PHYSICAL REVIEW B VOLUME 61, NUMBER 2 1 JANUARY 2000-II

Dependence of the Curie temperature on the Cu cover layer in *x*-Cu/Fe/Cu(001) sandwiches

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(Received 1 June 1999)

A strong reduction of the Curie temperature T_C has been observed for room-temperature-grown fcc Fe films on Cu(001) when covered with 1 monolayer (ML) Cu for all Fe thicknesses up to the fcc-bcc transition of the Fe film at ≈11 ML. At 2 ML Cu coverage this decrease of T_C partially recovers and approaches a constant lower value on further increasing Cu coverage. The correlation of this observed magnetic behavior with electronic and possible structural changes of the Fe film upon Cu coverage is discussed.

I. INTRODUCTION

The structural as well as the magnetic properties of fcc Fe films on Cu(001) and Cu/Fe/Cu(001) sandwiches have been investigated extensively during the past.[1–22] The reason for this wide interest is the rich variety of ferromagnetic, antiferromagnetic, or even more complex magnetic structures of fcc iron, which can be reached by small changes of the unit-cell volume[22a] or the tetragonal distortion.[23,24] Experimentally, fcc iron can be stabilized at room temperature only as small precipitates in a Cu matrix, where it is found to order antiferromagnetically below the Néel temperature of about 67 K.[25] It can also be prepared as ultrathin films grown on the Cu(001), $Cu_3Au(001)$,[26,27] Ni(001),[28,29] or the fcc Co(001) (Refs. 28 and 30) surface. Mostly the results are discussed in terms of the magnetic properties of the Fe film alone. The effect of the substrate was considered mainly as a template fixing the (in-plane) lattice spacing to the desired value. This view was supported by the experimental observation, that independent of the substrate (Cu, Ni, or Co) the same sequence of magnetic phases as a function of the Fe thickness were observed.[28] However, there is a direct interaction of the substrate or a nonmagnetic cover layer with the Fe film, which may significantly change the magnetic properties of the Fe film. In this paper we show that a cover layer of Cu on an Fe film on Cu(001) has a strong and complex influence on the Curie temperature (and therefore presumably also on the magnetic moment) of the Fe film.

Experimentally, it turned out that the magnetic properties of the Fe film depend strongly on the preparation method. Two standard preparation methods have been used mainly, either molecular beam epitaxial (MBE) growth of Fe at room temperature[31–39] or growth at low temperatures $(T \approx 100\,\mathrm{K})$.[40–42] In some investigations Fe films were prepared at temperatures significantly above room temperature $(T > 350\,\mathrm{K})$.[43,44] However it is now generally accepted that at this high temperature the Fe film is not stable (at least for thinner films) and it is covered partially by Cu.[10,37,45] The reason for the observed differences between room-temperature-grown Fe films and low-temperature-grown films are not so clear. Partially the magnetic and structural properties may be affected by the different morphology of the Fe films. While the room-temperature films grow nearly in a layer-by-layer mode, for growth at low temperatures the roughness is very much enhanced and morphological changes of the surface occur with increasing thickness.[14] In addition, these low-temperature-grown films may also be affected by adsorption of residual gas, such as hydrogen (which sticks on the surfaces only at these low temperatures) during the deposition.[46]

Therefore we consider room-temperature-grown Fe films as those, which are best characterized and which are closest to the idealized systems used in the theoretical models. For Fe films prepared in this way the following properties were found: At low Fe thickness below 4 monolayers (ML) (phase I) the Fe film is tetragonally distorted. The interlayer Fe distance is expanded by 5% to 1.87 Å.[12,13] At a thickness larger than 4 ML (phase II) the interlayer distance of the Fe film relaxes in its interior nearly to the value of ideal cubic symmetry. Only the interlayer distance of the first two layers remain expanded.[6] Parallel to the structural change the average magnetization of the Fe film drops to a value roughly equal to that of 2 ML Fe of phase I. Detailed experimental[11,16,31,36,38,47] and theoretical[48–52] investigations indeed revealed that the magnetic moment of the first and second layer couple ferromagnetically, while the deeper layers are antiferromagnetically aligned at temperatures lower than 200 K.[36] At a thickness of about 11 ML's the fcc Fe film transforms into a bcc phase (phase III).[7,32,34] Experimentally it is found that the easy axis of magnetization is perpendicular to the surface both for phase I and II and switches in plane only in the bcc phase III.

The Cu covered Fe films are less well investigated.[53–57] It was found that a 3-ML Fe film has nearly the same enlarged average magnetization as the uncovered film.[9] Qualitatively the same two magnetic phases, one at low Fe thickness having a high magnetic moment and one at thicknesses larger than 4 ML having a low magnetic moment is found as for the uncovered Fe film.[57]

In the present study we use the magneto-optical Kerr effect (MOKE) to determine the Curie temperature of uncovered fcc Fe films and Cu/Fe/Cu(001) sandwich systems. We describe in Sec. II the experimental setup and the results are presented in Sec. III. In Sec. IV we address the question of the origin of the observed unusual behavior of the Curie temperature with the thickness of the Cu cover layer. The paper ends with the conclusion in Sec. V that this effect probably cannot be explained entirely as a magnetovolume effect caused by the Cu overlayer induced structural changes. Direct electronic interaction of the Cu layer with the Fe film may be more important for the observed change in the magnetic behavior.

