathrm{K}$ for bulk fcc Co and Ni, respectively. Thus, there is a fundamental problem of how the magnetization of a system with coupled Co and Ni layers will behave as a function of temperature. Results of element-specific magnetization measurements on Co/Cu/Ni trilayers deposited on the Cu(100) substrate by means of the x-ray magnetic-circular-dichroism (XMCD) technique have shown a number of interesting effects. The authors observed that the magnetization of Co and Ni layers vanished at different temperatures. This behavior was characterized by two different ordering temperatures, a global Curie temperature T_C and the ordering temperature of Ni layer T_{Ni}^*, where $T_{\mathrm{Ni}}^*<T_{\mathrm{Ni}}^{\mathrm{bulk}}$. In addition, the Curie temperature of Ni layer in the $Cu_{2.8}/Ni_{4.8}/Cu(100)$ is reduced as compared to that of $Co_{2.8}/Cu_{2.8}/Ni_{4.8}/Cu(100)$ when Co layers were evaporated from the trilayer surface. The enhancement of T_{Ni}^* in trilayer as compared to bilayer, $\Delta T_{C,\mathrm{Ni}}$, was related to the interlayer exchange coupling across the spacer. The coupling was found to be either ferromagnetic (FM) or antiferromagnetic (AFM), depending on the spacer thickness d_{Cu} in the trilayer. Also, the exchange coupling followed by the $\Delta T_{C,\mathrm{Ni}}$ had an oscillatory behavior and was found to decay like d_{Cu}^{-2}. This functional form gives rise to an assumption that the Ruderman-Kittel-Kasuya-Yoshida (RKKY) model[11,18] of the interlayer exchange coupling is applicable for the studied trilayer. The measured magnetic moment of Ni layers in the $Cu_{2.8}/Ni_{4.8}/Cu(100)$ is also substantially reduced in comparison to the $Co_{2.8}/Cu_{2.8}/Ni_{4.8}/Cu(100)$ trilayer.

It is worth to note that the experimental results of Ney *et al.*[17] are in agreement with the theoretical prediction of Wang and Mills,[19] where a superlattice model consisting of two components with a higher T_C^{higher} and a lower T_C^{lower} in the uncoupled case was studied in the framework of the phenomenological Ginzburg-Landau theory. According to results of this work, the magnetic susceptibility of the system with two different T_C displays one singularity and one maximum near the bulk transition temperatures of each constituent. The above-mentioned experimental results have also stimulated several theoretical studies of Co/Cu/Ni/Cu multilayers. In particular, Jensen *et al.*[20] carried out a study of the phenomena described in Ref. 17 by means of the model calculations, based on the Heisenberg-type Hamiltonian and a many-body Green's-function approach. Using parameters adjusted in such a way as to reproduce experimental temperature dependence of magnetization in Co/Cu/Ni trilayers, the authors conclude that the observed magnetic properties indicate the strong effect of two-dimensional magnetic fluctuations in these layered magnetic systems. On the other hand, Wu *et al.*[21] have studied this effect in the framework of the itinerant-electron model within the single-band Hubbard model and employing the spectral-density approach. They established a relation between the Curie temperature shift and the strength of the interlayer exchange coupling.

However, to the best of our knowledge, so far there were

no attempts to investigate from first-principles the nature of the vanishing magnetization of Co and Ni at different temperatures and to study the oscillatory behavior of the interlayer exchange coupling in this trilayer. Thus, we have carried out a number of theoretical calculations of the magnetization and the total energy for the Co/Cu/Ni trilayer on the Cu(100) surface in the framework of the local-spin-density-functional theory using a first-principles interface Green's-function technique and the fixed-spin-moment (FSM) method. The results of the total-energy calculations, as well as calculated effective exchange parameters, show that there are two distinct energy differences in this system, the energy difference between a ferromagnetic (antiferromagnetic) and paramagnetic solutions, and the energy difference between a solution where only Ni layers are paramagnetic, while Co layers have nonvanishing magnetic moments, and the energy of a ferromagnetic sample. This is in agreement with an existence of two ordering temperatures in $Co_3/Cu_N/Ni_5$ trilayers on Cu(100) surface. In addition, there is a magnetic coupling between Co and Ni layers in this system, which oscillates between the AFM to FM as a function of the thickness of nonmagnetic Cu spacer.

