Influence of surface roughness and H_2 adsorption on the interlayer coupling in Ni/Cu/Ni trilayers on Cu(001)

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(Received 6 January 2000)

The interlayer coupling strength in Ni/Cu/Ni trilayers on Cu(001) depends strongly on the roughness of the surface of the topmost Ni layer and on the hydrogen coverage. Smoothing of the Ni surface increases mostly the coupling strength of the short period oscillation contribution. Hydrogen adsorption causes an enhancement and a phase change of this short period.

I. INTRODUCTION

The oscillatory interlayer exchange coupling (IXC) between ferromagnetic layers through a nonmagnetic metal spacer attracted considerable attention in the past decade.[1–3] Owing to advances in theory[4–8] and experiment[9–14] it is now well understood, that the interlayer thickness dependence of the exchange coupling is determined by the extremal wave vectors at the (bulk) Fermi energy surface of the spacer material. In case of a Cu(001) spacer layer there are two contributing extremal wave vectors leading to a period of about 5.9 ML (long period) and about 2.4 (short period).[4] This theoretical prediction has been confirmed many times for sandwich systems of Co/Cu/Co/Cu(001),[11,13,15–19] fcc Fe/Cu/Fe/Cu(001),[20] Fe/Cu/Co/Cu(001),[11] and Ni/Cu/Co/Cu(001).[11] While the periods depend only on the spacer material, the strength of the coupling is determined also by the ferromagnetic layer material and its thickness.[9,21,22] The coupling strength was found to depend sensitively on the roughness of the interfaces between the ferromagnetic layers and the spacer layer.[12,23] Also a cap layer influences the IXC.[10,24] This behavior can be explained by the model of Bruno.[6] The strength of oscillatory coupling is determined by the spin dependent reflection and transmission coefficient of the delocalized electrons at each interface in the trilayer structure. Therefore, the resulting coupling is determined by all and not only by the neighboring interfaces of the structure.

A strong dependence of the quantum well state (QWS) energies on the Ni thickness in Ni/Cu/Ni has been observed experimentally by inverse photoemission recently.[25] A single set of QWS was observed with mixed Ni and Cu character, showing that the quantum well states extend through the top Ni layer. In this paper we show, that changes of the surface of the magnetic Ni layer in a Ni/Cu/Ni/Cu(001) trilayer structure strongly affects the IXC. We found, a strong suppression of the short period contribution to the IXC for Ni/Cu/Ni/Cu(001) trilayers with smooth interfaces but a rough *surface*. H_2 adsorption causes an enhancement of the short period contribution with respect to the long period contribution.

II. EXPERIMENT

The Ni films were deposited onto a Cu(001) single crystal having a miscut of less than 0.2° in a molecular beam epitaxy (MBE) apparatus with a base pressure $<4\times10^{-11}$ mbar. The Ni films were grown at 293 K with rate of about 0.6 ML/min. The thickness of the Ni film was determined by means of the medium energy electron diffraction (MEED) oscillations during the growth with a precision of about ±0.1 ML while the pressure was kept below 2×10^{-10} mbar. After the growth of the first Ni film of the trilayer structure the sample was annealed at 450 K for several minutes, which has been shown to smooth the surface considerably without significant intermixing.[26,27] Then a wedgelike Cu film ranging from 4–13.5 ML thickness with a slope of about 1.5 ML/mm was grown at 173 K on top of the Ni film. The growth rate of Cu was calibrated prior to the growth of the wedge by means of MEED oscillations. Therefore, the thickness uncertainty is somewhat larger than that of the Ni films and amounts to about 10%. We have chosen this low growth temperature to avoid the pyramidlike growth of the Cu reported in the literature for room temperature growth.[13,28] The second Ni layer was then grown at 293 K again. The trilayer in this state will be called ''as grown'' throughout the paper. For some investigations the complete structure was annealed at 450 K to smooth the Ni surface.

Since we are interested in the influence of the surface morphology on the IXC, a thin top Ni film should show the biggest effect. However, the measurement of the Kerr effect on very thin Ni films becomes time consuming because of the very low magneto-optical parameter in the dielectric function. For a clean Ni film a spin-reorientation transition occurs at a thickness of 10–11 ML,[29] but covering of the surface with Cu or hydrogen causes a reduction of the thickness of this transition to 7.4 or 7 ML, respectively.[30] To avoid the additional complications of the reorientation transition we have chosen a thickness of 6 ML for both Ni films in the trilayer structure.

