Growth and magnetic properties of Co_xNi_{1-x} and Fe_xNi_{1-x} ultrathin films on Cu(100)

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We have grown ultrathin Co_xNi_{1-x} and Fe_xNi_{1-x} films on Cu(100) with varying stochiometry x. We find that these alloy films grow in a fcc phase on Cu(100). With the surface magneto-optic Kerr effect we measured the variation of the Curie temperature T_C as a function of the film thickness n in monolayers. Fitting an *empirical* scaling curve to our results we are able to extrapolate the value $T_C(n=\infty)$ for samples with different stochiometry. We use this framework in order to determine $T_C(n=\infty)$ for Fe_xNi_{1-x} alloy films, in particular for concentrations close to 65% Fe content. Bulk $Fe_{65}Ni_{35}$ shows a collapse of magnetic long-range order and a fcc-to-bcc structural transition, which is the so-called Invar effect. In ultrathin $Fe_{65}Ni_{35}$ films, we observe a ''quenching'' of the Invar effect, because growth on a Cu(100) substrate forces the film to adopt the Cu lattice spacing thereby suppressing the structural relaxation. [S0163-1829(97)00129-X]

INTRODUCTION

Molecular-beam epitaxy (MBE) has offered the possibility of stabilizing materials as thin films in new metastable phases, e.g., fcc Co/Cu(100) and fcc Fe/Cu(100).[1–5] In this paper we will show how we extended MBE techniques for growing metastable alloy films of Co_xNi_{1-x} and Fe_xNi_{1-x} on Cu(100) in a very controlled way. The motivation arises from two considerations. Firstly we want to test our *empirical* scaling concept for Co_xNi_{1-x} films introduced in an earlier publication.[6] Secondly using these scaling results we want to address the Invar problem in Fe_xNi_{1-x}. Since we will refer to Fe/Cu(100) in this publication we recall some results for this system.

The possibility of stabilizing fcc Fe on Cu(100) resulted in numerous studies which investigated the structure, growth, and magnetic properties,[5,7–14] sometimes reporting different or even contradictory results.[13,14] The driving force for these studies were first-principles calculations, which showed that fcc Fe can have several magnetic states.[15] Depending on the lattice constant (or atomic volume) antiferromagnetic (AFM), nonmagnetic (NM) or ferromagnetic with high-spin (HS)/low-spin (LS) phases are stable. At the Cu lattice constant these phases are very close in energy. This manifests itself in structural and magnetic instabilities in these Fe films which has been studied in detail by Müller *et al.*[5] and accounts for the different experimental results reported.

This behavior can be described in simple terms as the Fe atoms trying to adopt the bulk bcc atomic volume, while at the same time fulfilling the constraints imposed by the Cu substrate. A related phenomenon is the Invar effect in Fe_xNi_{1-x} bulk alloys. At a Fe concentration of 65% the magnetic moment deviates strongly from the Slater-Pauling curve, dropping quickly to zero as does the Curie temperature, at which point, a structural transition from fcc into bcc is observed.[16,17] In the ultrathin film limit Fe_xNi_{1-x}/Cu(100) grows pseudomorphically as we will show below. This enables us to ''clamp'' Fe_xNi_{1-x} into the fcc phase for Fe concentration beyond 65% and a fixed lattice constant imposed by the Cu substrate. Both points are relevant if we want to make a connection to very recent theoretical calculations.[18,19] The work of Abrikosov *et al.*[18] deals with fcc Fe_xNi_{1-x} alloys for all concentrations x in which the lattice remains fcc but the lattice constant is allowed to vary and adopts a value which gives the minimum of the total energy. They show that a collapse of the magnetic moment occurs again, but at the higher Fe content of 75%. At this concentration they find a lattice contraction of $\sim 3\%$. Due to the epitaxy on Cu(100) this relaxation is suppressed and we are able to show a modified phase diagram for Fe_xNi_{1-x} which reflects more the magnetic instabilities of this system.

EXPERIMENT

The experiments were performed in an UHV apparatus previously described[6,20] with a base pressure of 1×10^{-10} mbar, which was better than 4×10^{-10} mbar during alloy growth. The Cu(100) crystal was mechanically polished and subsequently electropolished before inserting into the vacuum system. A few cycles of Ar^+ sputtering (500 eV) and annealing to 750 K resulted in a sharp $p(1\times1)$ low-energy electron-diffraction (LEED) pattern and a contamination-free sample within the detection limit of the Auger spectrometer. The growth rate of the Co, Fe, and Ni sources were controlled by separate quartz crystal monitors (QCM), which were calibrated via reflection high-energy electron-diffraction (RHEED) oscillations of Co/Cu(100), Fe/Cu(100), and Ni/Cu(100).

The alloy films were grown near room temperature in order to avoid Cu segregation. We also observed RHEED oscillations during the growth of the alloy films which indicates good layer-by-layer growth, discussed below. The thickness determined by the calibrated QCM and by the RHEED oscillations of the alloy films agree to within 3%. This confirms our ability to control the thickness and the stochiometry very accurately.

The magnetic properties were investigated with the surface magneto optical Kerr effect (SMOKE). Our SMOKE apparatus can be operated in the polar and longitudinal geometry without sample movement.[6] The applied field was aligned along the [100] direction for in-plane measurements.