II. CALCULATION DETAILS

The surface Green's function method used in this work has been proposed by Lambrecht and Andersen[22] and further developed by Skriver and Rosengaard.[23] As the detailed description of the method may be found in a number of sources,[24–26] here we only outline the main features of this technique and specify the relevant parameters of our calculations. The main advantage of the Green's-function method is that it treats the surface of a semi-infinite sample rather than a slab as the model of a surface. The principal-layer technique[27] employed by the surface Green's-function method allows one to minimize the computational effort. As a result the calculation time scales linearly with the number of atomic layers. We have employed the linear muffin-tin orbitals basis set[28,29] in the tight-binding representation[30] with the orbital quantum number cutoff $l_{\max}=2$. The atomic-sphere approximation is used for the one-electron potential, and the core states are treated within the frozen-core approximation. The local-spin-density approximation is utilized for the exchange-correlation potential within the Vosko-Wilk-Nusair parametrization.[31] In the present implantation, besides the monopole and dipole, we also include higher multipole contributions to the electrostatic potential.[26]

We have used 505 k points in the irreducible part of the fcc Brillouin zone for the bulk calculations. The surface Brillouin zone integrals were evaluated using 136 k points in the irreducible part of the two-dimensional Brillouin zone for the (100) surface. The energy integrals were carried out in the complex plane using a semi-circular contour with 24 points. The thickness of the principal layer for the surface is equal to four physical layers. This set of parameters allows us to calculate total energy differences between different magnetic configurations with the accuracy 0.01 mRy. The lattice parameters for Cu, Ni, and Co are taken to be equal to the calculated equilibrium lattice parameter of Cu (3.59 Å) which is close to the experimental value for this metal (3.61 Å). We do not consider a possible lattice relaxation or distortion at the surface or the interfaces.

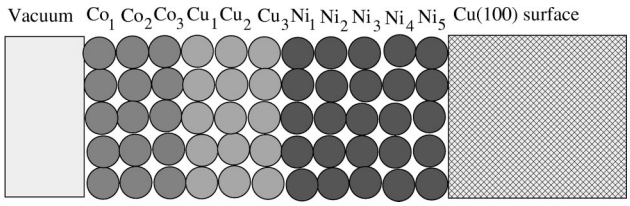

FIG. 1. Multilayer structures studied in this work are illustrated by an example of the surface geometry of $Co_3/Cu_N/Ni_5$ trilayer on the Cu(100) surface. Here Co_1 denotes a surface atom, while Co_3 and Ni_1 denote atoms at the interface towards the Cu spacer.

In order to be able to carry out calculations for a system with a predefined magnetic moment in a particular layer, we used the fixed-spin-moment method, described in the Appendix. The exchange coupling between the ferromagnetic and antiferromagnetic configurations of the multilayers was calculated from the total energy differences between these two solutions. Layer resolved effective exchange parameters J_0 were obtained by means of an expression derived by Liechtenstein *et al.*[32] Recently, this expression was used by number of authors for the study of exchange interactions and Curie temperature in systems with reduced dimensionality.[33–35]

III. RESULTS AND DISCUSSION

We have calculated the total energies and the layer-by-layer distributions of magnetic moments for a number of $Co_3/Cu_N/Ni_5$ trilayers and Cu_N/Ni_5 bilayers on the Cu(100) surface with different thickness of the Cu spacer layers $N=1$ to 5. These systems are most close to samples that were examined in Ref. 17, but in our study we neglect such phenomena as interface roughness and interdiffusion, and consider only integral numbers of layers of each material. Here we remark, that in principle the interface roughness can be treated via ensemble average, and the interdiffusion can be accounted for by means of the coherent-potential approximation.[36–38] However, this goes beyond the subject of the present study.

The surface geometry is presented in Fig. 1. As regards the magnetic properties, several constrains were imposed on the systems. First, we carried out fully self-consistent calculations for parallel, or ferromagnetic, orientations of all the moments in the multilayer. Second, the parallel spin alignment was assumed *inside* Co and Ni layers, but the antiparallel, or antiferromagnetic, orientations of moments was preset *between* Co and Ni layers. In the following we will refer to these calculations as self-consistent FM or self-consistent AFM cases, respectively. Next, we carried out calculations with constrained magnetic moment of Ni $\mu_{\mathrm{Ni}}=0$, but with fully relaxed magnetic moment on Co atoms. Here the fixed-spin-moment method described in the Appendix was used, and we will call this the constrained ferromagnetic (CFM) calculations. Finally, magnetic moment was suppressed on Co as well as on Ni sites, and the paramagnetic (PM) calculations were carried out.

