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on Cu(100)

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Growth and magnetic properties of $\text{Fe}_x\text{Ni}_{1-x}$ ultrathin films on Cu(100)

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We have investigated ultrathin $\text{Fe}_x\text{Ni}_{1-x}$ films grown epitaxially on Cu(100) with different stoichiometry. With the surface magneto-optic Kerr effect (SMOKE) we measured the variation of the Curie temperature T_C as a function of the film thickness n in monolayers (ML). Using the results of our previous investigations on finite-size scaling (Huang *et al.*), we are able to extrapolate the value $T_C(\infty)$ for samples with different Fe content. In particular, alloy films with Fe concentrations close to 65% remain ferromagnetic. This is in contrast to bulk $\text{Fe}_{65}\text{Ni}_{35}$, which shows a collapse of long range order, which is the so-called invar effect associated with a fcc to bcc structural transition. Growing these alloy films on a Cu(100) substrate forces them to adapt the Cu lattice spacing, thereby suppressing the structural relaxation. © 1996 American Institute of Physics. [S0021-8979(96)19908-1]

I. 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). Theoretical studies reveal that fcc Fe can have several magnetic states.¹ Depending on the lattice constant (or atomic volume) antiferromagnetic (AF), nonmagnetic (NM) or ferromagnetic with high spin (HS)/low spin (LS) phases are stable. At the Cu lattice constant, these magnetic phases are very close in energy, which made the Fe/Cu(100) system very attractive for experimental studies.²⁻⁹ The LEED I - V study by Müller *et al.*¹⁰ shows that Fe/Cu(100) films with thicknesses up to 4 ML order in a heavily distorted fcc structure, where the atomic volume of Fe is increased with respect to Cu. This has the result that the whole film is in a ferromagnetic state.^{9,11} Going to higher thicknesses at 300 K, Fe adopts the Cu atomic volume, except for the top layer, which still has an increased atomic volume before Fe transforms into the bcc phase. This means that the ferromagnetism is only located in the top layer⁹ while the rest of the film is antiferromagnetically ordered.⁸ In simple terms one can say that the Fe atoms try to adopt the bulk bcc crystal atomic volume while at the same time fulfilling the constraints imposed by the Cu substrate.

A well-known moment instability in the bulk is the invar effect in $\text{Fe}_x\text{Ni}_{1-x}$ 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 the fcc into the bcc phase is observed.¹² Also, the thermal expansion is very low.¹² From a comparison with FePt and FePd alloys, which do not show a collapse of the magnetic moment, but have a small thermal expansion and strong reduction of the Curie temperature¹² it seems that the moment instability is not an invar relevant feature. A recent study¹³ of 200 nm thick as-grown $\text{Fe}_{65}\text{Ni}_{35}$ films revealed that the magnetic moment follows the Pauling–Slater curve, but a small thermal expansion was still present. Again, stressing the point that

the collapse of the magnetic moment is not an invar relevant feature.

The bulk lattice constants of fcc $\text{Fe}_x\text{Ni}_{1-x}$ vary between 3.55–3.59 Å for concentrations $0.7 > x > 0.2$.¹⁴ This gives only a very small lattice mismatch with Cu. This fact motivates the growth of $\text{Fe}_x\text{Ni}_{1-x}$ films on Cu(100), for which we can expect good epitaxy with the above lattice matching argument. Furthermore, “clamping” $\text{Fe}_x\text{Ni}_{1-x}$ films on Cu(100) should extend the concentration range for which the fcc structure prevails.

At this point it is important to refer to the work of Abrikosov *et al.*,¹⁵ which discusses bulk fcc $\text{Fe}_x\text{Ni}_{1-x}$ through the whole concentration range. They find that the magnetic moment collapses at $x=0.75$ and the lattice parameter drops noticeably, this is due to the fact that the LS/NM solution becomes lower in energy. We see that “clamping” $\text{Fe}_x\text{Ni}_{1-x}$ to a fcc structure alone does not prevent the deviation from the Slater–Pauling curve, but by keeping the lattice constant fixed we expect a suppression of the invar effect.

II. EXPERIMENTAL

The experiments were performed in an UHV apparatus previously described¹⁶ with a base pressure of 1×10^{-10} mbar, which was better than 4×10^{-10} mbar during Fe, Ni codeposition. The Cu(100) crystal was mechanically polished and electropolished before inserting into the vacuum system. A few cycles of Ar^+ sputtering and annealing re-

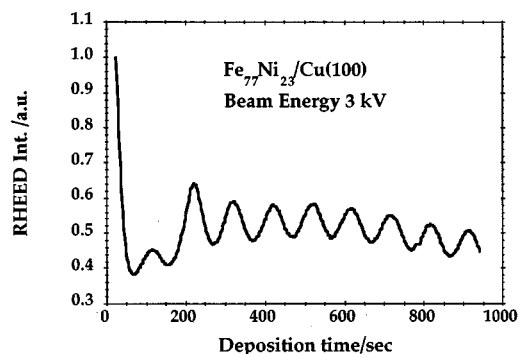


FIG. 1. Variation of the specular RHEED intensity during $\text{Fe}_{77}\text{Ni}_{23}$ growth.