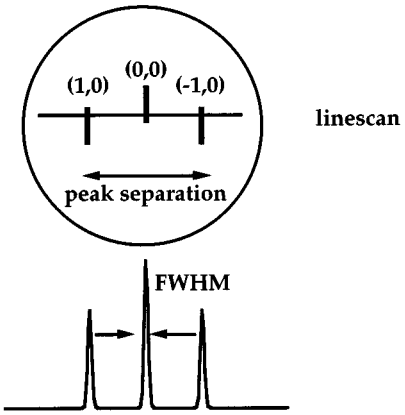

FIG. 1. Sketch of the RHEED experiment. The peak separation is proportional to the momentum transfer.

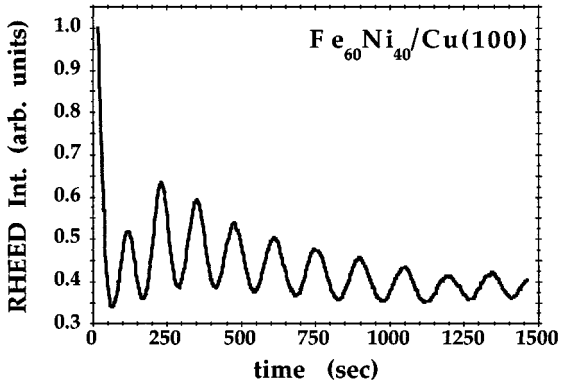

FIG. 2. Variation of the specular RHEED intensity during $Fe_{60}Ni_{40}$ growth, beam energy 3 kV.

Co_xNi_{1-x} alloy films were found to be magnetized in-plane, the same is true for Fe_xNi_{1-x} alloys with $x<0.5$. Fe_xNi_{1-x} films with $x=0.5$–0.75 were found to exhibit a small perpendicular component for thicknesses below 4 ML.[21] However upon increasing the sample temperature the perpendicular component vanishes below the Curie temperature. In summary the Curie temperature for both alloy systems was determined by the vanishing of the remanence in the longitudinal geometry.

We measured the remanence as function of temperature thereby determining the Curie temperature T_C for a given thickness and alloy concentration. In the early stages of the experiment the RHEED intensity of the specular beam was measured with a photodiode. Very recently we adapted a CCD camera based data acquisition system to the apparatus which allows us to do spatially resolved RHEED experiments similar to those reported by Fassbender *et al.*[22] We can determine intensities, full-width at half maximum (FWHM) of diffracted beams and the separation of diffracted beams in real time, see Fig. 1. The peak separation is proportional to the in-plane momentum transfer and therefore proportional to the inverse of the in-plane lattice constant. This means an increase of the peak separation is the result of a decrease of this lattice constant. Due to the grazing incidence of the electrons, RHEED essentially probes the top layer, therefore we are particularly sensitive to any changes of the lattice constant of this surface layer during growth. The angle of incidence was generally chosen to be around an anti-Bragg condition. An important aspect of the alloy growth is the question to what extent surface segregation plays a role. Using simple thermodynamic arguments we would expect little segregation to take place for Co_xNi_{1-x} and Fe_xNi_{1-x} alloys. Segregation is determined by three contributions to the free energy:[23]

(i) difference in the surface free energy of the two elements,

(ii) heat of solution,

(iii) difference in the atomic volume.

It turns out that for the alloy systems investigated here the first term is important. However, the surface free energy of Cu is significantly smaller than for Fe, Co, and Ni. Rather than segregation of one of the alloy elements we have to expect segregation of Cu. This is actually observed at temperatures $\sim$400 K for Fe/Cu(100) and Co/Cu(100),[10,24] which is also true for Fe_xNi_{1-x} alloys as our Auger spectra show. The fact that good epitaxial growth for, e.g., Co/Cu(100) is observed is only possible because ultrathin films are in a metastable state.

Work on Co_xNi_{1-x} and Fe_xNi_{1-x} bulk alloys has shown that in the first case surface segregation can be ignored,[25,26] whereas Fe_xNi_{1-x} alloys exhibit Ni segregation as Wandelt and Ertl show.[27] Their results indicate a Ni segregation of the order $\sim$7%.[28] Although this would not affect our conclusions concerning the Invar effect we have performed an analysis of Auger spectra. Using the integral electron gun of the CMA and a second gun which excited Auger electrons under grazing incidence ($\sim$15°) we investigated $\sim$10 ML thick alloy films.[29] Comparing the intensity ratios for Fe (47 and 650 eV) and Ni (60 and 848 eV) we found that we also detect a small amount of Ni segregation which was less than 5% for concentrations near the Invar concentration. Results for Ni-rich films showed a slightly higher Ni enrichment consistent with the findings of Wandelt and Ertl.[27] We therefore conclude that due to the small effect, surface segregation can be ignored.