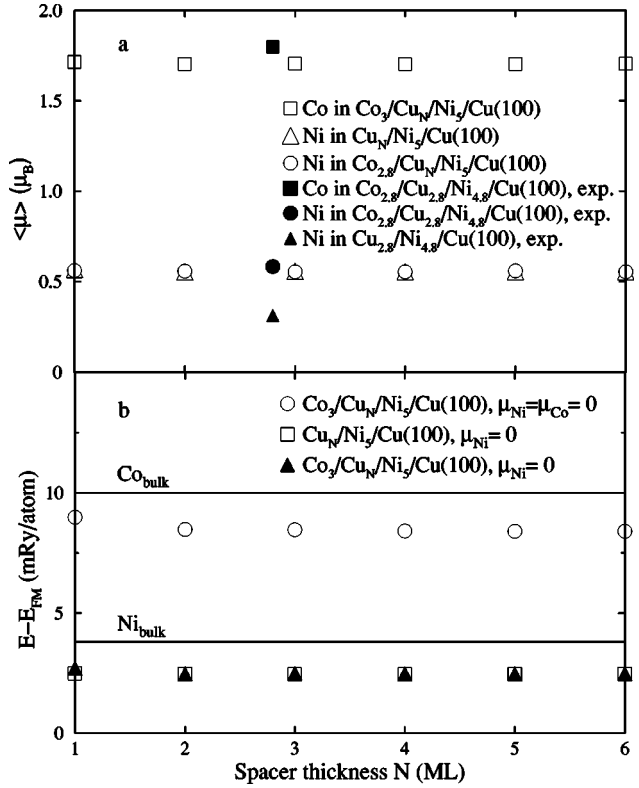

FIG. 2. (a) Averaged magnetic moments of Ni and Co layers in the Cu_N/Ni_5 bilayer (triangles) and the $Co_3/Cu_N/Ni_5$ trilayer (squares for Co and circles for Ni) on the Cu(100) surface as a function of the Cu spacer thickness N (in monolayers). Open symbols show calculated magnetic moments, whereas filled symbols correspond to the experimental results.[17] (b) Difference between the calculated total energies of various magnetic solutions for the Cu_N/Ni_5 bilayer (squares) and the $Co_3/Cu_N/Ni_5$ trilayer on the Cu(100) surface as a function of the Cu spacer thickness N (in monolayers). The FM solutions are used as the reference energies for the bilayers and the trilayers with the same spacer thickness. In this case magnetic moments of the Co and Ni layers were determined self-consistently. Open circles and open squares denote energies of the paramagnetic solutions for the trilayers and the bilayers, respectively. The energy of the constrained ferromagnetic solutions for the trilayers are given by the filled triangles. The energy differences between the FM and PM solutions for the bulk fcc Co (11 mRy/atom) and Ni (3.6 mRy/atom) are shown for comparison by horizontal lines. See text for more details.

A. Magnetic properties of trilayers

First, we discuss the dependence of the averaged magnetic moments for the $Co_3/Cu_N/Ni_5$ trilayer on the Cu(100) surface on Cu spacer thickness. Results of calculations are shown in Fig. 2(a). The calculated magnetic moments for Co and Ni are in good agreement with the experimental results reported in Refs. 17,39. Also, in Refs. 40,41 magnetic moments of Cu/Co/Ni/Cu, Cu/Ni/Co, Cu/Co/Cu/Ni/Cu multilayers on the Cu(100) surface were studied using the polarized-neutron-reflection method and no considerable reduction of magnetic moments of Ni layers was observed. For the Cu/Co/Ni/Cu sample the obtained magnetic moments were close to their bulk values, i.e., $(1.70\pm0.20)\mu_B$ and