To determine the IXC energy *in situ* magneto-optical Kerr effect (MOKE) measurements in the longitudinal geometry with an angle of incidence of about 71.5° was applied. We measured the Kerr rotation at $\lambda=670$ nm. The direction of the external magnetic field was nearly parallel to the surface along the ⟨110⟩ azimuth. The Curie temperature T_C for a 6 ML Ni film is about 370 K.[31] On covering this film with a Cu layer the T_C is strongly reduced. In Ref. 32 a reduction of about 30 K down to 273 K was observed for a 4.3 ML thick

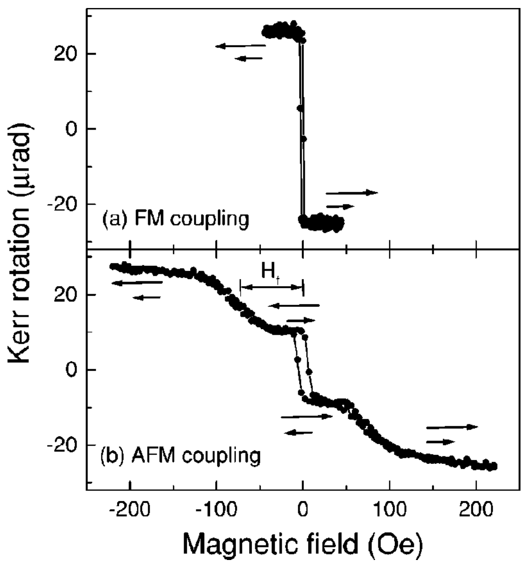

FIG. 1. Kerr hysteresis loop from a 6-ML Ni/*x*-Cu/6-ML Ni/Cu(001) trilayer structure with a Cu interlayer thickness (a) of 5.3 ML in the ferromagnetic coupling range and (b) of 9.4 ML in the antiferromagnetic coupling range measured at 220 K. The arrows represent the magnetization direction of the upper (long arrow) and lower (short arrow) Ni film. Note, despite the same thickness of both films the magnetic moment of the lower film is reduced. H_f indicates the flip field.

Ni film when covered with 2.8 ML Cu. A reduction of the Curie temperature by about 60 K has been found in Ref. 30 for even thicker Ni films. All MOKE measurements in this paper were performed at 220 K, which is well below the Curie temperature of a single Ni film, even when covered with a Cu cap layer.

III. RESULTS

Figure 1 shows the Kerr rotation hysteresis loops measured on a 6.1-ML Ni/*x*-Cu/6.1-ML Ni/Cu(001) trilayer for a Cu spacer thickness of (a) 5.3 ML in the regime of ferromagnetic (FM) coupling and (b) of 9.4 ML in the antiferromagnetic (AF) coupling regime for the ''as grown'' structure. Despite the fact, that both Ni layers have the same thickness, their magnetic moment differs. Covering a Ni film with a Cu layer causes a strong reduction in the Curie temperature[30,33] and magnetic moment.[32] Therefore, it is reasonable to assume, that the upper Ni layer has the higher magnetic moment. This is indicated by the longer arrows in Fig. 1. The Kerr signal from the lower (buried) Ni film is only about 0.46 of that of the upper Ni film resulting in a reduction of the total Kerr signal for AF coupling to 0.37 of that for FM coupling. The coupling energy is of the order of 2 μJ/m^2 at this thickness of 9.4 ML, which is about 60 times smaller than the value for Co/Cu/Co/Cu(001) at the second AF peak at 12 ML.[17] The maximum coupling strength depends critically on the pressure during the evaporation of the Cu interlayer. For only slightly higher CO partial pressure (less than a factor of 2) during deposition of the Cu film we observed a decrease of the flip field H_f to about 40 Oe. Nevertheless, we observed qualitatively the same effects on these structures of lower quality although the magnitude of