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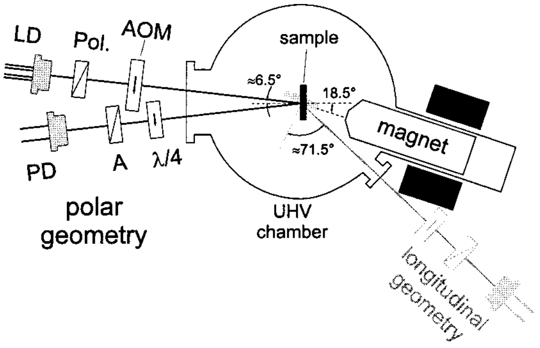

FIG. 1. Schematic view of the experimental Kerr setup. LD: laser diode ($\lambda = 670\,\mathrm{nm}$); Pol.: dichroic sheet polarizer; AOM: acousto-optical modulator; λ/4: quarter-wave-retardation plate; A: dichroic sheet polarizer; PD: photodiode.

II. EXPERIMENT

Fe films of constant thickness as well as wedgelike samples were grown at $T = 298\,\mathrm{K}$ on a Cu(001) single crystal in a molecular beam epitaxy (MBE) apparatus (base pressure $< 4 \times 10^{-11}\,\mathrm{mbar}$). Since it has been shown that these fcc Fe films are unstable when heated to temperatures considerably higher than room temperature,[37,58] the Fe films were kept below 313 K during the whole measuring period. The flux of the Fe *e*-beam evaporator was calibrated by means of medium-energy electron diffraction (MEED) prior to the growth of the wedges. The thickness of the single Fe films were controlled directly by MEED. The growth rate was about 0.8 ML/min at a pressure of less than 2×10^{-10} mbar during evaporation. On top of the Fe films a Cu wedge was grown under the same conditions as for the Fe film. In the Kerr imaging experiment described below a double wedge structure was prepared by deposition of a Fe wedge and subsequent azimuthal rotation of the sample by $74 \pm 1°$ and deposition of the Cu wedge. The thickness of all wedges were cross checked by Auger analysis after completion of the measurement. From all that we estimate that the absolute error in the thickness calibration of the single layers is below 0.2 ML. For wedges it is below 0.2 ML plus 10% of the thickness of the wedge.

The setup for the MOKE measurements is shown in Fig. 1. A lock-in phase modulation technique has been used. The resulting photodiode signal at the modulation frequency is approximately proportional to the Kerr ellipticity change. Two different geometrical setups have been used which we name polar and longitudinal geometry in this paper. For the polar geometry the angle between the incident light beam and the sample normal was about 6.5°. The axis of the dipole magnet was at an angle of about 18.5° with respect to the sample normal parallel to the optical plane. For the measurements in the longitudinal geometry the sample was rotated. There the angle between incident light beam and the sample normal was about 71.5°. For this geometry the axis of the magnet was nearly parallel to the surface of the sample. The maximum external field which could be reached at the sample was about 300 Oe.

For the *in situ* Kerr imaging the same setup as described in Ref. 59 was used (see Fig. 1 of Ref. 59). The sample was illuminated with linear polarized light. The incident angle was about 20° with respect to the surface normal. The light reflected from the sample passed a polarization analyzer. A special ‘‘long-distance’’ microscope objective forms an image of the crystal onto the chip surface of a charge-coupled device (CCD) camera. Images for opposite magnetization either in the remanent state or with an external magnetic field of about 300 Oe normal to the surface were taken for the analyzer set close to maximum extinction.

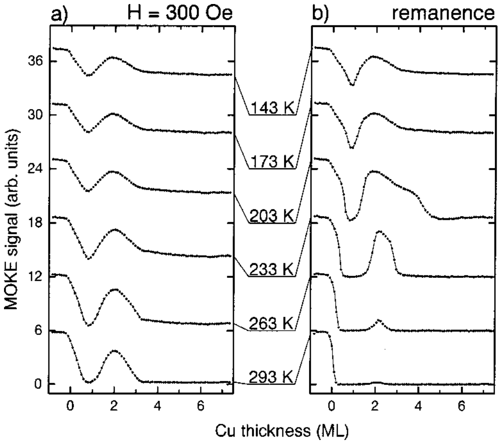

FIG. 2. Polar MOKE signal from a x-Cu/3-ML Fe/Cu(001) sandwich structure as a function of the Cu cover layer for a selected number of temperatures. (a) MOKE signal with an applied external field of 300 Oe; (b) MOKE signal in remanence. The curves are offset by 6 arbitrary units with respect to each other.

III. RESULTS

Figure 2 shows the MOKE signal from a x-Cu/3-ML Fe/Cu(001) sandwich obtained in the polar geometry (see Fig. 1) as a function of the Cu cover layer thickness for a selected number of sample temperatures. In Fig. 2(a) the MOKE signal measured with an external field of about 300 Oe is shown while in Fig. 2(b) the same is plotted for the remanent MOKE signal. In both cases a deep minimum at about 1-ML Cu coverage followed by a maximum at about 2 ML can be seen. No significant temperature hysteresis is observed in the measurements taken at increasing and decreasing temperature. For this 3-ML Fe film the external field of 300 Oe was sufficient to magnetically saturate the film for all investigated temperatures and thicknesses of the Cu cover layer.