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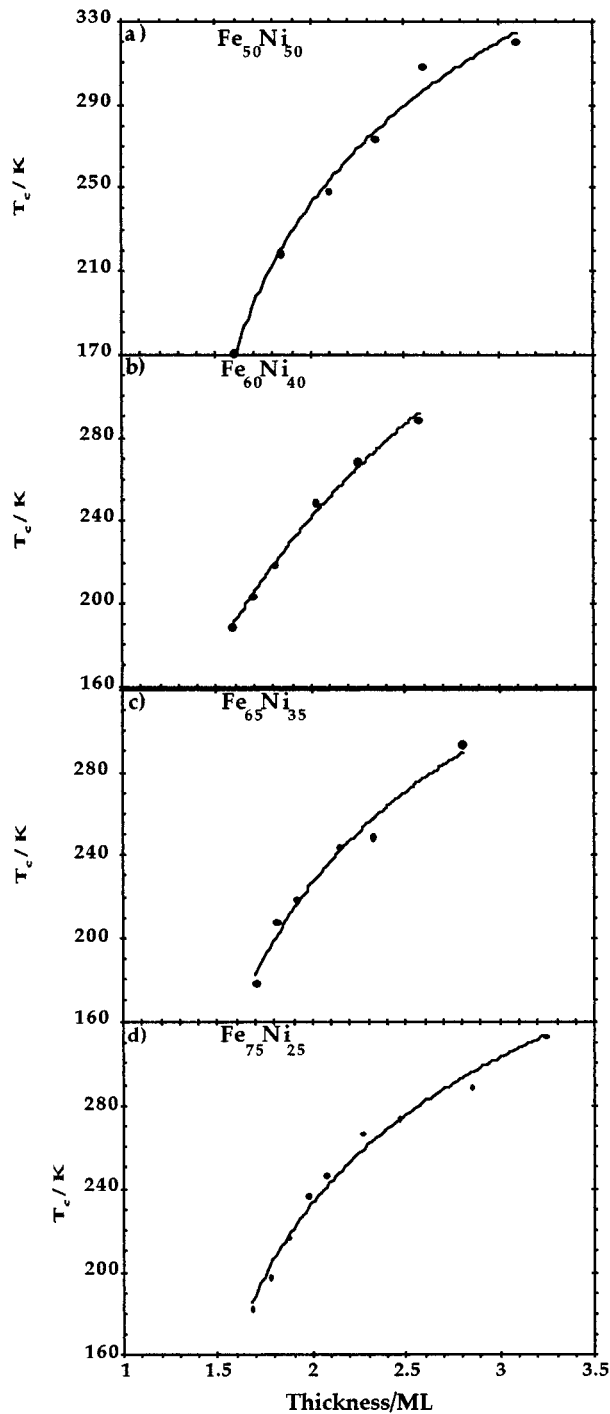


FIG. 2. Thickness dependence of T_C for different alloy concentrations. The solid line represents the fit to the scaling law; see the text.

sulted in an ordered and clean sample, as judged from LEED and Auger spectroscopy. The growth rate of the Fe and Ni sources were controlled by quartz crystal monitors (QCM), which were calibrated via RHEED oscillations of Fe/Cu(100) and Ni/Cu(100).¹⁶

The alloy films were grown at 350 K in order to avoid Cu segregation, we also observed RHEED oscillations during the growth of $\text{Fe}_x\text{Ni}_{1-x}$ films, which indicates good layer-by-layer growth; see Fig. 1. The thickness determined

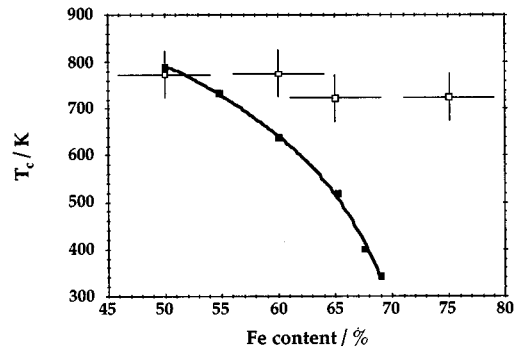


FIG. 3. Comparison between bulk Curie temperatures¹² (solid squares) and $T_C(\infty)$ of the thin film data (open squares). Error bars show variation of T_C on fit parameters and accuracy of composition.

by the calibrated QCM and by the RHEED oscillations of the $\text{Fe}_x\text{Ni}_{1-x}$ films agree to within 3%. This confirms our ability to control the thickness and the stoichiometry very accurately.