RESULTS AND DISCUSSION

A. Growth and structure

The first goal in our investigation was to ensure good growth of Co_xNi_{1-x} and Fe_xNi_{1-x} alloys in the fcc phase. It is known that the lattice constant for bulk fcc Co_xNi_{1-x} varies almost linearly with concentration x between the values of the pure elements in the fcc phase. Ni has a lattice mismatch of 2.6% and Co has 1.7% when compared with Cu.[30] Similarly the bulk lattice constants of fcc Fe_xNi_{1-x} varies between 3.55 and 3.59 Å for concentrations $0.7>x>0.2$.[30] This gives only a very small lattice mismatch (0.6–1.6 %) with Cu, in particular close to the Invar region. The above lattice matching argument encourages growth of these alloys on a Cu(100) surface. Furthermore ''clamping'' Fe_xNi_{1-x} films on Cu(100) should extend the concentration range for which the fcc structure prevails, which is the key point for our discussion of the Invar effect.

We have observed RHEED oscillations for Co_xNi_{1-x} and Fe_xNi_{1-x} grown on Cu(100) which are more pronounced in the case of Fe_xNi_{1-x} alloys. In Figs. 2 and 3 we show the

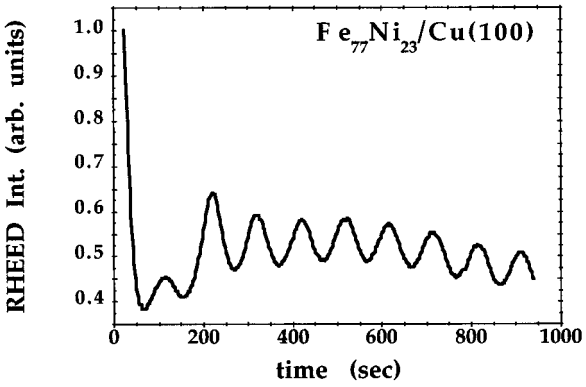

FIG. 3. Variation of the specular RHEED intensity during $Fe_{77}Ni_{23}$ growth, beam energy 3 kV.

RHEED intensity during the growth of two Fe_xNi_{1-x} alloy films with concentrations 60 and 77 % Fe. We can clearly observe RHEED oscillations, which indicate layer-by-layer growth. Fe agglomeration in the early stages similar to that observed for Fe/Cu(100) seems to be absent, since a pronounced drop in intensity at the position of the first monolayer is missing. From this point of view the $Fe_{77}Ni_{23}$ alloy films are qualitatively identical with the RHEED data for Fe_xNi_{1-x} films below 65%. However we observe that the intensity of the first maximum for the $Fe_{77}Ni_{23}$ alloy is more strongly reduced as compared with $Fe_{60}Ni_{40}$ alloy. We come back to this point in our discussion below. However it is important to note that there is no sudden transformation into the bcc phase, which would also be accompanied by a drop in the RHEED intensity like for Fe/Cu(100).[14,24] This is in contrast to bulk $Fe_{77}Ni_{23}$ which has transformed into the bcc phase. This observation is important because it confirms that our idea of clamping Fe_xNi_{1-x} on Cu(100) extends the concentration range for which the fcc structure prevails. We will show below results of the spatially resolved RHEED experiments, which confirm this statement. Using this setup we are able to monitor the specular and diffraction streaks simultaneously. A change of the structure would change the position of the diffraction streaks dramatically, which we do not observe. The observation of $p(1\times1)$ LEED patterns confirmed epitaxial growth for both alloy systems, however the sharpness of the spots deteriorated slightly with increasing thickness. Pseudomorphic growth of Fe_xNi_{1-x}/Cu(100) has also been reported by Dresselhaus *et al.*[31] After establishing our ability to grow Co_xNi_{1-x} and Fe_xNi_{1-x} in a stable fcc phase we investigated the magnetic properties. We want to discuss the Co_xNi_{1-x} system first, which is in bulk well-behaved both structurally and magnetically.

B. Magnetism of Co_xNi_{1-x} films

The thickness dependence of the Curie temperature in the limit of large thicknesses is well known both theoretically and experimentally also known as finite-size scaling.[32,33] However theoretically little is known how T_C evolves with thickness in the monolayer regime. In our previous study[6] we have shown that the variation of the Curie temperature $T_C(n)$ with film thickness n in ML's can be described with an *empirical* scaling formula:

$$\frac{T_C(n)}{T_C(\infty)}=\frac{1}{1+[(n-n')/n_0]^{-\gamma}}. \tag{1}$$

The thickness of a film is given by the number of monolayers n, consequently $T_C(n)$ denotes the Curie temperature at this thickness. Here n' is an empirical constant approximately equal to 1.1, n_0 is another empirical constant, which has a value of approximately 3.4. This follows from our previous work on Ni/Cu(100).[6,34] Although we originally used the known bulk value $T_C(n=\infty)$ for Ni, we found that treating $T_C(n=\infty)$ as an independent fit parameter gave identical results. Leaving only $T_C(n=\infty)$ and γ as independent fit parameters Eq. (1) serves as a tool for determining $T_C(n=\infty)$ if a direct measurement is not possible. At this point we want to make few comments about the above formula. We notice that in the limit $n\rightarrow\infty$ Eq. (1) reduces to the well-known finite-size scaling formula if we set λ equal to γ:[32]

$$\frac{T_C(\infty)-T_C(n)}{T_C(\infty)}=\left(\frac{n}{n_0}\right)^{-\lambda}. \tag{2}$$