Going to Fe films thicker than 4 ML a structural and a magnetic phase transition occurs as discussed in the introduction. Figure 3 shows the MOKE signal from a x-Cu/7-ML Fe/Cu(001) sandwich in this second phase of the Fe layer. Here the Curie temperature of the uncovered Fe film, $T_C \approx 280\,\mathrm{K}$ is much lower than that in phase I. Therefore at the highest temperature of 293 K the MOKE signal with applied field is already significantly reduced and no remanent Kerr signal is detected. However, at lower temperatures qualitatively the same as for the 3-ML Fe film is observed. At temperatures lower than 170 K the maximum external magnetic field of 300 Oe was not sufficient to reverse the magnetization of the uncovered 7-ML Fe film due to the much larger coercive field H_c in the phase II range compared to the thinner Fe films in phase I. For a given temperature H_c decreases upon Cu coverage and therefore the magnetization of this Cu covered film can be reversed

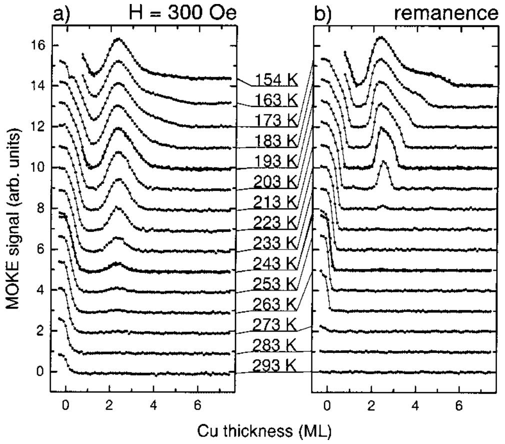

FIG. 3. Same as Fig. 2 for a x-Cu/7-ML Fe/Cu(001) sandwich structure. The curves are offset by one arbitrary unit with respect to each other. [The very small negative MOKE signal in (a) at large Cu thickness is partly caused by a small longitudinal Kerr contribution as the external field is applied at an angle of 18° to the surface normal and partially by a Faraday effect from the UHV windows.]

with the 300 Oe external field.

We note that this decrease of H_c at constant temperature is caused mainly by the strong drop of the Curie temperature (see below). For the 3-ML film at low temperatures, however, we have seen an initial increase of H_c upon small Cu coverage while it also drops on thicker Cu cover layers. This is shown in Fig. 4 on MOKE hysteresis loop from a 3-ML Fe film at 160 K. H_c for the uncovered film is about 75 Oe. A small coverage of 0.4 ML Cu causes H_c to increase to about 120 Oe. At 1 ML Cu coverage H_c is very small but this is accompanied with a strong reduction of T_C (see below). In addition the shape of the hysteresis loop deviates significantly from the nearly rectangular shape seen for the uncovered film and for the thicker Cu coverages. The H_c for Cu coverages thicker than approximately 2 ML is nearly constant and at a lower value of 30–40 Oe compared to that of the uncovered Fe film.

From the measured MOKE curves vs Cu thickness shown in Figs. 2 and 3 the MOKE signal vs temperature curves shown in Fig. 5 were derived. While the Curie temperature T_C for the uncovered 3-ML film is larger than the maximum temperature of 313 K used in the experiment, for all Cu covered Fe films the remanent MOKE signal drops to zero below 300 K as shown in Fig. 5(a). For 1 ML coverage the remanent signal disappears at a temperature T_r of about 200 K but for a Cu coverage of 2 ML this happens at a higher temperature of about 260 K. At even thicker Cu cover layers T_r goes gradually back to 200 K. Qualitatively, the same can be observed for the temperature dependence of the remanent MOKE signal of the thicker Fe film of 7 ML shown in Fig. 5(b). Here the temperatures of vanishing remanence are generally lower but a minimum in T_r is again observed at 1 ML Cu coverage.

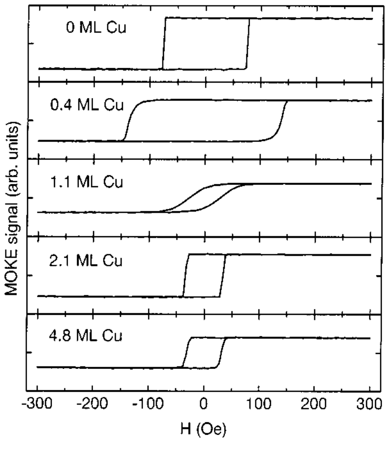

FIG. 4. MOKE hysteresis loops in the polar geometry from a 3-ML Fe film on Cu(001) at $T=160$ K for Cu coverages from 0 to 4.8 ML (top to bottom).