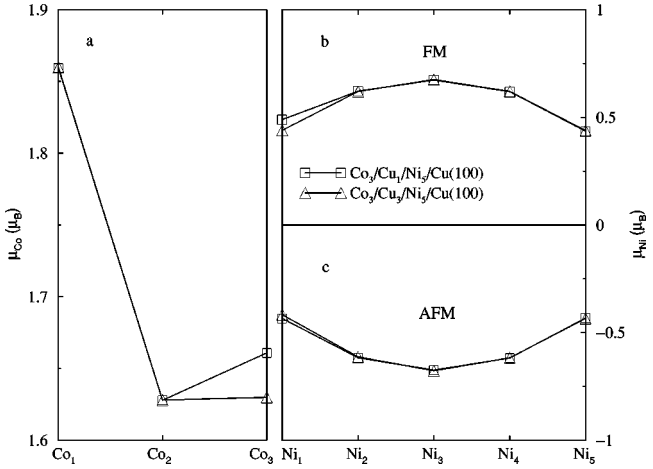

FIG. 3. Layer-resolved distribution of magnetic moments in the $Co_3/Cu_N/Ni_5$ trilayer on the Cu(100) surface for $N=1$ ML (squares) and $N=3$ ML (triangles) thick Cu spacer. Atoms are numbered according to Fig. 1. Magnetic moments of Co are shown in panel (a). They are given only for FM case. Panel (b) shows magnetic moments of Ni aligned ferromagnetically to those of Co. Panel (c) shows Ni moments for the antiferromagnetic case.

$(0.57\pm0.05)\mu_B$ for Co and Ni, respectively. For the Cu/Co/Cu/Ni/Cu structure magnetic moments for Ni and Co were slightly reduced and experimental moments of $(1.57\pm0.08)\mu_B$ for Co and $(0.50\pm0.04)\mu_B$ for Ni were found. Thus, our theoretical results are in excellent agreement with available experiments.

In Fig. 3 we present the layer-by-layer distribution of magnetic moments of Ni and Co atoms for the 1 and 3 ML thick Cu spacers. In this figure Co_1 denotes a surface atom, while Co_3 and Ni_1 denote atoms at the interface with Cu spacer, see Fig. 1. Magnetic moments of Co are displayed only for the FM case. The Co moments obtained in our AFM calculations were found to be close to those of the FM samples. For the 1 ML Cu spacer the magnetic moment of the surface and the interface Co atoms are increased as compared to their values in the bulk ($1.86\mu_B$ and $1.67\mu_B$, respectively). For the 3 ML Cu spacer only the surface Co atom has a larger magnetic moment. The other Co atoms have magnetic moments that are close to the calculated bulk value ($1.61\mu_B$). Magnetic moment of Ni atoms are shown in Figs. 3(b) and 3(c) for the FM and AFM cases, respectively. In both cases Ni_3 atoms have bulklike magnetic moments, but for Ni_5 atoms they are reduced (approximately to $0.43\mu_B$) and are practically independent of the spacer thickness. This indicates that multiple scattering effects such as confined quantum-well states only have a minor effect on the magnetization.[5] The magnetic moments of the Ni_1 atoms are also reduced and they are different for AFM and FM structures. Besides they depend on the spacer thickness. For the AFM case it is $0.41\mu_B$ for 1 ML spacer and $0.43\mu_B$ for 3 ML spacer. In the FM case it is $0.49\mu_B$ and $0.45\mu_B$ for 1 ML and 3-MD-thick Cu spacers, respectively. Note, that calculated magnetization profiles can be described within a superposition principle of independent binary interface magnetization

profiles and that the magnetic moments of the interface atoms can be estimated from the Slater-Pauling-interface curve.[5]

Our calculated moments for Ni atoms are in excellent agreement with the values that were assumed in the study of Co/Cu/Ni trilayers by Jensen *et al.*,[20] where interface Ni atoms have magnetic moments $0.46\mu_B$, while interior Ni atoms have magnetic moments $0.61\mu_B$. Though the magnetic moment within Co layers ($2.02\mu_B$) used by the authors of Ref. 20 is higher than our calculated value, this set of moments made it possible to explain the enhancement of T^*_{Ni}, in the $Co_3/Cu_3/Ni_5$ trilayer on the Cu(100) surface by means of the model calculations.

Note that theoretical calculations of the magnetization of Ni and Co layers on the Cu(100) surface give somewhat different results but they are in agreement with each other in the sense that magnetic moment of an interface atom as a rule is reduced and vice versa, it increases for a surface atom. In particular, Tersoff and Falicov[42] investigated Ni thin films on the Cu(100) surface using tight-binding method and found that the interface Ni atom has a magnetic moment of the order of $0.45\mu_B$. Ernst *et al.*[43] pointed out that self-consistence calculations of layer-resolved magnetic moments in a 3-ML-thick Ni films on the surface using the Korringa-Kohn-Rostoker Green's-function method give a value of $0.46\mu_B$ for the interface Ni atom. Our results for the interface Co and Ni atoms are in agreement with calculations of Niklasson, Johansson, and Skriver[5] where magnetic moment of a Co atom at Co/Cu interface was found to be close to the magnetic moment of a Co atom in the bulk, but the value of magnetic moment of Ni atom at Ni/Cu interface was reduced compared to the bulk Ni magnetization. Magnetic profile for the Ni layers also agree with that calculated for Cu/Ni/Cu sandwiches.[44,45]

