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Magnetic instabilities in fcc $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ thin films

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We present the results obtained on $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloy films epitaxially grown on Cu(100). They are characterized by a fcc structure pseudomorphic to the substrate over a wide range of concentration and thickness. In particular, the martensitic transition which in bulk alloys occurs around the "Invar" concentration ($x \approx 0.65$) is suppressed. We report the concentration dependence at low temperature of the total magnetic moment and of its Fe-$3d$ and Ni-$3d$ projected components in such thin fcc $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloy films. Magnetic instabilities that might be associated with noncollinear spin alignments of Fe atoms are clearly observed for $x > 0.73$, where the magnetic moment decreases with increasing Fe concentration. In this Fe-rich concentration range the layers are still ferromagnetic and a magnetic moment is still observed, even on Ni atoms and at room temperature, up to $x = 0.86$. We also show how the variation of the magnetization in this region is correlated with a very small variation of the atomic volume (~1%).

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I. INTRODUCTION

For more than two decades, the study of artificial systems such as multilayers and/or superlattices, as well as metallic ultrathin films deposited on appropriate substrates has been attracting increasing interest. As the magnetic properties of thin films are very sensitive to the constraints imposed by the substrate, the first challenge was to change the crystallographic structure of a system in order to favor the appearance of a magnetic order which is different from the one in the bulk. Indeed, the magnetic and the structural properties are strongly correlated, especially in the case of $3d$ metals. Many $3d$ compounds present magnetic-volume instabilities characterized by high-magnetic-moment–high-atomic-volume (HM) states and/or low-magnetic-moment–low-atomic-volume (LM) states. In many cases, the energy difference between those states is so small that fluctuations between them become thermally accessible via low-energy excitations.[1] This situation results in several anomalies of the physical properties observed as a function of composition and external parameters such as temperature, magnetic field, and pressure.[2] The $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloys are considered as archetypes of this class of materials: for concentrations near $x = 0.65$, the thermal expansion coefficient is very small in a wide range of temperature (0–450 K). This is the so-called "Invar" effect.[3] The prevailing explanation attributes this effect to anharmonic lattice vibrations compensating the thermal excitations between a HM ferromagnetic state and a LM state.[2,4] Similarly, the departure of the temperature dependence of the total magnetic moment from a Bloch behavior, in the $\mathrm{Fe}_{0.65}\mathrm{Ni}_{0.35}$ Invar alloy, has been related to the existence of almost degenerate HM and LM states.

Many theoretical models have predicted the existence, in $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ systems, of stable or metastable states separated by a very small total energy difference (only 10 meV) but characterized by different equilibrium volumes. However, the transition from a HM to a LM phase as a function of temperature has not so far received any experimental evidence. Moreover, while there is a general agreement about the nature of the HM state, the magnetic order in the LM state is still controversial. *Ab initio* calculations done for the HM phase at $T = 0$ K in the case of an ordered $\mathrm{Fe}_3\mathrm{Ni}$ alloy, indicate that the magnetic moments range from $1.4\mu_\mathrm{B}$ to $1.9\mu_\mathrm{B}$ for Fe and from $0.5\mu_\mathrm{B}$ to $0.6\mu_\mathrm{B}$ for Ni.[5–8] The LM phase is predicted nonmagnetic[5,6] or magnetic with $0.5\mu_\mathrm{B}$ on Fe and no moment on Ni.[7,8] More recently, the dependence of the total energy with magnetization and volume of fcc $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloys has been calculated as a function of concentration.[9,10] At $T = 0$ K, a HM ground state is found in the case of Ni-rich alloys, with a concentration dependence of its moment following a Slater-Pauling curve. The energy difference between the HM and the LM states decreases as the iron concentration is increased. Above the instability concentration where the HM and LM states are degenerate the ground state is expected nonmagnetic.

The understanding of the magnetic behavior of $\mathrm{Fe}_x\mathrm{Ni}_{1-x}$ alloys is complicated by the fact that, as the Fe concentration x is increased, a fcc-bcc martensitic transition takes place near the instability limit of $x = 0.70$. The change in the crystal structure is accompanied by a strong downward deviation of the magnetic moment from the Slater-Pauling curve and a fast decrease of the Curie Temperature T_c. Simultaneously, a deviation of the lattice parameter from the Vegard law is observed. It has been suggested that such behavior may be connected to magnetic-volume instabilities as well.[11]