In this context λ is related to the critical exponent of the pair-correlation function. This concept is strictly valid only for thick films, the threshold being given by the dimensionality crossover from three dimensions to two dimensions (3D)–(2D), which has been found to be around 5 ML.[6,35] As we will show below the thickness regime covered in this work is mainly below this limit, therefore the use of Eq. (2) is not warranted. Furthermore there is clear evidence that a minimum coverage is required before long-range order at a finite temperature is established.[6,14,35–42] The exact coverage depends on the early stages of the growth, but there is consensus that around a thickness of 1 ML ferromagnetism occurs. A recent paper stated that submonolayer magnetism for the Fe/W(110) system is possible.[43] This does not contradict the above statement, since the observation required growth at elevated temperatures which resulted in a decoration of step edges. We come then to the conclusion that we have to introduce a ''boundary'' condition, which allows long-range order to exist only if a minimum coverage is exceeded. This is the justification for the introduction of the parameter n' in this work and we will use the value 1.1 as mentioned above.[44]

Interestingly the analysis of the data for Ni/Cu(100) with Eqs. (1) and (2) gives similar results.[34,45] Identifying γ^{-1} with υ our *empirical* scaling yields $\upsilon=0.80$ as compared with $\upsilon=0.705$, Schulz *et al.* actually fix υ to the value of the 3D Heisenberg system. Another reasonable agreement exists in their value of $C_0=5.2$ which in our notation equals 3.2, and we found to be 3.4 as stated above. We should point out that we used all data points for Ni/Cu(100) whereas Schulz *et al.* used coverages larger than 5 ML. This observation encouraged us to develop a concept which allows us to determine T_C for infinite thickness. Obviously it would be desirable to measure T_C for very thick alloy films. This would give us directly $T_C(n=\infty)$ which we could compare with T_C(bulk), but the experimental window is limited since at 400 K Cu segregation is noticeable for the Fe_xNi_{1-x} alloys as Auger spectra show. Furthermore it has been shown that a Cu layer causes Fe/Cu(100) films to reorder.[46] Since structure and magnetic properties are intimately related, we have

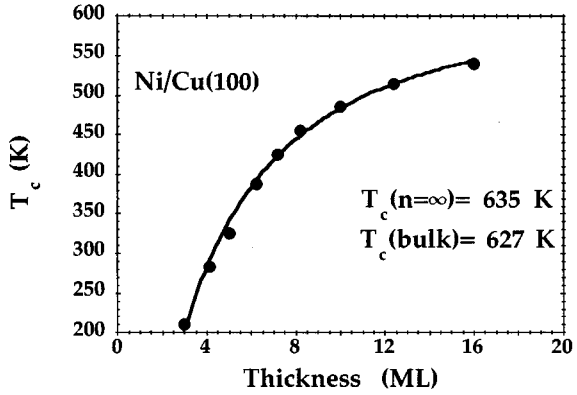

FIG. 4. Thickness dependence of T_C for Ni/Cu(100) films, solid cure is fit to the *empirical* finite-size scaling formula.

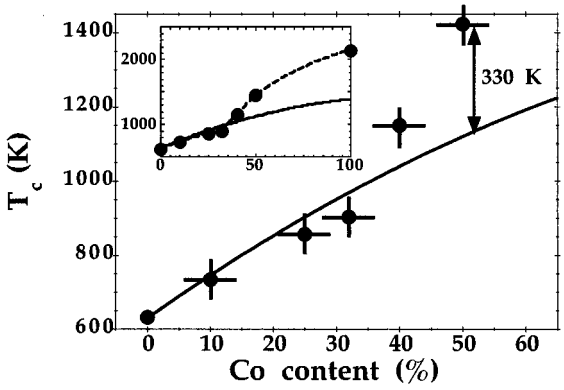

FIG. 5. Concentration dependence of $T_C(n=\infty)$ for Co_xNi_{1-x} films between 0 and 65 % Co content. The errors show variation of $T_C(n=\infty)$ on fit parameters and accuracy of composition. The solid line are the bulk values for T_C. Inset shows the overall concentration dependence of $T_C(n=\infty)$ and T_C(bulk).

to avoid Cu segregation. This is particularly important for Fe_xNi_{1-x} alloys, where we expect structural instabilities associated with the Invar effect. Although the concept of our *empirical* scaling in these pseudomorphic layers has been discussed by us in an earlier publication,[6] we want to discuss in more detail its accuracy in determining $T_C(n=\infty)$. Ni/Cu(100) is particularly well suited, because of the relatively low Curie temperature and the thermal stability of these films up to 500 K.[47,48] As a result we have a large set of data points available and we can compare $T_C(n=\infty)$ with T_C(bulk), which we have shown to give very good agreement,[6] see Fig. 4. If we want to use the scaling formula (1) as a tool to determine $T_C(n=\infty)$ for Fe_xNi_{1-x} films we need to know the validity and accuracy of this concept. Another test is the omission of the four data points with the highest T_C of the Ni/Cu(100) data set. Surprisingly the result for $T_C(n=\infty)$ has changed by only a few K. The choice of removing the last four data points of the Ni data was not arbitrary. The highest T_C of the reduced data set is 425 K and this value is more or less the maximum temperature to which the Fe_xNi_{1-x} films have been exposed. This means if $T_C(n=\infty)$ is determined to be below 635 K, this value should be rather accurate as judged from the Ni/Cu(100) case, see Fig. 4.