From our calculated average and layer-resolved magnetic moments one can see that they depend only weakly on the Cu spacer thickness. There is a small increase of magnetization of interface Co and Ni atoms for the case of 1 ML Cu spacer, that disappears with the increasing thickness of the spacer. This is so because Co and Ni *d* electrons that mainly contribute to the magnetization, are tightly bounded to their sites and, therefore, Co and Ni *d*-wave functions cannot overlap directly in case of a thick spacer layer. Thus, it is unlikely that the presence of Co can influence strongly the values of local magnetic moments on Ni when the separation between them becomes large, as indeed observed in our calculations.

B. Energetics of different magnetic configurations in the trilayer

In this section we discuss the results of our total-energy calculations for different magnetic configurations of $Co_3/Cu_N/Ni_5$ trilayer from which we can make some qualitative conclusions regarding the behavior of T_c. Note that the best way of studying theoretically influence of the temperature on magnetic properties of a system would be to carry out simulations of spin dynamics in the framework of *ab initio* scheme[46] or using model Heisenberg-type Hamiltonians with parameters calculated by *ab initio* methods.[47,48] However, such calculations would be very time consuming, and they are beyond the scope of the present study. It is also well known that for most systems the exchange splitting and the local magnetic moments do not vanish above the critical temperature, and approaches based on the calculation of the Curie temperature directly from the energy difference between the paramagnetic and the ferromagnetic solutions grossly overestimate its value. At the same time, it has been pointed out earlier that the behavior of the energy difference between the PM and the FM solutions correlates with the behavior of the Curie temperature,[49] and we assume this correlation in the discussion below. In addition, we calculate layer-resolved effective-exchange parameters J_0 that estimate an energy needed for the rotation of one spin moment at a site in layer Λ on the angle θ from the ferromagnetic orientation, and can be related to T_c within the mean-field approximation.[32,34]

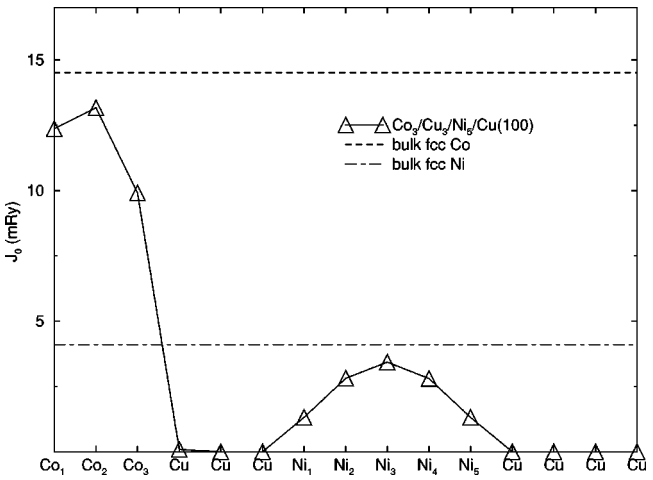

FIG. 4. Layer-resolved effective exchange parameters J_0 in the $Co_3/Cu_3/Ni_5$ trilayer on the Cu(100) surface. Atoms are numbered according to Fig. 1. The effective-exchange parameters for the bulk fcc Co (14.5 mRy) and Ni (4.1 mRy) are shown for comparison by horizontal lines. J_0^{Ni} in Cu_3/Ni_5 bilayers on the Cu(100) surface are very close to those in the trilayer on the energy scale of this figure.

In Fig. 2(b) the total energies of different magnetic configurations of $Co_3/Cu_N/Ni_5$ trilayer (relative to the energy of FM configuration) are shown as a function of the Cu spacer thickness. The PM configuration is about 8.56 mRy/atom higher in energy, and this value is quite close to the PM-FM energy difference in the bulk fcc Co, also shown in Fig. 2(b). Thus, the global T_C for the trilayer is going to be close to the bulk T_C of Co. The same conclusion may be drawn from the calculated effective-exchange parameters plotted in Fig. 4 for $Co_3/Cu_3/Ni_5$ trilayer. One can see that J_0 for Co layers is quite high, and is close to J_0 of bulk fcc Co, also shown in Fig. 4 with the dashed line. Note that our value for the effective-exchange parameter of the bulk fcc Co (14.5 mRy) is in good agreement with earlier calculations of this quantity that also leads to the T_C in the range 1300–1650 K, depending on the approximation used,[50] in good agreement with the experimental value 1380 K.