Such a complication can be overcome by growing thin Fe_xNi_{1-x} alloy films on an appropriate substrate.[12] It has been demonstrated that a fcc metastable phase can be stabilized for $x>0.65$ if a Fe_xNi_{1-x} film is deposited on Cu(100).[13,14] Recently, we have also been able to confirm by auger electron diffraction (AED) and x-ray photoelectron diffraction (XPD) that 10–30 ML thick Fe_xNi_{1-x} films grown on Cu(100) conserve their fcc crystal structure at the liquid nitrogen temperature, with no segregation of bcc clusters.[15,16] It has been shown that a similar situation occurs even in the case of Fe_xNi_{1-x}/Cu superlattices.[17] Thus, by using thin films epitaxially grown on copper, it becomes possible to considerably extend the investigation of the physical properties of fcc Fe_xNi_{1-x} alloys in a domain that would not be accessible with bulk samples. The exciting point is that this domain is precisely the one where a crossover between the total energies of the LM and HM states is expected.

In this paper, the possibility of magnetic-volume instabilities already proposed in the literature[18] is carefully examined. First, the growth mode, structure and atomic volume of the Fe_xNi_{1-x} films were determined by using reflection high energy electron diffraction (RHEED), Extended x-ray absorption fine structure spectroscopy (EXAFS), and x-ray diffraction (XRD). Second, the magnetic properties of these films were investigated by using x-ray magnetic circular dichroism (XMCD) and superconducting quantum interference device (SQUID) measurements.

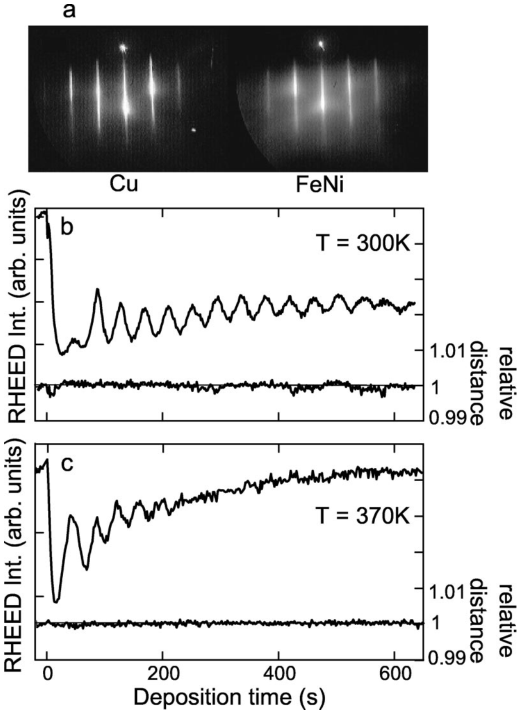

FIG. 1. (a) RHEED pattern observed on the initial Cu(100) surface and for 30-ML-thick $Fe_{84}Ni_{16}$ growth on Cu along [011] azimuth and RHEED oscillations observed during the growth of $Fe_{75}Ni_{25}$ alloy film on Cu (b) at 300 K and (c) at 370 K.

II. EXPERIMENTAL

All the *in situ* experiments (growth, structural, and magnetic analyses) were performed under UHV conditions with a base pressure of the order of $(1-5)\times10^{-10}$ hPa depending on the apparatus. The samples have been prepared and analyzed in several equipments: either at LURE in Orsay, at the LPM in Nancy, or at the ESRF in Grenoble.

A. Growth

It is well known that the bulk lattice constant of fcc Fe_xNi_{1-x} alloys varies between 3.55–3.59 Å in the concentration range $0.2<x<0.7$. Cu(001) was used as a substrate to evaporate thin fcc Fe_xNi_{1-x} films since, in this concentration range, the lattice mismatch between the alloy and the Cu substrate is less than 2%. Two different kinds of substrates are used: a Cu(100) single crystal or a 300 Å Cu buffer layer epitaxially grown on a MgO(100) single crystal. The first type of substrate is prepared by *in situ* cycles of Ar^+ sputtering and annealing. The growth of the Cu buffer layers on MgO(100) has been the object of another study.[16] The growth and/or annealing temperature have been optimized to obtain a buffer layer with a surface quality (roughness) that was comparable to that of the single crystal.

Fe and Ni were sublimated from separate effusion or electronic bombardment cells with typical growth rates from 0.3 to 5 Å/min, calibrated by using RHEED oscillations and/or quartz microbalance located at the place of the sample. The alloys were grown at room temperature (RT) in order to avoid interdiffusion at the interface with Cu. Their composition was fixed by varying the cells temperature, thus varying the evaporating rate of each element. The Fe and Ni relative concentrations were directly determined from the growth rate and density of each element. AES and x-ray photoelectron spectroscopy (XPS) were used to check the chemical quality of the films obtained and also to verify *a posteriori* the relative concentration of Fe and Ni in the alloys. The relative error done on the concentration calculated from the Fe and Ni flux or the spectroscopic techniques is about 10%. No oxygen contamination was found, although very small traces of carbon are sometimes present on the surface.