To further test the validity of formula (1) we have measured T_C as function of thickness for different Co_xNi_{1-x} alloy films. We have determined $T_C(n=\infty)$ and compared it with the bulk values, see Fig. 5. The vertical error bars reflect the variation of the determination of $T_C(n=\infty)$ on the fit parameters, whereas the horizontal error bars indicate the variation of the concentration during the deposition. For up to 32% Co content $T_C(n=\infty)$ and T_C(bulk) agrees reasonably, at this point we have $T_C(\text{bulk})\approx 950$ K. At higher Co concentrations we see a systematic deviation and for the pure Co films we extrapolate $T_C(n=\infty)\approx 2100$ K, which is more than 700 K too high. This trend of overestimating is obviously related to the fact that the available range of thicknesses is reduced if T_C(bulk) becomes larger. While the available temperature window remains the same, the limiting factor is the onset of Cu surface segregation. Summarizing the results for Co_xNi_{1-x} films, we can state that we are able to determine $T_C(n=\infty)$ accurately as long as the ''real'' value does not exceed 950 K. The accuracy is given by error bar of the fit parameters, which gives ~ 50 K. We find the parameter γ to be 1.25 for Ni which varies between 1.04 and 1.50 for Co concentrations up to 40%. As observed before[6] there is no clear concentration dependence for γ. In fact we found the value more affected by the choice of n' and the width of the temperature interval covered by the actual experiment; however this did not change the value of $T_C(n=\infty)$ significantly. For Fe_xNi_{1-x} alloys the parameter γ varies between 0.59 and 0.92, this is smaller than for Co_xNi_{1-x} alloys. This we associate with the reduced available temperature interval (compared with Co_xNi_{1-x}) which is limited by the onset of Cu surface segregation. In conclusion the parameter γ scatters significantly and only in the limit of very large thicknesses [e.g., Ni/Cu(100)] can we make a connection to a theoretical prediction as discussed before.

C. Magnetism of Fe_xNi_{1-x} films

We can now proceed and use formula (1) for Fe_xNi_{1-x} alloy films and compare the results with the bulk behavior. In

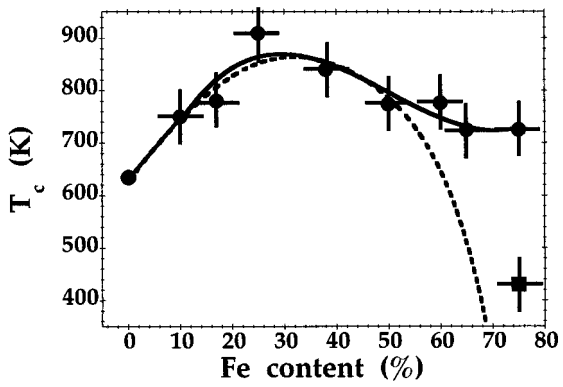

FIG. 6. Comparison between bulk Curie temperatures (Ref. 16) (dashed curve) and $T_C(n=\infty)$ (solid points) of the ultrathin film data. Error bars show variation of $T_C(n=\infty)$ on fit parameters and accuracy of composition. The solid line is guide for the eye. Solid square is $T_C(n=\infty)$ for the $Fe_{75}Ni_{25}$ alloy for higher thicknesses, see text and Fig. 7.

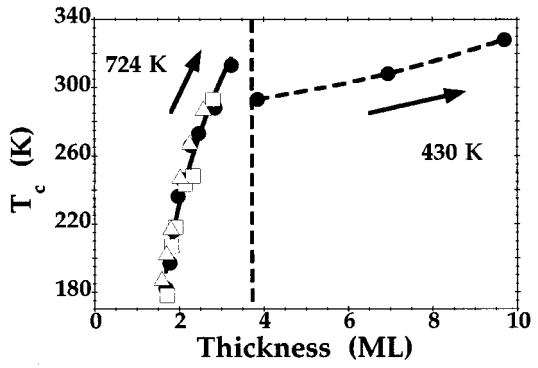

FIG. 7. Thickness dependence of T_C for a $Fe_{75}Ni_{25}$ alloy (solid symbols) in the thin-film limit. The solid line is fit to the *empirical* scaling law (Refs. 6 and 57), the dashed line is a guide for the eye. The squares (triangles) are data for $Fe_{65}Ni_{35}$ ($Fe_{60}Ni_{40}$), their scaling curves are very close to the one for the $Fe_{75}Ni_{25}$ alloy.