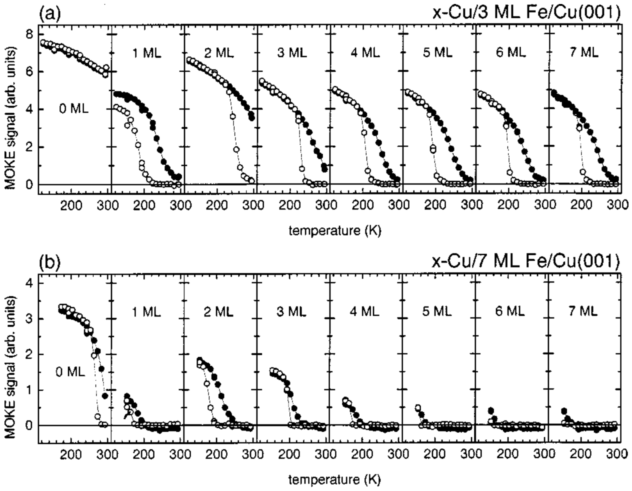

FIG. 5. Polar MOKE signal from a x-Cu/3-ML Fe/Cu(001) (a) and x-Cu/3-ML Fe/Cu(001) (b) sandwich structure versus the sample temperature for an uncovered 3-ML Fe film (left) and with a Cu cover layer of 1 to 7 ML thickness. The solid (open) symbols represent the measurement with an (with no) external field of $H=\pm 300$ Oe applied.

The temperature at which the remanent magnetization vanishes, however, does not indicate the Curie temperature of the film. It has been shown in the case of Ni films on Cu(001), for example, that the observed sharp drop of the remanent magnetization is caused by domain formation at temperatures below T_C.[60] The solid symbols in Fig. 5 represent the MOKE measurement with an applied external field of 300 Oe. The difference between T_r and the inflection point of the saturation measurements amounts up to ≈50 K for the 3-ML Fe film in Fig. 5(a) and less than 20 K for the 7-ML film. The true T_C can be obtained by extrapolating $M(H)$ down to zero external field. However, because of the insufficient homogeneous external magnetic field in our experimental setup we did not attempt that. Instead in Fig. 6 the temperature T_r (open symbols) and the temperature T_s (filled symbols), at which the MOKE signal with applied magnetic field of 300 Oe dropped to 20% of the maximum value, is plotted. From the shape of hysteresis loops at different temperatures we are convinced that $T_r<T_C<T_s$ with the latter one, T_s, quite close to the real Curie temperature. The choice, 20% of the maximum value, is not very critical for the determination of T_r since the remanent Kerr signal drops very rapidly near T_r. For the MOKE signal with applied field the 20% of the maximum value are close to the inflec-

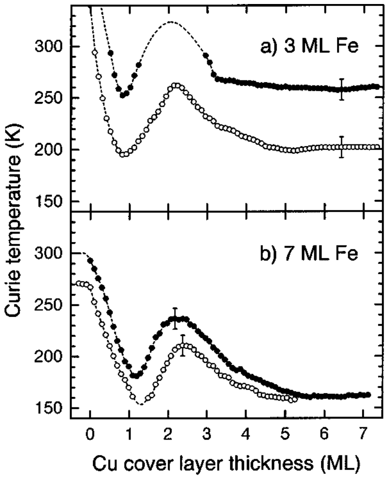

FIG. 6. Curie temperature of Cu/Fe/Cu(001) sandwiches as function of the Cu cover layer thickness derived from the data shown in Figs. 2 and 3. The solid symbols represents an estimate of the Curie temperature derived from the Kerr data at 300 Oe external field as explained in the text. In the thickness range from about 1.5 to 3 ML the dashed curve in (a) results from an extrapolation of the measured Kerr data below 313 K to higher temperatures. The open symbols represent the temperature T_r at which the remanent Kerr signal disappeared.

tion point of the MOKE vs temperature in the investigated temperature range. We estimate the error by this procedure to be less than ±10 K.

A very similar behavior of T_C vs Cu cover layer can be seen in Fig. 6(b) for the 7-ML Fe film despite the fact that the magnetic ordering in the uncovered 3-ML Fe film and the 7-ML Fe film is vastly different. Remember, for the (uncovered) 7-ML Fe film the Fe layers are coupled partially antiferromagnetically leaving only two ferromagnetically coupled ''live layers'' at the surface while the 3-ML film is completely ferromagnetically ordered.[31] The lower T_C for the 7-ML film therefore is not surprising. However, for the 7-ML Fe film upon Cu coverage one may expect a complete reordering of the magnetic alignment of the individual Fe layers to a more symmetric structure.[52] The very similar course of T_C vs Cu coverage for the 3- and 7-ML Fe film seems to indicate that the change of the Curie temperature is mainly effected by the changes at the surface or Cu/Fe interface.