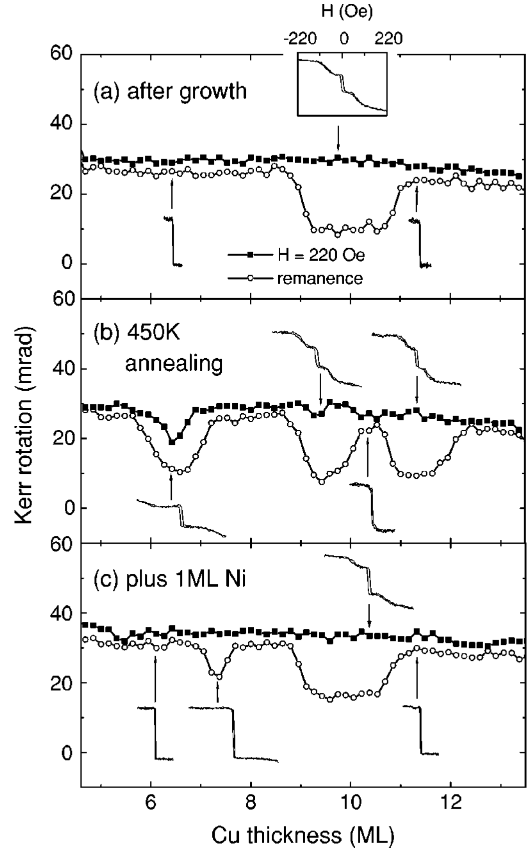

FIG. 2. The Kerr rotation with 220 Oe external field applied (solid squares) and the remanent Kerr signal (open circles) measured (a) after growth at 300 K, (b) after annealing at 450 K, and (c) after growing one additional ML Ni on top at 293 K. All measurements were performed at 220 K. Hysteresis loops for Cu thickness, marked by arrows, are included. They are all drawn to the same scale as the inset on the top.

the changes in the IXC strength upon the different surface treatment, which will be discussed below, were different.

A. Effect of surface roughness on the interlayer coupling

In Fig. 2 the remanent Kerr rotation (M_r) (open circles) and the Kerr rotation with an applied field of 220 Oe (M_s) (solid squares) are plotted versus the Cu interlayer thickness for a 6.1-ML Ni/*x*-Cu/6.1-ML Ni/Cu(001) trilayer structure (a) as grown, (b) after annealing to 450 K, and (c) after growth of an additional 1 ML Ni at 300 K. The insets show the full hysteresis loops for the Cu thickness indicated by the vertical arrows. All hysteresis loops are drawn to the same scale, which is shown for one loop in Fig. 2(a). The regions of AF coupling can be identified in the remanent Kerr signal vs Cu thickness curve by the reduction to ≈ 0.4 of the signal in the FM regions. For the as grown structure there is only one region of AF coupling at about 9–11 ML visible in the investigated Cu interlayer thickness range from 5.5–13.5 ML. As shown by Ref. 26 annealing at 450 K causes a reduction of the mean square roughness by more than a factor of 2. For a thickness of the Ni film equal or less than 6 ML annealing results in an almost perfect flat surface over 100 nm with only a few monatomic islands. This change of the surface morphology causes a change in the IXC as can be seen in Fig. 2(b). Now regions of AF coupling appear at about 6.5, 9.5, and 11.5 ML Cu interlayer thickness. (At 6.5 ML Cu thickness the maximum field of about 220 Oe was not sufficient to align the two Ni layers parallel.) Obviously, smoothing of the surface by annealing has enhanced the short period component of the IXC. The assumption, that interdiffusion at the interfaces occur upon annealing, does not give the right answer because interdiffusion acts in a similar way as interface roughness and causes a *reduction* of the short period contribution.[34]

That the change in the IXC is *not* caused by a modification of the interior Ni/Cu and Cu/Ni interfaces becomes furthermore evident in Fig. 2(c). There the Kerr signals are plotted after an additional 1 ML Ni has been deposited at $T=293$ K onto the annealed film. The region of AF coupling appear now almost in the identical region from about 9–11 ML as for the as grown structure. (The small dip at 7.3 ML does not indicate a significant AF coupling as can be seen in the corresponding hysteresis loop.) We note, that already the deposition of 1/2 ML Ni is sufficient to restore the ''as grown'' distribution of AF and FM regions. However, annealing of this sample after deposition of the additional 1 ML Ni did not cause a change in the AF coupling regions. The annealed 7 ML Ni/x-ML Cu/6 ML Ni/Cu(001) sample showed only one AF coupling region at 9–11 ML in the investigated thickness range.

For the lower quality Ni/Cu/Ni structures no significant change of AF and FM regions was observed upon annealing in the thickness range from 9–11 ML but AF coupling occurred at 6 ML.

B. Influence of H_2 adsorption

In Fig. 3 M_r and M_s vs Cu interlayer thickness of the annealed 6.1-ML Ni/x-Cu/6.1-ML Ni/Cu(001) trilayer are compared with those from the the same sample after exposing it to about 2 Langmuir (L) of H_2 at 220 K. (The exposure was determined from the ion gauge reading without any further corrections.) Hydrogen adsorbs dissociatively on the Ni(001) surface in a fourfold coordinated site.[35] An H_2 exposure of about 2 (uncorrected) L at 220 K was sufficient to saturate the surface. We observed no significant changes for larger H_2 exposure. As it is evident from Fig. 2(b) the hydrogen slightly changes the regions of AF and FM coupling. The AF region at about 6 ML is broadened and shifted towards 7 ML and the AF region at about 11 ML is shifted upwards by 1 ML. This effect of the hydrogen is fully reversible. After desorption of the hydrogen at 330 K and cooling down again to 220 K the initial M_r curve was obtained as can be seen in Fig. 3(c). Obvious changes in the M_r vs Cu interlayer thickness were observed already for H_2 exposures of a fraction of a Langmuir.