In Fig. 1(a), we compare two RHEED patterns obtained along the [011] azimuth, respectively, for Cu(100) and for a 30 ML Fe_xNi_{1-x} film with 86% Fe concentration. As expected, the surface structure of the alloy film is found to be pseudomorphic with the buffer layer. We have compared the in-plane lattice spacing measured after the alloy deposition with the in-plane lattice spacing measured on the initial Cu buffer layer surface in all the alloy concentration range reported in this paper without detecting any difference. Furthermore, as shown in Fig. 1(b) in the case of the epitaxial growth of a $Fe_{75}Ni_{25}$ film at room temperature, RHEED oscillations were clearly observed, indicating a layer by layer growth mode up to at least 15 ML. No variation of the in-plane lattice spacing was detected during the deposition.[19] In order to test any possible structural instability associated to small temperature variations, a similar experiment was performed using a 370 K growth temperature [Fig. 1(c)]. In this case the RHEED intensity oscillations rapidly disappear. As no roughness was evidence on the RHEED pattern during all the deposition time, this broadening is probably due to a progressive change of the growth mode by step flow. As for former experiments, no in-plane lattice spacing variation is observed at this growth temperature. A similar behavior is observed for all concentrations ($0.45<x<0.9$) and thickness (up to about 50 ML) investigated. From these RHEED results, we can safely conclude that there is no martensitic transition in the Fe_xNi_{1-x} thin films, in agreement with earlier results.[13]

B. Structure

RHEED is used to study the in plane structural quality of our films. The three-dimensional local environments of the thin alloy films have been studied by x-ray absorption near-edge spectroscopy (XANES) and EXAFS or by XRD.

EXAFS. EXAFS and XANES spectra were obtained on alloy films grown *in situ* on a Cu single crystal at the DCI storage ring at LURE. These experiments were performed, at 80 K, at the Fe and Ni *K* edge.

EXAFS is very sensitive to the local environment around a defined atom. The distinction between an fcc and a bcc structure is thus straightforward since these structures, in FeNi alloys, differ both by the number of nearest neighbors (NN's) and by the lattice constant. Moreover, by using the linear polarization of the synchrotron light, it is possible to separate the out-of-plane versus in-plane bond contributions and to determine the in-plane and out-of-plane NN's distances (R_{in} and R_{out}). Unfortunately, because of the small difference between the backscattering phases and amplitudes of Fe and Ni, it is not possible to separate their contributions to the local environment. We essentially used the experiments at the Ni edge in order to obtain an extra check of the alloys concentration.

The *in situ* XANES and EXAFS experiments were performed on 5- and 30-ML-thick Fe_xNi_{1-x} films deposited on Cu(100) for three different Fe concentrations 80, 75, and 60%. In Fig. 2(a) the XANES spectra measured at the Fe *K* edge on these FeNi films are reported. For comparison, we report in Fig. 2(b) the same XANES spectra measured at 80 K, at normal incidence, for three FeNi bulk polycrystals with different concentrations $x=0.75$, 0.65 (Invar concentration) and 0.45. The magnetic phase diagram of bulk FeNi alloys,[8] indicates the occurrence of a structural transition from fcc to bcc above $x=0.70$. This transition is clearly seen on the XANES spectra of bulk samples that are typical of either a fcc structure for ($x=0.65$, $x=0.45$) or a bcc structure ($x=0.75$). We do not observe the characteristic bcc spectral shape in the absorption spectrum of the thin films, indicating that the structure is fcc. These results show that no structural transition occurred in our thin films for the whole thickness and concentration range we have studied.

In the limit of accuracy of the EXAFS technique (±0.02 Å), no modification of the structure of the films is detected and the atomic volume variation we obtain (from 11.32 to 11.63 Å^3) as a function of the alloy concentration is within the experimental error bar (5%).