In a recent paper we demonstrated that Kerr imaging can be used with advantage for MOKE measurements on wedge-like samples.[59] Figure 7 shows a color map of the Curie temperature (precisely, the temperature T_s) derived from Kerr images of a Cu/Fe/Cu(001) double wedge prepared as described in Sec. II. (While the Fe and Cu wedge were grown at an angle of 74° with respect to each other the data shown in Fig. 7 are transformed to Cartesian coordinates.) At about 4.5 ML Fe thickness our maximum magnetic field was not sufficient to reverse the magnetization even for temperatures close to T_C. Therefore we might have underestimated T_C in the regions marked with the color ''below 130 K.'' However, it is clearly seen that for *all* Fe thicknesses below the fcc-bcc transition at about 10–11 ML the Curie temperature is strongly reduced upon Cu coverage and a maximum T_C occurs at about 2 ML Cu coverage despite the fact that the Fe film changes its magnetic structure at about 4 ML thickness. The magnetically most stable configuration in this plot above 0 ML Cu coverage is that of a 2 ML Fe film covered with a 2-ML thick Cu film. Figure 7 suggests that a Cu covered Fe film of about 5 ML's and much less pronounced (and barely visible in Fig. 7) of 8 ML thickness is magnetically especially stable while a 6-ML film shows the lowest Curie temperatures. For a 1-ML Cu coverage one can see that for all integer ML of Fe thickness the Curie temperature is above 170 K with the exception of the 6-ML film. Between complete layers the Curie temperature drops below 130 K. Above 11 ML the Fe film transforms into the bcc phase, which again has a T_C far above 313 K. The magnetization for this phase is in the plane. Because of the angle of incidence of 20° a small longitudinal Kerr signal from the in-plane magnetization is detected which makes it possible to determine T_C in the bcc phase as well.

Because in Figs. 2–7 mainly the perpendicular component of the magnetization was measured one may argue that only this component vanishes at the temperature determined in Figs. 2, 3, and 7. To exclude this possibility we measured the MOKE signal in the longitudinal geometry as indicated in Fig. 1 as well. The result is shown in Fig. 8. Generally, due to the high refractive index of metals the Kerr signal from the normal component of the magnetization is much larger than the Kerr signal caused by the in-plane component of the magnetization. For the polar geometry in Fig. 1 this is approximately a factor of 30 and for the longitudinal geometry with an angle of incidence of about 71.5° this is still a factor of approximately 4. The direction of the external magnetic field in this geometry is nearly parallel to the surface plane. There is, however, a small component of the magnetic field perpendicular to the sample surface, which is sufficient to reverse the magnetization close to T_s. The MOKE signal from this polar component of the magnetization is seen in Fig. 8. The remanent signal drops to zero at exactly the same temperatures as in Fig. 5(a) for the polar geometry. There are differences in the MOKE curve measured with external field, because in the longitudinal geometry the field component normal to the surface is much smaller. Nevertheless, in no case is a MOKE signal measured at a higher temperature as in the polar geometry. We conclude that there is no in-plane magnetization above T_r or T_s.

As mentioned in the introduction, the magnetic transition from phase I to phase II of the uncovered Fe film is accompanied by a structural transition. To test if this occurs as well on Cu coverage of the Fe film we did IV-LEED measurements of the specularly scattered electron beam. The result for a 3-ML Fe film is shown in the top panel of Fig. 9 for the beam energy range of 100–600 eV. The angle of incidence for the electron beam was 6°. The numbers 4 to 7 indicate the position of the kinematical Bragg peaks assuming an interlayer distance of 1.87 Å.[13] In the experimental spectrum a pair of split peaks is observed around each of these Bragg peak positions. In previous publications the positions of the maxima of such pairs have been utilized to estimate the tetragonal distortion of the Fe film with respect to the Cu substrate.[18,20,61] For example, for the fifth-order kinematical peak the spectrum exhibits two maxima at about 247 and 278 eV. The peak at the lower energy of about 247 eV is a signature of an expanded interlayer distance of the Fe layer