The hysteresis loops at about 6 ML and 12 ML show little hysteresis. This may be attributed to the fact, that hydrogen adsorption causes a strong reduction of the Curie temperature by about 70 K down to 290 K.[33] The temperature $T=220$ K, at which the measurements were performed, may be therefore not much lower than the Curie temperature of the trilayer system. In this case a reliable determination of the coupling strength from the measured hysteresis loops is difficult to obtain, since the Curie temperature of exchange coupled layers may be reduced in the regions of AF coupling

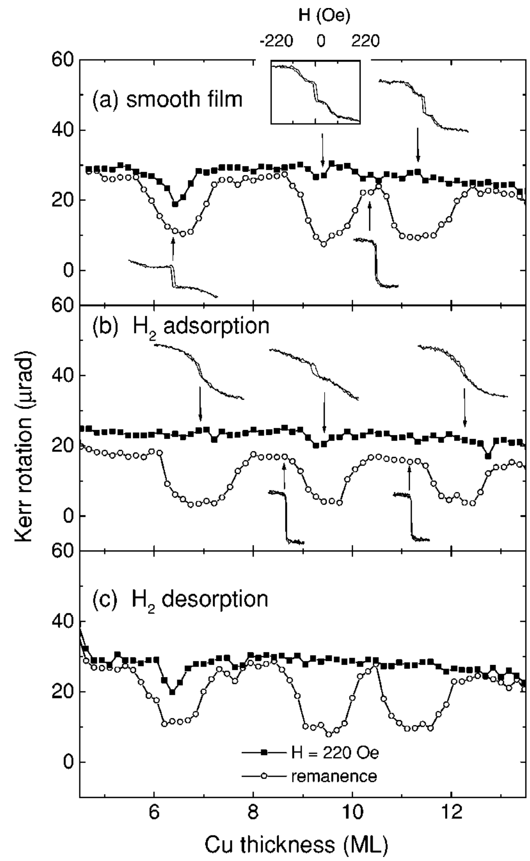

FIG. 3. The Kerr rotation measured with 220 Oe external field (solid squares) and the remanent Kerr signal (open circles) measured (a) after annealing at 450 K, (b) after exposing to 2 L H_2, and (c) after desorption of the hydrogen at 330 K. All measurements were performed at 220 K.

with respect to the FM coupled regions.[36] The reduced remanent MOKE signal at 7 and 12 ML could be caused just by this effect. However, the hysteresis loop at 9.6 ML shows a clear hysteresis, indicating, that 220 K is below the T_C. The Kerr rotation measured with applied magnetic field is reduced by about 20% and the remanent signal by about 60%. Under the assumption that the Kerr signal scales linear with the magnetic moment of the films, this would indicate a reduction of the magnetization of the top Ni film by about 30% upon hydrogen coverage, while the magnetization of the bottom layer is nearly unchanged.

At 11 ML the coupling switches from AF to FM coupling upon H_2 adsorption, proving directly the influence of H_2 adsorption on the IXC. The amplitude of the Kerr loop is essentially the same as that of the uncovered structure at about 10.5 ML. The measurements with the external field applied parallel to the ⟨100⟩ direction gave the same result excluding the possibility of a change of the easy axis of magnetization from the ⟨110⟩ azimuth direction to ⟨100⟩. Also no remanent polar Kerr signal was observed above 6 ML.

It seems, that H_2 adsorption increases the amplitude of the short period component of the IXC relative to the long period contribution. When exposed to H_2 the ''as grown'' sample showed the same additional AF coupling regions at ≈ 7 and ≈ 12 ML as the annealed film after H_2 adsorption although the regions of AF coupling were smaller for the