The presence of $p(1 \times 1)$ LEED patterns confirmed pseudomorphic growth, which agrees with the observation of Dresselhaus *et al.*, who investigated 30 ML thick $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ films, with $x=0.47$, 0.64 , and $x=0.87$.¹⁷

Small C,O contamination were detectable at the end of the experimental run. With the surface magneto-optical Kerr effect (SMOKE) we measured the remanence as a function of temperature, thereby determining the Curie temperature T_C .

III. RESULTS AND DISCUSSION

Although the magnetic measurements focused on the invar concentration at $x \approx 0.65$, we investigated the growth of $\text{Fe}_x\text{Ni}_{1-x}/\text{Cu}(100)$ through the whole concentration range. We found RHEED oscillations of the (0,0) beam for up to 10 ML, which is exceptional in metal epitaxy and confirms good epitaxial growth even better than, e.g., Ni/Cu(100). As an example we show the RHEED oscillation for a $\text{Fe}_{77}\text{Ni}_{23}/\text{Cu}(100)$ film. This concentration is above the bulk invar concentration, where the transition to bcc has already occurred. This curve is qualitatively identical with the

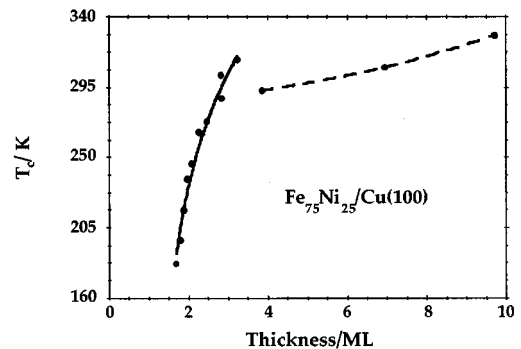


FIG. 4. Thickness dependence of T_C for a $\text{Fe}_{75}\text{Ni}_{25}$ alloy in the thin film limit and intermediate thickness. The solid line is fit to scaling law; the dashed line is a guide for the eye.

RHEED data for $\text{Fe}_x\text{Ni}_{1-x}$ films, with x below 65%. We can therefore rule out a sudden transformation into the bcc phase, which would also be accompanied by a drop in the RHEED intensity, like for $\text{Fe}/\text{Cu}(100)$.^{9,18}

In our previous studies,^{16,19} we have shown that the variation of the Curie temperature $T_C(n)$ with film thickness n can be described with the empirical scaling law:

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

Here n' is an empirical constant approximately equal to 1.¹⁶ The value of n' determines the thickness for which T_C starts to be finite. n_0 is another empirical constant, which has a value of approximately 3.5.¹⁶ $T_C(\infty)$ and λ are treated as independent fit parameters.

Equation (1) allows us to determine the $T_C(\infty)$ and compare this value with the bulk T_C of a given alloy concentration. Obviously, it would be desirable to measure T_C for very thick alloy films. This would give us directly $T_C(\infty)$, but the experimental window is limited, since at 400 K, Cu segregation is noticeable, as Auger spectra show. In Fig. 2 we show T_C as a function of the number of monolayers for several alloy films near the invar concentration. The solid line represents the fit to Eq. (1). We see that all these curves show a large slope near the onset of long-range order. We can now plot $T_C(\infty)$ as a function of concentration and compare it with the Curie temperature of bulk $\text{Fe}_x\text{Ni}_{1-x}$ alloys,¹² see Fig. 3. The vertical error bars reflect the uncertainty of the determination of $T_C(\infty)$, whereas the horizontal error bars indicate the variation of the concentration during the deposition. The important point here is that $T_C(\infty)$ does not follow the bulk behavior and varies little within the accuracy of the fit with Eq. (1). Also important is that the alloy films are all ferromagnetic, even beyond the invar concentration, which is in contrast to the bulk behavior, but is also at odds with the theoretical work of Abrikosov and co-workers.¹⁵

Extending the measurements for a $\text{Fe}_{75}\text{Ni}_{25}$ alloy film up to 10 ML thickness reveals a strong deviation from the scaling law; see Fig. 4. At 4 ML T_C has dropped when compared with the scaling curve, and increases only slowly with further deposition. These results indicate a structural relaxation/spin reduction occurring between 3 and 4 ML thickness.

ACKNOWLEDGMENT

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