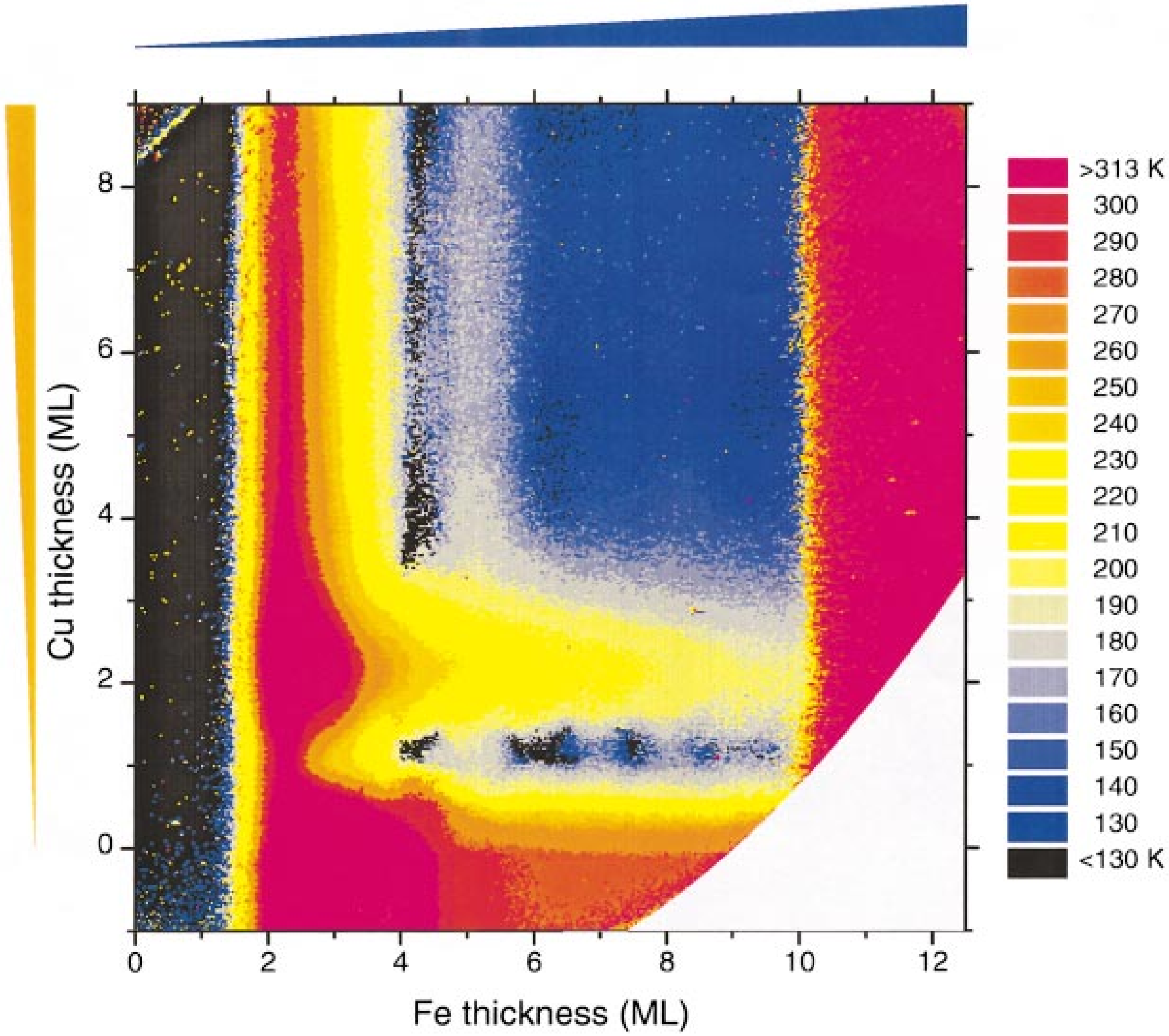

FIG. 7. (Color) Color map of the Curie temperatures from a Cu/Fe/Cu(001) double wedge. The Kerr data for this image were obtained by a Kerr-imaging setup as described in Ref. 59 with a maximum external field of $H=\pm 300$ Oe. The Curie temperature in the light gray area in the lower right corner is not determined because these points correspond to coordinates outside of the crystal. (While the Fe and Cu wedges were grown at an angle of 74° with respect to each other, the data in this figure are transformed to an orthogonal coordinate system.)

and can be seen throughout phase I. This peak disappears in phase II because the interior of the Fe film relaxes to the fcc lattice spacing of the Cu. The single expanded top Fe layer does not produce any sharp peaks in the IV-LEED curve. The energy region around this fifth-order kinematical peak is repeated in the bottom panel of Fig. 9 on an expanded energy scale. On top of the 3-ML Fe film a Cu wedge was grown and the IV-LEED measurements obtained at different positions on this wedge, corresponding to different thickness of the Cu cover layer, are plotted in the bottom panel of Fig. 9 as well. When covered with 1 ML Cu the peak near 247 eV disappears. One may be tempted to interpret this as a structural transition of the film into the fcc phase. However, no recovery of the low energy peak is observed for the 2 ML covered Fe film as one would expect if a structural change of the Fe layer is responsible for the increase of T_C at 2-ML Cu coverage after the drop at 1-ML Cu coverage. There is no parallelism between the MOKE data and the structural IV-LEED data. However, it must be considered that covering the Fe film with a Cu layer introduces an additional scattering layer with a different interlayer spacing. One can convince oneself easily by a simple calculation within the kinematical scattering approximation that this completely changes the IV-LEED curve which becomes very similar to that of the Cu substrate in agreement with a dynamic LEED calculation. Without an extensive complete IV-LEED structural analysis no quantitative information about the interlayer distance of the Fe film can be obtained. However, by looking at the position of the higher energy peak at about 278 eV in

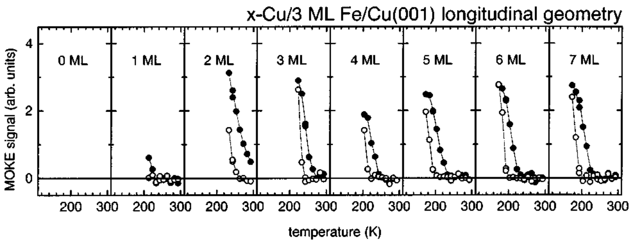

FIG. 8. Same as Fig. 2 in the longitudinal Kerr geometry. Due to the insufficient external field component normal to the surface in this geometry the magnetization direction of the Fe film could not be reversed below ≈180 K.

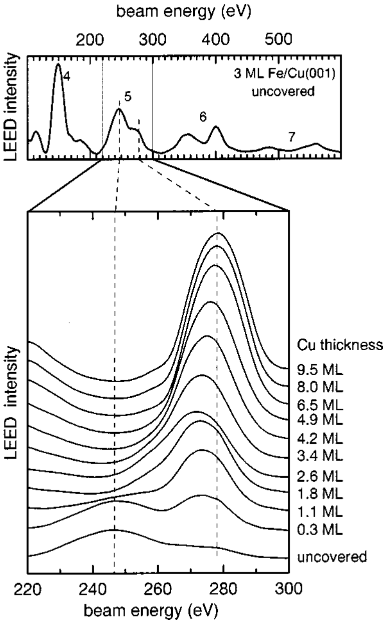

FIG. 9. Top panel: IV-LEED intensity curves for the specularly scattered electron beam from a 3-ML Fe film on Cu(001) measured at $T=150\,\mathrm{K}$. The numbers 4 to 7 indicate the order of the ‘‘kinematical peaks’’ at 173, 278, 407, and 558 eV expected for a 1.805-Å interlayer spacing and an inner potential of 9 eV. Bottom panel: The same curve as in the top panel on an expanded energy scale (bottom curve) together with the IV-LEED curves of the same 3-ML Fe film for increasing thickness of a Cu cover layer. The dashed lines indicate the two maxima of the split fifth-order ‘‘kinematical peak’’ of the uncovered 3-ML Fe film.