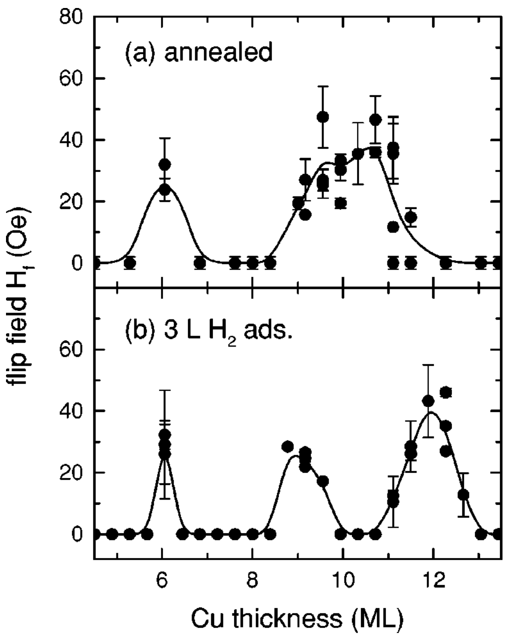

FIG. 4. Flip field H_f vs the Cu interlayer thickness for the lower quality films, (a) after annealing at 453 K, and (b) after exposure to 3 L H_2 at 123 K. The measurements were performed at 123 K.

rough H_2 exposed film. Measurements on the slightly less perfect samples at lower temperatures (120 K) showed a strong enhancement of the short period component of the IXC upon H_2 adsorption. In Fig. 4 the flip field H_f is plotted vs the Cu interlayer thickness for these films, (a) after annealing at 453 K, and (b) after exposure to 3 L H_2 at 123 K. AF coupling was observed at about 6, 9, and 12 ML while at about 10 ML the coupling changed from AF to FM coupling although annealing did not have a significant influence on this structures.

IV. DISCUSSION

The structure of ultrathin Ni films on Cu(001) has been thoroughly investigated by Ref. 26 recently: At room temperature Ni films start to grow in a nearly layer-by-layer mode up to 3–4 ML but trilayer growth becomes dominant at a thickness of 6 ML. For this nominal thickness of 6 ML Ni atoms of the 5th, 6th, and 7th layer form the surface. The relative fractions are 0.26, 0.51, and 0.23. The average island size is of the order of 3 nm. Annealing at 450 K smoothes the surface considerably. The root mean square roughness decreases by more than a factor of two. In Ref. 26 no indication for an intermixing or surface segregation of Cu has been observed. In the literature, however, subsurface growth of Ni has been reported for a Ni film thickness below 3 ML.[37,38] For thicker Ni films, however, we can exclude such an effect from our own investigations.[29,30] In particular, we found a strong change of the magnetocrystalline anisotropy upon coverage of a Ni film with one (or more) monolayers of Cu. Therefore we believe, that no strong intermixing occurred in samples of our investigation and that the buried Ni/Cu and Cu/Ni interface in the Ni/Cu/Ni/Cu(001) structure are not significantly altered upon annealing. Furthermore the fact, that by deposition of an additional fraction of a ML of Ni at low temperatures, which causes only a modification of

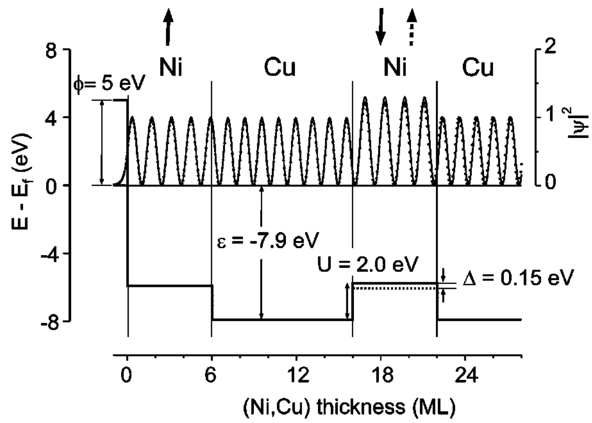

FIG. 5. One-dimensional spin-dependent potential for 6-ML Ni/10-ML Cu/ 6-ML Ni/Cu(001) used for the calculation of the long period contribution to the interlayer exchange coupling. The solid lines represent the state for AF alignment of the two Ni layers and the dotted line for FM alignment.

the surface, apparently the ''as grown'' state can be recovered, supports the view, that the annealing only influences the surface.