Let us now analyze results obtained in our fixed-spin-moment calculations for the CFM case, where we determined μ_{Co} self-consistently but constrain $\mu_{\mathrm{Ni}}=0$. One can see, that the difference in energy between this solution and the FM solution is much lower, of the order 2.5 mRy/atom, as compared to the PM-FM energy difference. This means that it costs much less energy for the system to demagnetize only Ni layer than to demagnetize the whole system. Note that in our case the energy difference between the CFM and FM solutions is lower than the PM-FM energy difference in the bulk fcc Ni [3.5 mRy/atom, Fig. 2(c)] in approximately the same proportion, as T^*_{Ni} is lower than the T_C of bulk Ni. Thus, our total-energy calculations show that there are two distinct energy differences in this system, the energy difference between the ferromagnetic and the paramagnetic samples, and the substantially smaller energy difference between the ferromagnetic sample and the sample where only Ni layers are paramagnetic, while Co layers have nonzero magnetic moments, and this is in complete agreement with Ref. 17, where the magnetization of Co and Ni layers was found to vanish at different temperatures.

This conclusion is also supported by calculated effective-exchange parameters for Ni layers, Fig. 4. It is clearly seen that J_0^{Ni} are much smaller than J_0^{Co}, being on the average twice as small as J_0^{Ni} in the bulk. This is in qualitative agreement with the experimental observation for T^*_{Ni}, which is roughly half of the bulk Ni T_C. Here again our calculated bulk effective-exchange parameter for the Ni is in good agreement with previous calculations.[32,50] It is also known that the mean-field estimates of T_C for the Ni lead to the lower values as compared to the experiment, most probably due to the neglect of the Stoner excitations.

C. Effect of Co cap layer and exchange coupling

When one evaporates the Co cap layer, and goes from $\mathrm{Co_3/Cu}_x\mathrm{/Ni_5}$ trilayer to $\mathrm{Cu}_x\mathrm{/Ni}$ bilayer, theory does not show any significant differences for magnetic moments of Ni layers, as can be seen in Fig. 2(a). The energy difference between the PM and FM calculations for the bilayer is close to the CFM-FM energy difference in trilayers [Fig. 2(b)], indicating that T^*_{Ni} in the trilayer may be close to T_C in the bilayer. The experimental results reported in Refs. 17,39 show that the temperatures at which Ni magnetization vanishes in the bilayers and trilayers are indeed close to each other ($\Delta T_{C,\mathrm{Ni}}$ is much smaller than the difference between the T^*_{Ni} and T_C of the bulk Ni). At the same time, according to the experiment the interlayer exchange coupling effects T^*_{Ni} and this effect is also qualitatively seen in our calculations. The difference in total energies between solutions with $\mu_{\mathrm{Ni}}=0$ (CFM solutions) and FM solutions in the bilayer relative to this difference in the trilayer depends weakly on the thickness of the spacer Cu layer, but it clearly oscillates (see Fig. 5) as a function of this thickness.

A substantial decrease of Ni magnetization in the bilayers as compared to that of the trilayers is observed in the experiment, and this disagrees with our calculated trends. One reason for the reduced magnetic moments of Ni layers observed experimentally may be an intermixing of Ni and Cu atoms at the interface. However, based on the results of the scanning tunneling microscopy and on the XMCD experiments Lindnerd *et al.*[51] have shown that the intermixing is limited and it does not affect the magnetic moments of 4–5 ML Ni films on the Cu(100) surface. Giving also the fact that our calculations for the trilayers are in very good agreement with the experiment, as well as having in mind discussion in Sec. III B, where we have shown that the presence of Co cannot influence strongly the values of local magnetic moments on Ni atoms, we attribute the observed disagreement between the theory and the experiment in case of a bilayer to the following. As was noted, for example, by Koizumi *et al.*,[52] the intensity of XMCD is proportional to the mean magnetic moment of specified elements projected onto the direction of the incident x rays. Therefore, it is difficult to evaluate the absolute values of local magnetic moments from this type of experiments. Thus, the discrepancy between theoretical and experimental results may be caused by noncollinearity of local moments inside the Ni films, for example, due to the fact that bilayers are not fully saturated.