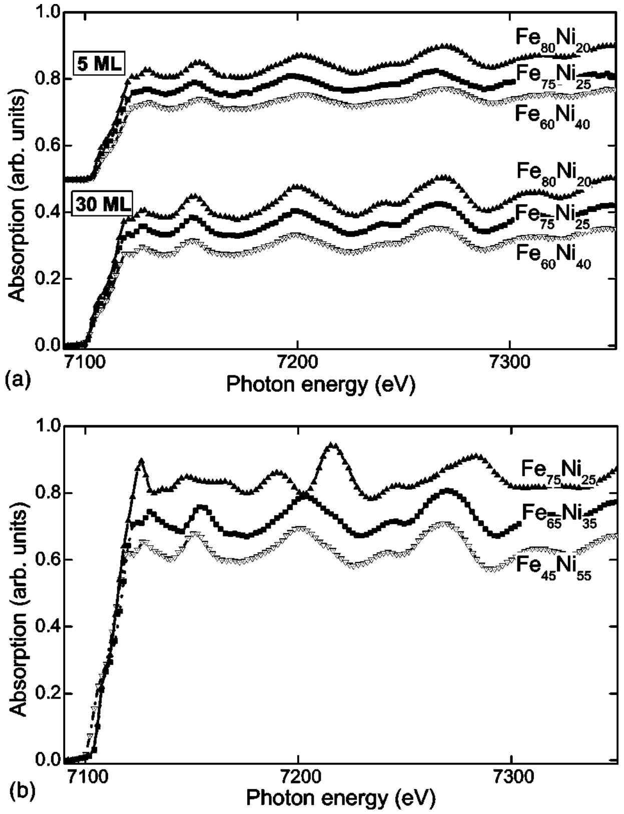

FIG. 2. Fe *K* edge XANES recorded at 80 K (a) for 5- and 30-ML Fe_xNi_{1-x} alloys grown on Cu(100) (from top to bottom $x=0.8$, 0.75, and 0.6) and (b) for three bulk Fe_xNi_{1-x} alloys (from top to bottom $x=0.75$, 0.65, and 0.45).

XRD. The structure of FeNi alloys was also studied, at room temperature, by x-ray diffraction (XRD) experiments performed at the Laboratoire de Cristallographie de Grenoble (at grazing incidence and detection with $\lambda=1.5418$ Å) and at the LPM (in the θ-2θ mode). Experiments were done on 50-ML-thick Fe_xNi_{1-x} alloy films on Cu/MgO and for several concentrations $x=0.81$, 0.75, 0.66, and 0.53. The samples were capped by a 50-ML-thick Cu layer. The experiments at grazing incidence were performed to get the $(hk0)$ peaks, and therefore the in-plane lattice spacing. A typical diffraction pattern obtained for $x=0.75$ is shown in Fig. 3(a). The first peak at $\theta=21.55°$ corresponds to a (200) reflection from MgO and the other one to a (200) diffraction plane of a sample with a fcc structure with $a_{\mathrm{RT\text{-}in}}=3.608\pm0.005$ Å. The XRD experimental error is estimated ±0.005 Å from the diffraction peaks of the MgO substrate. It should be noted that the contribution from the Cu and FeNi film cannot be separated. This means that the in-plane lattice spacing of Cu and FeNi films are equal. This is in agreement with RHEED in-plane lattice spacing measurement. The experiments in the θ-2θ mode were performed on the same samples in order to get the (001) distances and therefore the out-of-plane distance. The spectrum corresponding to the $x=0.75$ concentration is shown in Fig. 3(b). Two distinct contributions are now clearly visible in the (002) diffraction peak, one is due to Cu and the other to FeNi. A Gaussian fitting of these two features leads to a constant out-of-plane lattice spacing for Cu ($a_{\mathrm{Cu}}=3.611\pm0.003$ Å), and to a FeNi lattice spacing which varies between 3.591 ($x=66\%$) and 3.580 ($x=81\%$). These