The influence of the roughness of the interfaces between the ferromagnetic layers and the spacer layer has been discussed in many publications.[34,39–41] Also the fact, that the IXC depends not only on the spacer layer and the adjacent interfaces to the ferromagnetic layers, but also on the thickness of the ferromagnetic layers,[40,9,42] the presence of a cap layer,[7,10] or by an embedded ferromagnetic layer of another material in one of the ferromagnetic layers[43] has been addressed and was explained in terms of a spin-dependent reflection of the electron waves in the whole layer stack.[42]

In the following we apply the model of Ref. 6 to the present case of a 6-ML Ni/x-Cu/6-ML Ni/Cu(001) trilayer film in its simplest form, the free electron model. In this approximation in the limit of large spacer thickness and weak confinement, the IXC is given by[7]

$$E_F - E_{AF} = 2J_i = \frac{2\hbar^2 k_F^2}{\pi^2 m} \mathrm{Im}\left[e^{2ik_F D} \int_0^\infty d\kappa\, \kappa \Delta r_{Ai} \Delta r_{Bi} e^{-2\kappa D} \right], \tag{1}$$

with D the interlayer thickness, k_F the Fermi wave vector of the spacer material, and the index $i=1,2$ indicates the contribution from the belly and neck of the Fermi surface contour. Δr_{Ai} and Δr_{Bi} are the differences of the reflection coefficients for majority and minority electrons from the top layer and the bottom layer, respectively, including all multiple reflections and are calculated using the potential described below. For the total IXC we took simply $J=J_1 + wJ_2$ ignoring differences in the Fermi surface curvature and group velocity of the band. Instead we introduced a weighting factor w, which accounts for the different relative weight of J_1 and J_2 as a free parameter. In Fig. 5 the potential used for the calculation of Δr_{Ai} and Δr_{Bi} is sketched for the belly contribution to J for 10 ML spacer thickness. $\epsilon = 7.9$ eV corresponds to an extremal wave vector of $k_f = 0.83$ in units of the Brillouin zone boundary (BZ) and to an asymptotic oscillation period of 5.9 ML. The potential barrier $U = 2.0$ eV was estimated from the Fermi wave vectors k_F of bulk nickel: The calculation by Ref. 44 gave for the Fermi wave vector of the majority spin electrons $k_F^{\uparrow} = 0.73$ BZ, by Ref. 45 $k_F^{\uparrow} = 0.78$ BZ, and by Ref. 46 $k_F^{\uparrow} = 0.68$ BZ. Therefore this value is somewhat uncertain and we have chosen a value in this range, $k_F^{\uparrow} = 0.718$ BZ, which best fits our data. For the exchange splitting we took $\Delta = 150$ meV from the exchange splitting of the Δ band at the Fermi energy of Ref. 46. This value is not very critical, because it essentially scales the strength of the IXC without affecting the phase of the oscillation. The resulting wave function $|\psi|^2$ (of the majority electrons of the surface Ni layer) for AF alignment of the two Ni layers (solid line) and for ferromagnetic alignment (dots) is plotted in Fig. 5. Note, despite the relatively large potential step between the Cu and the Ni (compared, for example, to Co/Cu), the transmission coefficient from the Cu interlayer into the Ni layer is still very close to 1. The stronger reflection coefficient in the case of Co/Cu comes from the energy gap in the minority channel, which opens up at about 0.6 eV below E_F and makes the above assumption of weak confinement invalid.[6] For Ni this gap opens at a lower energy of of about -1.0 eV for both, minority and majority electrons. Therefore, from Fe, Co, and Ni the approximation of weak confinement at $k_{\parallel} = 0$ is best fulfilled in the case of Ni. For the short period contribution at $k_{\parallel} \approx 0.52$ BZ there is strong confinement in the case of Co/Cu for the minority electrons. For Ni/Cu neither the majority nor the minority electrons are fully confined to the Cu spacer layer at E_F but the gap may open up already at some 100 meV below E_f.[46] Nevertheless, we used the same weak confinement approximation as for $k_{\parallel} = 0$, with $\epsilon = 4.0$ eV, $U = 1.5$ eV, and the same exchange splitting $\Delta = 150$ meV.

In order to discuss the properties of Eq. (1) in detail, we have to substitute the explicit expressions of Δr_{Ai} and Δr_{Bi}. In the limit of a small Δ Eq. (1) can be approximated to

$$J_i \approx \frac{\hbar^2 k_F^2}{\pi^2 m} \mathrm{Im}\Bigg[r_v \sin(2\Delta k' L_1) e^{2ik'L_1} e^{2ik_F D} \times \left\{ \frac{r_\infty \sin(2\Delta k' L_2) e^{2ik'L_2}}{(D+L_1+L_2)^2} + \frac{i\Delta r_\infty}{(D+L_1)^2} \right\} \Bigg]. \quad (2)$$

L_1 and L_2 are the thickness of the top and buried FM layers, respectively. r_∞ is the (average) reflection coefficient from the barrier between a semi-infinite FM layer and the spacer layer and r_v is the reflection coefficient from the surface.[7] The (average) wave vector $k' = (k_\infty^{\uparrow} + k_\infty^{\downarrow})/2$ in the FM material at E_F is defined by $\hbar^2 k'^2/2m_e = \epsilon_F - V - \Delta/2$. $\Delta k' = (k_\infty^{\uparrow} - k_\infty^{\downarrow})/2$, and Δr_∞ is the difference in the reflection coefficient for spin-up ($\uparrow$) and spin-down ($\downarrow$) electrons, $\Delta r_\infty = (r_\infty^{\uparrow} - r_\infty^{\downarrow})/2$. Note, because the QWS extend through the entire Ni/Cu/Ni trilayer, the coupling J decays approximately as $(D+L_1+L_2)^{-2}$ and not as D^{-2}.