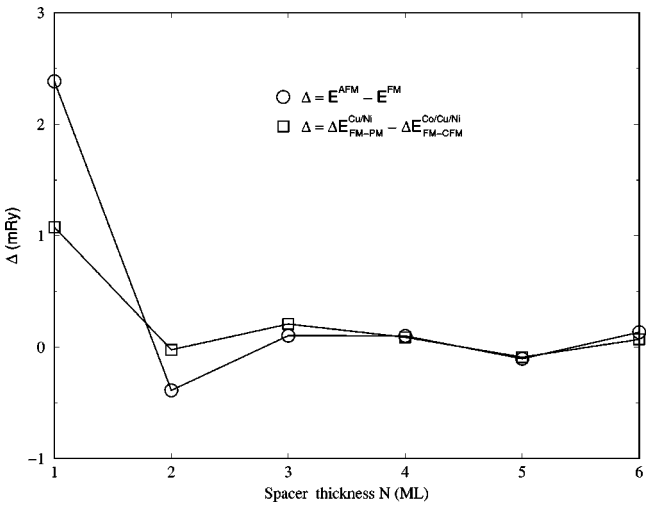

FIG. 5. Interlayer exchange coupling in the $\mathrm{Co_3/Cu}_N\mathrm{/Ni_5}$ trilayer on the Cu(100) surface as a function of the Cu spacer thickness *N*. The difference of total energies between the AFM and FM configurations of spins in the Ni layers with respect to the Co layers is shown by the open circles, and the positive sign corresponds to the FM coupling between the layers. Open squares denote the difference in total energies between solutions with $\mu_{\mathrm{Ni}}=0$ and FM solutions in the bilayer relative to this difference in the trilayer.

This suggestion is also supported by the calculated effective-exchange parameters shown in Fig. 4. It is seen that J_0 of surface and interface Ni atoms are rather small and, therefore, it is easy to rotate these moments from the direction of global magnetization. Therefore, these moments must have stronger tendency towards fluctuations as compared, for example, to the bulk Ni. Our conclusion is in accordance with the conclusions of Ref. 20 where strong magnetic fluctuations in the Ni layers play an important role in the explanation of the experimental observations. At the same time, these fluctuations can be at least partially suppressed in the presence of Co layers due to the interlayer exchange coupling. Note that the experiment was carried out for the spacer

thickness 2.8 ML. The experimental interlayer exchange coupling in this case is in fact close to its maximal value,[17] and may be sufficient to align Ni moments along one direction.

In Fig. 5 we show calculated interlayer exchange coupling for the trilayer as a function of the Cu spacer thickness. In qualitative agreement with the experiment[17] our results indicate that in the case with 2 ML Cu spacer the AFM alignment of magnetic moments between Co and Ni layers is favorable energetically, while in the case of thicker Cu spacers the system can change the magnetic state, and the FM, as well as the AFM alignments between spins in the Co and Ni layers are possible. Note that periods of these oscillations agree with those for the energy differences between the FM and CFM solutions with and without Co cap layers. Finally, we note that Co and Ni layers are FM coupled in the case of a 1-ML-thick Cu spacer, while one may expect the AFM coupling from the extrapolation used in Ref. 17. The obvious reason for this is that Co and Ni still feel each other directly through such a thin spacer, as is also seen from our calculated layer resolved magnetic moments, Sec. III A, and, therefore, RKKY theory cannot be applied for the analysis of this situation.

IV. CONCLUSIONS

In conclusion, we have investigated magnetic properties of the Co/Cu/Ni trilayers and Cu/Ni bilayers on the Cu(100) surface by means of the first-principles Green's-function technique and the fixed-spin-moment method. The averaged magnetic moments $\langle\mu\rangle$ for Co/Cu/Ni trilayers are in good agreement with the experimental results, but there is poor agreement for Cu/Ni/Cu bilayers. We suggest that this could be due to the fact that Ni films are not fully saturated in the bilayers.

We have also studied layer-resolved magnetic moments and effective-exchange parameters of Co and Ni in the trilayers. Total energy of different magnetic configurations were calculated as a function of Cu spacer thickness. We have found two distinct energy differences ΔE in this system, $\Delta E^{\text{FM-PM}}$ and $\Delta E^{\text{FM-CFM}}$, where FM denotes fully self-consistent calculations, while CFM stays for the self-consistent calculations for the Co layers with the constrain that magnetization of Ni layers is zero. The effective exchange parameters in Co and Ni layers are also very different. The existence of these distinct energy scales is in correspondence with two critical temperatures experimentally observed in Co/Cu/Ni trilayers on Cu(100) surface. At the same time, the Co and Ni layers are exchange coupled. This coupling is relatively strong for 1 ML Cu spacer, but decreases and oscillates for larger thickness.