Fig. 9 one sees that its maximum first shifts towards lower energies for increasing Cu coverage before at about 2 ML’s the peak shifts towards higher energies again and finally reaches the peak position of the Cu substrate. The observation of a minimum energy for the peak position around 2-ML Cu coverage is opposite to the effect one would expect in a simple kinematical picture if only an additional layer with the Cu bulk interlayer distance is added to the film. An average Fe interlayer spacing enlarged by less than 0.01 Å with respect to that of the Fe layer covered with 1 ML Cu would result from a kinematical analysis of the observed inward shift of about 2 eV. A dynamic LEED model calculation leads to a similar number. Such energy shifts have been observed for other Fe thicknesses as well.[62] We conclude that there are only minor structural differences in the Fe layer covered with 1 ML or covered with 2 ML Cu.

There are definite strong structural changes of the Fe film when covered with 1-ML Cu compared to the uncovered Fe film: Depending on the thickness a 4×1, 5×1, and 2×1 (Refs. 6, 12, and 13) superstructure is observed for the uncovered Fe films which we could not detect after covering the film with Cu. However, these superstructures disappear already on small Cu coverages and never reappear again for thicker Cu cover layers.

IV. DISCUSSION

A similar behavior of the MOKE signal upon Cu coverage was already observed by Swartzendruber *et al.*[63] on low-temperature-grown 6-ML Fe film on Cu(001). For these low-temperature-grown films the Fe exhibited in-plane magnetic anisotropy. This changed when covered with 1 ML Cu to perpendicular magnetization in connection with a strong drop of the saturation MOKE signal by more than a factor of 6 at 150 K and an increase of the MOKE signal by a factor of 2 with respect to the value for the 1 ML covered film when covered with 2 ML of Cu. Several possible reasons have been invoked for the observed behavior: (1) There might be structural changes of the Fe film causing simultaneously a transition into a nonmagnetic or antiferromagnetic state when covered with Cu (magnetovolume effect). (2) The electronic interaction of the Cu causes only a change of the magnetic state of the Fe film without affecting significantly the structure of the Fe film. (3) Finally, the quantum well states of the Cu overlayer may cause especially strong interaction with the *d* states of the Fe in the range of 1–4 ML’s. In the following we discuss the relevance of these three points in detail.

A complete reordering of a 3-ML-thick Fe layer upon coverage with 10 ML Cu has been reported by Magnan *et al.*[44] The analysis of the extended x-ray-absorption fine structure (EXAFS) spectrum of the uncovered Fe film reveals a highly reconstructed film in agreement with the LEED analysis of Ref. 12. On the other hand, the Cu covered 3-ML Fe shows the signature of a well-ordered fcc structure. In Ref. 44 a 3-ML Fe film grown at 370 K was investigated as well from which we can assume now that it is partially (fraction of a ML) covered with Cu.[37,22] This film also showed the same EXAFS spectrum as that of the 10 ML Cu covered Fe film. From that we conclude that for 1 ML as well as for thick Cu overlayers the 3-ML Fe undergoes structural changes in the direction of a more uniform fcc structure. Unfortunately, there are presently no EXAFS or IV-LEED measurements for a 2-ML-thick Cu overlayer. Only our own observation of the small peak shift in the IV-LEED spectrum in Fig. 9 between 1 and 2 ML Cu coverage may indicate a (small) change in the Fe unit-cell volume of the order of 0.5% from which we feel that this does not sufficiently change the magnetic moment. Assuming a linear dependence of the Kerr signal with the magnetic moment of the Fe film an increase of the magnetization of the Fe film covered with 2 ML Cu of the order of 40% with respect to that covered with 1 ML Cu can be estimated from the data in Fig. 5. We cannot rule out the possibility of a spectroscopical enhancement as observed in the overlayers of Au on Co(0001) (Ref. 64) and Au/Fe(001).[65]

There is no reason to assume only a uniform change of the whole Fe layer upon Cu coverage. In fact, in several publications on Cu/Fe/Cu(001) multilayers a two-phase model is assumed.[55,57] The EXAFS study of Ref. 57 found a strong correlation between the Fe-Fe bond length and the magnetic moment of the Fe. The transition from phase I to phase II of the uncovered film has been discussed in this picture recently.[21,66] It could be shown that the simultaneous presence of these two phases is responsible for the strong increase of the coercive field in the region of coexistence. A similar but weaker effect on the coercive field is present upon coverage with small amounts of Cu (see Fig. 4) which may point to the presence of two magnetic phases for Cu coverages below 1 ML. However, no such strong increase in the coercive field is observed for larger Cu coverages.