The result of the calculation of J with a weighting factor $w = 1/2$ is shown in Fig. 6 as squares. Theoretically a weighting factor of $w \approx 4$ is expected.[6] The lower value of $w = 1/2$ can be explained by interface roughness. An alternative description with the theoretically expected value $w \approx 4$,

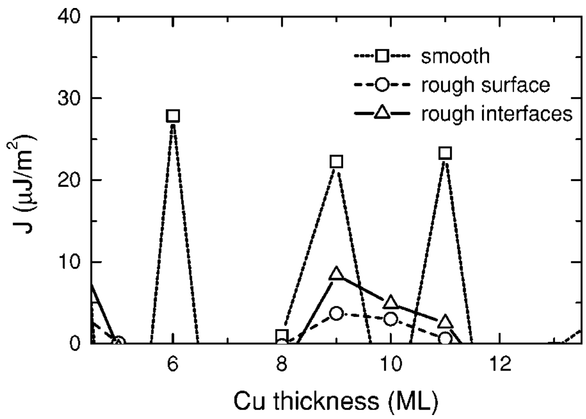

FIG. 6. Calculated interlayer exchange coupling J as a function of the Cu interlayer Cu distance. For the roughness parameters see text. Squares for the flat Ni surface, circles for a rough Ni surface, and triangles for rough interfaces.

but which includes the effect of interface roughness (see below), gives a virtually identical result. The calculation reproduces the observed ranges of AF coupling at 6 ML, 9 ML, and 11 ML for the smooth, annealed film. The coupling strength is too high compared to the experimentally observed values, which may be attributed partly to the neglected residual imperfections of the interfaces and the surface. On the less perfect trilayer structures grown under slightly worse vacuum conditions the change from AF to FM coupling at 10 ML upon annealing was not observed. This observation is in agreement with our simple model, if we assume a reduction of the short period component by a larger residual roughness of the interior Ni/Cu and Cu/Ni interfaces as discussed below.

To simulate the effect of the surface roughness, we took the above mentioned experimentally determined weighting factors of 0.26, 0.51, and 0.23 for the 5 ML, 6 ML, and 7 ML thick fraction of a nominally 6 ML thick Ni film[26] in the ''as grown'' state and calculated an averaged $J_{\mathrm{av}}(D) = 0.26 J_{5\ \mathrm{ML}}(D) + 0.51 J_{6\ \mathrm{ML}}(D) + 0.23 J_{7\ \mathrm{ML}}(D1)$. This is shown in Fig. 6 as circles. The triangles are the result of the calculation assuming an interface roughness with the same parameters as for the surface roughness: $J_{\mathrm{av}}(D) = 0.26 J_{6\ \mathrm{ML}}(D-1) + 0.51 J_{6\ \mathrm{ML}}(D) + 0.23 J_{6\ \mathrm{ML}}(D+1)$. The strength of the short period contribution is reduced by surface roughness more than by interface roughness.

The stronger influence of surface roughness can be easily understood with the aid of Eq. (2). The oscillatory part of the interlayer thickness dependence is entirely contained in the exponential $\exp(2ik_F D)$. Variations in D lead to a much stronger attenuation of the short period contribution compared to the long period contribution: A variation of the interlayer thickness by 1 ML causes a phase shift of about 0.82π, close to antiphase condition, for the short period but only to about 0.34π for the long period contribution. For the surface roughness the variation of J with the Ni thickness has to be considered. The most important contribution comes from the exponential $\exp(2ik' L_1)$. (Since the exchange splitting is small, the thickness dependence of the sin function can be neglected.) In the Ni layer the wave vector $k' = 0.46$ BZ at $k_{\parallel} = 0.52$ BZ which corresponds to a phase shift of about 0.92π, which is even closer to complete destructive interfere than for the Cu layer. The wave vector, $k' = 0.713$ BZ, for the $k_\parallel = 0$ contribution gives a phase shift of about 0.57π. Therefore, the presence of surface roughness in the top Ni layer reduces the long period contribution more efficiently than interface roughness does, but surface roughness causes an even stronger suppression of the short period contribution.