Fig. 6 we show the comparison of $T_C(n=\infty)$ and published values for T_C(bulk).[16] The first thing to notice is that the highest value for $T_C(n=\infty)$ is 908 K for $Fe_{25}Ni_{75}$. Referring to the preceding paragraph we are still within the temperature range for which we expect our *empirical* scaling formula to work. If we limit ourselves to Fe concentrations up to 30% only, we find good agreement between $T_C(n=\infty)$ and T_C(bulk). For these small Fe concentrations Fe_xNi_{1-x} is well within the regime where it is well behaved,[16] i.e., the magnetic moment follows the Slater-Pauling curve. This is another test of the validity of formula (1), which we will use as a tool for Fe_xNi_{1-x} films with higher Fe concentrations. Coming back to Fig. 6, we see that for Fe concentrations up to 50% the bulk values and thin-film data agree, at higher concentrations T_C(bulk) falls rapidly, but the value for $T_C(n=\infty)$ stays almost constant at around 720–770 K. At this point we would like to refer to the work of Dumpich *et al.*[49] They investigated thick (200 nm) fcc Fe_xNi_{1-x} films, in particular $Fe_{65}Ni_{35}$. They find that the as-grown film has a Curie temperature of $T_C \sim 700$ K, which agrees well with our scaling result, see Fig. 6. This again shows that it is justified to use the *empirical* scaling formula (1) as a tool.

Also important is that the alloy films are all ferromagnetic even beyond the bulk Invar concentration which is in contrast to the bulk behavior. In this sense we have ''quenched'' the Invar effect in the ultrathin limit. In Fig. 7 we show the thickness dependence for several concentrations near the bulk Invar concentration of 65% Fe. We see in the ultrathin limit almost identical behavior. Extending the measurements for a $Fe_{75}Ni_{25}$ alloy film up to 10 ML thickness reveals a strong deviation from the scaling law, see Fig. 7. At ~ 4 ML, T_C has dropped when compared wit the scaling curve and increases only slowly with further deposition. This is in contrast to the $Fe_{60}Ni_{40}$ and $Fe_{65}Ni_{35}$ alloys, which do not show this. In order to quantify this deviation more clearly we have scaled the value of $T_C(n=9.7$ ML) with the scaling curve of Ni/Cu(100) at this thickness. This allows us to determine a second $T_C(n=\infty)$ for the $Fe_{75}Ni_{25}$ alloy, this has been included in Fig. 6. As indicated in Fig. 7, the values are 724 and 430 K, respectively. But even without the results of the scaling curve it is apparent that we observe some sort of magnetic transition. Visual inspection of Figs. 4 and 7 shows that the $T_C(n)$ of the $Fe_{75}Ni_{25}$ alloy increases faster with thickness as Ni/Cu(100) for coverages up to ~ 4 ML. Beyond this thickness the $T_C(n)$ curve of the $Fe_{75}Ni_{25}$ alloy drops quickly below the Ni curve. At this point it should be mentioned that the variation of the Kerr intensity with thickness for all Fe_xNi_{1-x} alloys did not show indications of a ''magnetic live surface layer'' (Ref. 21) as, for example, seen by Thomassen *et al.*[14]

If we associate this behavior with a change of the magnetic moment, then we observe a transition from a high-spin (HS) state to a low-spin (LS) state. We identify this with an Invar-like transition at an increased Fe concentration of $\sim 75\%$ when compared with the bulk. Interestingly the concentration agrees very well with the calculation of Abrikosov *et al.*; they find a moment collapse at 75% Fe.[18]

D. RHEED experiments on Fe_xNi_{1-x} films with spatial resolution

We want now to make a connection to the theoretical work of Abrikosov *et al.*[18] In their work they avoid the structural complications associated with the transition from the fcc phase into the bcc phase by assuming a fcc lattice throughout the whole concentration range. They find that the magnetic moment collapses as in the ''real'' bulk system, but at a higher Fe concentration of 75%. At this point the system undergoes a change of the fcc lattice constant, which is reduced by $\sim 3\%$. We should point out that Abrikosov *et al.* do not include an antiferromagnetic (AFM) phase in their calculation for the Fe_xNi_{1-x} alloy, which is certainly important in the limit of pure fcc Fe.[18,50] Antiferromagnetic phases in Fe/Cu(100) have been observed by utilizing Mössbauer spectroscopy.[51,52]

First we see that we have good agreement about the Fe concentration where a transition occurs, the value is significantly larger than what is experimentally observed in the bulk.[16] But, in contrast, we do not observe a collapse of the magnetic moment, since T_C remains finite for the $Fe_{75}Ni_{25}$ alloy film. This would indicate that beside their high-spin (HS)/nonmagnetic (NM) solution the possibility of the existence of a low-spin (LS) solution or coexistence of different phases.[53] However we still would expect a reduction of the lattice constant if the deviation of T_C from the scaling curve is caused by a reduction of the magnetic moment. This follows immediately from the dependence of the magnetic moment on the lattice constant for a $Fe_{74}Ni_{26}$ alloy as plotted by Abrikosov *et al.*[18] If the lattice constant is varied continuously between the values for the high-spin (HS)/nonmagnetic (NM) phase they show a monotonic variation.

In order to address this point we have employed the RHEED apparatus which allows us to do spatially resolved experiments. We then can determine the peak separation of the (1,0) and $(-1,0)$ RHEED streaks which is inversely proportional to the in-plane lattice constant, see Fig. 1.