The Mössbauer study of Ref. 55 on a Cu/6.5-ML Fe/Cu multilayer measuring only the two interior Fe layers of the Fe film found a low as well as a high spin phase when cooling below 150 K. The quadrupole splitting indicated a small interlayer compression in agreement with the findings for the uncovered Fe layers in phase II.[6] There is still some unresolved question about the magnetic anisotropy: While in Ref. 55 an in-plane easy axis was found in our present investigations we found a perpendicular magnetization component. However, we also found that small differences in the preparation of the sample may lead to an in-plane component at larger Cu thickness.

To summarize the above discussion it is shown that Cu coverage causes a change in the magnetic moment as well as in the structure of the Fe film. There is some evidence that this may be described by a magnetovolume effect. However, the local maximum in the Curie temperature at 2 ML Cu coverage seems not to be accompanied by strong structural changes. In the following we want to stress the point that there may be magnetic changes present beyond the magnetovolume effect: A theoretical investigation by Fu and Freemann[2] revealed that the surface of a Fe film on Cu(001) has an enhanced magnetic moment of $2.85\mu_B$. The coverage of Cu decreases the magnetic moment of the Fe at the Fe/Cu interface by $0.25\mu_B$. A more recent spin-polarized relativistic Korringa-Kohn-Rostoker calculation by Szunyogh, Ujfalussy, and Weinberger[52] showed that the magnetic moment of the surface Fe layer is indeed reduced by more than 20% upon coverage with an (infinitely thick) Cu layer. However, a complete redistribution of the magnetic moment over the film thickness occurs and generally an increase of the averaged magnetic moment results. In this publication an antiferromagnetic ordering of the individual Fe layers in the Fe film was assumed which is the ground state for a film thickness larger than 2 ML according to their calculation. In an earlier publication the same group presented calculations for the ferromagnetic aligned layers as well.[48] For the magnetic moment per unit cell for a 3-ML Fe film they got $7.86\mu_B$ for the uncovered film, $7.58\mu_B$ for 1-ML Cu, $7.63\mu_B$ for 2 ML Cu, and $7.51\mu_B$ for an infinitely thick Cu overlayer, i.e., a *minimum* of the magnetic moment for 1 ML and a *maximum* for 2 ML Cu coverage very similar to behavior of the MOKE data presented above. In this calculation the authors did not take any layer relaxation effects into account. The nonmonotonous change of the magnetic moment in the calculation with the Cu coverage is therefore entirely of electronic origin and no structural changes have to be invoked.

We believe that such an entirely electronic effect is mainly responsible for the maximum in the Curie temperature at 2 ML Cu coverage, since this is independent of the structure of the Fe film. The same effect is observed independent of the Fe thickness. Delocalized quantum well states in the Cu overlayer as mentioned by Ref. 63 are probably less important for the observed effect since we did not see any further oscillations at larger Cu thickness.

The variations in the magnetic moment in the theoretical calculation by Ref. 48 are rather small. A reduction of the magnetic anisotropy would also cause a decrease in T_C. In fact, according to the Mermin-Wagner theorem in absence of any anisotropy long-range ordering could not persist at any finite temperature in this two-dimensional system of ultrathin ferromagnetic films. It is only the presence of a magnetic anisotropy, which, leads to a finite Curie temperature.[67] In fact, the (uniaxial) magnetic anisotropy is quite large for Fe films on Cu(100) and amounts to about 120 μeV/atom for a 3-ML Fe film at room temperature.[66] Nevertheless, a change of the surface by covering with a cap layer usually strongly change the magnetic surface anisotropy and a change of T_C can be expected. According to Ref. 67, T_C depends only logarithmically on the magnetocrystalline anisotropy. Significant changes of T_C solely due to the change of the anisotropy can be expected only if the anisotropy nearly vanishes.

V. CONCLUSIONS

We observed a strong decrease of the Curie temperature of fcc Fe films when covered by 1 ML Cu for the entire thickness range of the fct/fcc phase. This drop in T_C is partly reverted for a 2 ML Cu coverage while for all thicker Cu cover layers T_C reaches a constant lower value. The coverage of the Fe layer with 1 ML Cu is accompanied by structural changes of the Fe layer. However, no further strong structural changes are observed for an increased Cu cover layer thickness. Therefore we are inclined to the idea of assigning the maximum in T_C at a Cu coverage of about 2 ML to a change of the electronic structure alone, i.e., a change in the hybridization of Fe and Cu states at the interface, and not to a magnetovolume effect. This view is supported by the observed fact that this maximum in T_C at 2-ML Cu coverage was found in whole Fe thickness range from 2 to 10 ML's despite the vastly different structural and magnetic properties of phase I (below 4 ML) and phase II (above 4 ML) of the uncovered Fe film. Further structural characterization of Cu/Fe/Cu(001) sandwiches for thin Cu overlayers are necessary to clarify the possible contribution of a magnetovolume effect on the magnetic changes.

ACKNOWLEDGMENTS

We thank P. Ohresser for performing the IV-LEED model calculations and B. Schmidt for assistance in the measurements. One of us (M.S.) acknowledges financial support from the Deutsche Forschungsgemeinschaft.

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