The interlayer coupling is also strongly affected by thickness fluctuations of the buried Ni layer. The dominant first term in the curly brackets of Eq. (2) contains a similar exponential $\exp(2ik'L_2)$, which oscillates rapidly with the thickness L_2 of the buried Ni layer. Therefore a small amount of roughness on either of the interior interfaces causes a strong reduction of the short period contribution, which may explain our finding that for only slightly less perfect growth conditions the short period is suppressed and does not appear after smoothing the surface.

We included roughness only in the simplest form in the above model, not considering the lateral correlations of the thickness fluctuations. However, for the (001) surfaces and relatively thin Ni films it is expected, that this influence J_1 and J_2 in the same way.[47] Recently Wildberger *et al.* calculated the IXC of Ni layers in Cu(001).[48] They found that the reflection coefficients for $k_\parallel = 0$ are very low for majority as well as for minority electrons in agreement with the simple free electron model presented here. For the $k_\parallel \approx 0.54$ BZ contribution the authors of Ref. 48 showed that for a Ni thickness of 1 ML the majority and minority electrons are not confined to the Cu layer as well. However, for the minority electrons the amplitude of the reflection coefficient increases rapidly and oscillates with increasing thickness. This leads to a strong thickness dependence of the short period contribution as a function of the Ni thickness. In the free electron model the factor $\exp(2ik'L_1)$ accounts for this strong thickness dependence partially. The result of Ref. 48, however, that for thick Ni films the minority electrons are close to total confinement while the reflection coefficient for the majority electrons remains small, is not contained in our model and may introduce additional phase shifts. Further investigations are necessary to clarify the effect of roughness in this case.

The effect of H_2 adsorption cannot be explained by the hydrogen induced changes of the work function of 0.17 eV.[49] The phase shifts introduced by an increased work function are much to small to have any significant influence on the range of AF and FM coupling regions. In a theoretical work of Ref. 50 it was found that hydrogen adsorption on Ni(001) surfaces strongly reorders the Ni surface and leads to an increase of the island size. However, it is unlikely that such an effect contributes significantly to the observed change in the interlayer coupling upon H_2 adsorption, because these effects are fully reversible, when the hydrogen is desorbed again. We also mention, that an increase of the Ni island size without any interlayer transport would not cause a change in the interlayer coupling. At least in the model presented here only the vertical roughness, i.e., thickness variations of the Ni film have an influence on the interlayer coupling.

The observed shifts of the AF and FM coupling regions would correspond to an additional phase shift of the minority and majority electrons contributing to the short period oscillation of about $\Delta\Phi_2 = \pi/2$ upon reflection from the surface. This phase shift could be caused by an upward shift of the corresponding energy bands near the surface. However, H_2 adsorption does not only cause this shifts of AF regions but the strength of the short period is considerably enhanced with respect to the long period contributions. This effect may be explained by the change in confinement of the minority electrons at $k_\parallel \approx 0.54$ BZ upon hydrogen exposure: Hydrogen adsorption delocalizes the the surface states of Ni.[51] Particularly, in Ref. 51 it was found, that upon hydrogen exposure the $\overline{\Gamma}_4\overline{\Delta}_2\overline{X}_4$ band looses its surface character in a wide range around the middle of the $\overline{\Delta}$ line. Therefore this state contributes more to the delocalized quantum well states which may lead to an enhancement of the short period contribution of the IXC.

Finally, we note that the apparent coupling strength as measured by the flip field H_f depends not only on the interlayer coupling strength J but on the magnetic moment of the Ni films as well. Since this moment is influenced (reduced) by the adsorption of hydrogen, this may cause an overall change in the flip field. However, for the observed shifts in the AF coupling regions the relative strength of the short and long period contribution must change.

V. CONCLUSION

We have shown in this paper that the interlayer coupling in 6-ML Ni/Cu/6-ML Ni/Cu(001) depends not only on the thickness of the intermediate Cu layer and the smoothness of the adjacent Cu/Ni and Ni/Cu interface but also strongly on the properties of the Ni surface. A rough surface significantly reduces the coupling strength of the short period oscillation relative to the long period contribution. H_2 adsorption enhances the short period contribution considerably.

ACKNOWLEDGMENTS

One of the authors, Y.Z.W., acknowledges the MPI Halle for financial support during his stay. He also acknowledges the partial support from the National Natural Science Foundation (Grant Nos. 19625410 and 19734002). This work was supported in part by the EC through Grant No. ERB-EMRX-CT96-0015 (TMR NOMOKE).

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