In Fig. 8 we show evolution of the intensity of the (0,0) beam and the peak separation between the (1,0) and $(-1,0)$ diffraction peaks for a $Fe_{66}Ni_{34}$ alloy film. This concentration is at the bulk Invar concentration, but below the fcc Invar concentration of Abrikosov *et al.*[18] We see oscillations of the peak separation of the value at the minima are

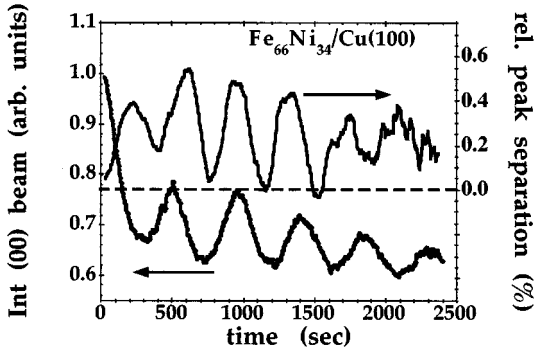

FIG. 8. Variation of the RHEED intensity of the (0,0) beam and peak separation for a $Fe_{66}Ni_{34}$ alloy, beam energy 4.5 kV.

close to the initial value. In a simple picture one would not expect oscillations of the peak separation at all. The origin of these oscillations are due to a relaxation of the edge atoms of islands in the top layer as discussed by Fassbender *et al.*[22] This means the amplitude of the oscillation should not be mistaken as an uniform contraction of the top layer; it is the average of edge atoms and atoms within islands sampled over the electron-beam diameter.

Although we probe only the top layer, it is clear that the underlying layers must also adopt the Cu lattice spacing. The important result is now that $Fe_{66}Ni_{34}$ alloy film is still clamped to the Cu(100) substrate lattice, since the peak separation is the same as that of the clean substrate. We now compare these results with the data for a $Fe_{80}Ni_{20}$ alloy. In Fig. 9 we plot the intensity of the (0,0) beam and the peak separation between the (1,0) and $(-1,0)$ diffraction peaks during the growth of the alloy. In contrast to Fig. 8 we do not see a regular pattern for the peak separation. The first monolayer seems to be clamped to the Cu(100) substrate, since the peak separation goes back to its original value. However going to higher coverages the peak separation gradually increases, which means the in-plane lattice constant is reduced. Compared to the Cu lattice constant we observe a reduction between 0.7 and 1 % at 4 ML $Fe_{80}Ni_{20}$. For both alloys (66 and 80 % Fe content) we are also able to observe oscillations in the intensity and FWHM of the (1,0) and $(-1,0)$ beams. This also does not support an explanation of the $T_C(n)$ behavior of the $Fe_{75}Ni_{25}$ alloy in terms of different growth regimes like Fe/Cu(100).[14] In both cases we have good epitaxial growth and the main difference between the $Fe_{66}Ni_{34}$ and $Fe_{80}Ni_{20}$ alloy films is the thickness dependence of the peak separation.

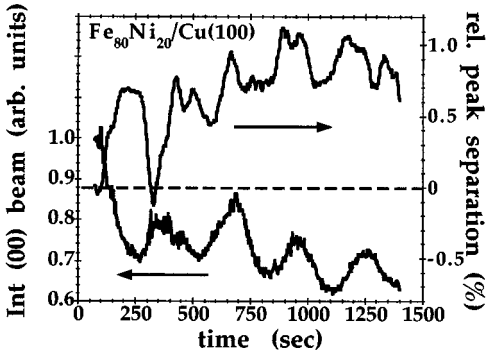

FIG. 9. Variation of the RHEED intensity of the (0,0) beam and peak separation for a $Fe_{80}Ni_{20}$ alloy, beam energy 4.5 kV.

It should be pointed out that the oscillation period for the intensity and the peak separation for the $Fe_{66}Ni_{34}$ alloy are not identical. In fact the period for the peak separation is only 85% of the intensity period. In the picture of Fassbender *et al.* this points towards some change of the island morphology during the growth. It is interesting to note that for concentrations below 66% Fe the period of the intensity and peak separation is identical. It appears that upon approaching the Invar concentration the morphology is affected by the magnetic instability. We also observe that in the early stages of the growth of $Fe_{77}Ni_{23}$ and $Fe_{80}Ni_{20}$ the first maximum in the intensity is reduced, see Figs. 3 and 9. This behavior is commonly associated with some amount of bilayer growth. The resemblance with the RHEED intensity of $Fe_{77}Ni_{23}$ and Co/Cu(100) together with scanning tunneling microscope results on Co/Cu(100) would indicate only a small amount of bilayer growth of the order 10%.[54] We therefore conclude that the structural differences are more important than small deviations from perfect layer-by-layer growth.

There are now two points which need to be discussed here. First the thickness dependence of T_C for a $Fe_{75}Ni_{25}$ alloy film shown in Fig. 7 shows a rather sharp deviation from the scaling curve at 3 ML. The peak separation of the $Fe_{80}Ni_{20}$ has significantly increased at around 1.7 ML. It appears that the length scales do not quite agree. It might be related to the fact that we compare slightly different alloy concentrations. We would expect the $Fe_{80}Ni_{20}$ alloy to be more ''unstable'' and trying to adopt the low-spin phase at lower coverages.