V. ACKNOWLEDGMENTS

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APPENDIX: THE FIXED-SPIN-MOMENT METHOD

In order to understand different aspects of magnetism it is sometimes a great advantage to be able to constrain the magnetic moment to specified values. Experimentally this can be achieved by applying an external field to the sample. In theoretical first-principles calculations the problem is formally to minimize the functional

$$F[n(\mathbf{r}),m(\mathbf{r})]=E[n(\mathbf{r}),m(\mathbf{r})]-\mu\left(\int n(\mathbf{r})-N\right)-\sum_Q h_Q\left(\int m(\mathbf{r})-M_Q\right), \tag{A1}$$

where $E[n(\mathbf{r}),m(\mathbf{r})]$ is the total energy spin-density functional and μ,h are the Lagrangian multipliers with respect to the charge density $n(\mathbf{r})$ and the magnetization density $m(\mathbf{r})$. By minimizing $F[n(\mathbf{r}), m(\mathbf{r})]$, in the case of a non-site-dependent magnetization $h_Q=h$, with respect to the spin-dependent densities $n^{\pm}(\mathbf{r})$, where $m(\mathbf{r})=n^{+}(\mathbf{r})-n^{-}(\mathbf{r})$, we have that

$$\frac{\delta F}{\delta n^{\pm}}=\mu\pm h. \tag{A2}$$

The conventional fixed-spin method[53,54] makes use of this spin-dependent chemical potential using two different Fermi levels in order to fix the number of particles of the two spin channels separately. The disadvantage with this method, which can be shown to be identical to applying a uniform external magnetic field, is that it is not possible to fix the local moments at individual sites. Another possibility of constraining the magnetization is by applying external fields locally at individual atomic sites or layers. In this way we can change the moments of specified regions. However, one major problem in applying an external exchange field is to determine the actual field strength that has to be applied to produce a specified local magnetization density. A solution to this problem is to guess a field and perform the electronic-structure calculation until a self-consistent solution is found with a specific magnetic moment. If the moments are too high or low a new calculation has to be performed with smaller or higher field strengths. The process is repeated until a solution with the desired moment distribution is achieved. If only a single global external exchange field is applied this might possibly be achievable. However, if we want to specify several individual moments the method is impractical. To circumvent this problem we have developed

a fixed-spin-moment scheme where the external field is automatically adapted during each self-consistency cycle of the density-functional minimization in order to achieve an efficient convergence to a solution with a predefined site-dependent magnetic-moment distribution. The external exchange field is balanced according to a so-called PID regulator, which is a standard technique used in control theory.[55] The applied field is determined by three terms: (i) a *P* term proportional to the difference between the actual moment $m_Q(n)$ at a specific site Q and iteration n and the desired moment M_Q,

$$V^P(M_Q,n)=C_P[m_Q(n)-M_Q], \tag{A3}$$

(ii) an *I* term proportional to the integrated difference over all previous iterations,

$$V^I(M_Q,n)=C_I\sum_{i=0}^{n}[m_Q(n)-M_Q], \tag{A4}$$

(iii) and finally the *D* term that is proportional to the differential, i.e., the change of the moment difference between two iterations,

$$V^D(M_Q,n)=C_D\{[m_Q(n)-M_Q]-[m_Q(n-1)-M_Q]\}. \tag{A5}$$

C_P, C_I, and C_D are the proportionality constants that have to be chosen in an appropriate way in order to achieve fast convergence.[56] The spin- and site-dependent one-electron potential, which is applied during the next self-consistency cycle, is given by

$$V_Q^{\pm}(n,M_Q,\mathbf{r})=V_Q^{\pm}(n,\mathbf{r})\pm V^{\mathrm{PID}}(M_Q,n), \tag{A6}$$

where

$$V^{\mathrm{PID}}(M_Q,n)=V^P(M_Q,n)+V^I(M_Q,n)+V^D(M_Q,n), \tag{A7}$$

is the PID regulated external exchange field. $V_Q^{\pm}(n,\mathbf{r})$ is the unconstrained spin-dependent one-electron potential. The presented fixed-spin-moment method above is straightforward to implement and has worked very efficiently in our calculations.

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