Secondly our lattice contraction is 0.7–1 % as compared to the 3% calculated by Abrikosov *et al.* This discrepancy might be due to the fact that we compare experimentally a HS phase with a LS phase, since T_C remains finite. Their calculation does not include a LS solution, however we expect a smaller lattice contraction between a HS phase and LS phase as discussed before. From this point of view we conclude that the deviation from the scaling curve in the case of the $Fe_{75}Ni_{25}$ alloy is caused by a reduction of the magnetic moment, which in turn manifests itself in a small in-plane lattice contraction. Preliminary LEED *I-V* results based on Bragg-peak analysis[10] of the (0,0) beam indicate that between 2 and 6 ML a contraction of the perpendicular lattice constant takes place.[55] This further confirms the view that we have observed a moment-atomic volume instability.

As a test of our instrument we have performed a similar RHEED experiment for the pure Fe/Cu(100) system. In Fig. 10 we see that at the beginning the peak separation decreases first. This means in real space that the lattice constant is increased. The Fe atoms try to adopt the bulk atomic volume, but experience the constraint of the Cu substrate. This also manifests itself in the rather complicated surface reconstructions known for this system.[5,14,24] At around 3.9 ML we see a dramatic reduction of the peak separation after which oscillations set in. It is now important to refer to the work of Müller *et al.*[5] In their careful LEED *I-V* analysis they find an increase of the atomic volume of the surface layer, this is exactly what we probe with the RHEED experiments. The thickness also coincides with the observation of a magnetic

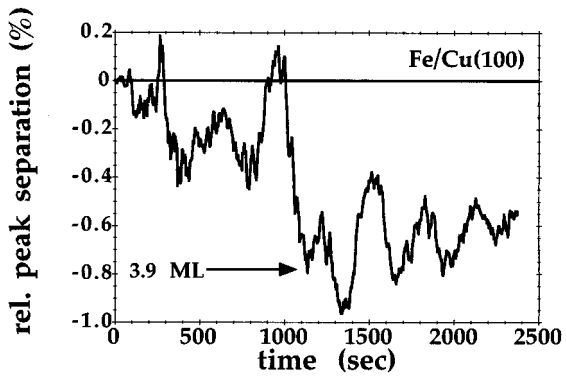

FIG. 10. Variation of the peak separation for a Fe/Cu(100), beam energy 4.5 kV.

''live'' layer by Thomassen *et al.*[14] At this thickness only the top layer is magnetic. The driving force is the attempt by the Fe atoms to adopt the bulk atomic volume while being grown on Cu(100). However, we should point out that in contrast to the work of Müller *et al.*[5] who report an increase of the interlayer distance, we have determined an increase of the in-plane lattice constant. This makes the observation of a magnetic live layer even more plausible.[13] The increased in-plane lattice constant together with the increased layer separation found by Müller *et al.* means that the Fe top layer is ''decoupled'' from the underlying layers. It should be pointed out that LEED *I-V* studies usually do not consider a layer dependence of the in-plane lattice constant.[56] Therefore our observation of an increase of the in-plane lattice constant of the top layer does not contradict the findings of Müller *et al.*[5]

First we see that our instrument is capable of detecting in-plane lattice constant changes of the order of 1%. We also see the differences between Fe/Cu(100) and Fe_xNi_{1-x}/Cu(100). Fe/Cu(100) is structurally more unstable, hence the changes in the magnetic properties are more clearly visible. The lack of such a clear signal for the $Fe_{80}Ni_{20}$ alloy indicates that the mechanism of the moment instability is more subtle than for the system Fe/Cu(100).

SUMMARY

We have shown that Co_xNi_{1-x} and Fe_xNi_{1-x} films can be stabilized in the fcc phase when grown as ultrathin films on Cu(100). Since Co_xNi_{1-x} alloys are well behaved both magnetically and structurally in the bulk we have used Co_xNi_{1-x}/Cu(100) as a test system for our scaling formula (1). By adjusting only two parameters we are able to extrapolate values for $T_C(n=\infty)$, which are in good agreement with bulk values as long as $T_C(n=\infty)$ does not exceed $\sim$900 K. This allows us to determine the concentration dependence of $T_C(n=\infty)$ for Fe_xNi_{1-x}/Cu(100) films, which we compare with T_C(bulk). We find again that for Fe concentrations below 30% Fe the results derived from the scaling formula (1) and the bulk values agree reasonably. Using this scaling formula for Fe_xNi_{1-x} alloys near 65% Fe content, we observe that the moment collapse is suppressed in the ultrathin limit since $T_C(n=\infty)$ hardly varies near the Invar concentration. However a deviation from the scaling curve for $Fe_{75}Ni_{25}$ alloy at thicknesses larger than 4 ML occurs. Our RHEED experiments show a small $\sim$0.7% in-plane lattice spacing contraction for a $Fe_{80}Ni_{20}$ alloy. We therefore conclude that a structural relaxation is responsible for the different thickness dependence of T_C for $Fe_{75}Ni_{25}$/Cu(100). In other words the structural change results in a different magnetic phase.

The concentration for which we observe a deviation from the scaling curve agrees with the concentration where Abrikosov *et al.* find the moment collapse to take place. Since T_C remains finite in our work we conclude that an additional low-spin phase exists. This would also explain the difference in the magnitude of the lattice contraction between our experiments and the calculation of Abrikosov *et al.*

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