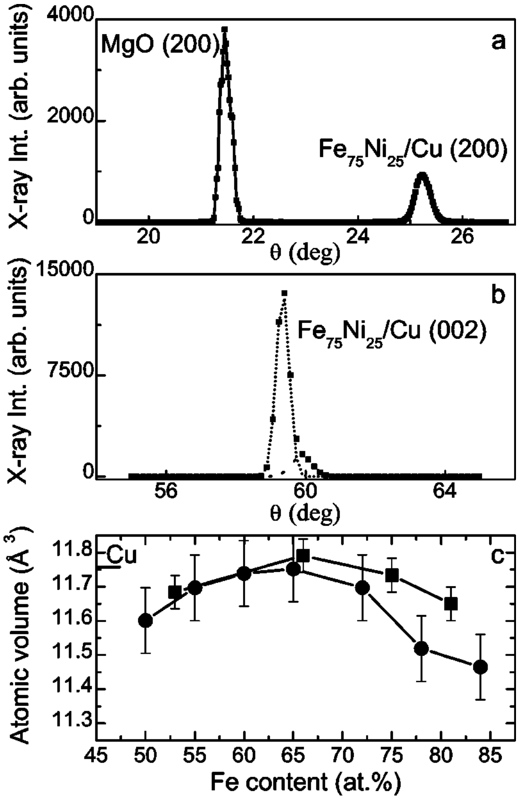

FIG. 3. XRD measurement taken at room temperature for 50 $Fe_{75}Ni_{25}$ ML/165 Cu ML/MgO(100) capped by 50-ML-thick Cu layer: (a) at grazing incidence (λ=1.5418 Å) and (b) in the $\theta-2\theta$ mode (λ=1.7889 Å). Lines are the results of fitting by using Gaussian peaks. (c) Evolution of the atomic volume obtained from XRD analysis on thin Fe_xNi_{1-x} alloy films grown on Cu(100) as a function of Fe concentration: black square (■) 50-ML thick (this study); black circle (●), obtained from RHEED and LEED measurements for an alloy thickness of 6 ML (Ref. 18).

values strongly depend on the fitting parameters and we may expect an error of about ±0.01 Å. We would like to stress that we did not detect any trace of bcc structure or even any mixture of fcc-bcc phases from these XRD data. Moreover, no experimental evidence for the possible growth of an ordered compound, e.g., Fe_3Ni, is found. The calculated atomic volume deduced from XRD is reported in Fig. 3(c) as a function of the Fe concentration. Our results are similar to the atomic volume variation obtained by Schumann *et al.*[18] from combined RHEED and LEED measurements on thick Fe_xNi_{1-x} films (6 ML). We can thus notice that there is no significant difference between a 6-ML-thick film and a 50-ML one. The atomic volume of the sample slowly decreases by 1% as the Fe concentration increases ($V_{RT}=11.79\pm0.05$ Å^3 for $x=0.66$ to $V_{RT}=11.65\pm0.05$ Å^3 for $x=0.81$).

C. Magnetism

The Fe and Ni local magnetic properties are determined by performing XMCD experiments both at the dragon beamline (ID12B) at the ESRF (Ref. 20) and at the SU23 beamline of the Super-Aco ring at LURE. In order to investigate the concentration dependence at low temperature of the Fe-3d and Ni-3d projected contributions to the total magnetic moment, XMCD measurements were performed both at the Fe and Ni $L_{2,3}$ absorption edges of thin Fe_xNi_{1-x} films $(0.5<x<0.9)$. The thickness of the samples (10 to 20 ML) was chosen in order to ensure a high Curie temperature ($T_c>450$ K) and a sufficient in-plane magnetic remanence along the (010) crystallographic axis. Similarly, the occurrence of possible effects due to the interface with Cu (enhanced magnetic moments, perpendicular anisotropy) was reduced by using such large thickness. Let us note that, in this thickness range, XMCD is sensitive to the whole volume of the sample and thus the surface of the alloy should weakly contribute to the spectra. This means that even small segregation of Ni on top, or the presence of a magnetic live surface layer, should not affect our conclusions about the bulk properties of the fcc alloys. The magnetic properties obtained in this way by XMCD are thus representative of the metastable fcc bulk phase.[13]

At ESRF, the XMCD experiments were performed at 10 K in the remanence mode using the circular polarized light emitted by the helical undulator Helios I (circular polarization rate ~85%).[21] The XMCD spectra were obtained at remanence by reversing the light helicity or by flipping the magnetization of the samples with a ~0.5 T pulsed magnetic field. XMCD measurements at room temperature were performed in the saturation mode on the SU23 beamline at LURE with a fixed circular light polarization (circular polarization rate is ≈52%) and switching an applied magnetic field of 1 T. All the dichroic signals are obtained measuring the drain current from the sample. They are normalized to the intensity at the Fe and Ni L_3 edges calculated after background removal (the method is the same as that described in Ref. 22), and corrected for the incomplete polarization and the nonparallel alignment between the sample magnetization and the photon spin. From general sum rules,[23] XMCD allows one to obtain the value of shell- and element-projected magnetic moments as soon as the shell occupancy is known. The Fe-3d and Ni-3d total magnetic moments can therefore be obtained assuming that the contribution of the magnetic dipole operator T_z is negligible, as theoretically[24] and experimentally[22] confirmed for 3d metals with cubic symmetry. Moreover, let us notice that the charge transfer between Fe and Ni is expected to be small.[8,10] This means that the Ni- and Fe-3d occupation numbers should not exhibit a significant dependence on the concentration and/or temperature and thus we can safely use the occupation numbers found in the literature. The total Fe-3d and Ni-3d magnetic moments were calculated using an occupation number of 7.4 electrons for Fe, as determined by fixed-spin-moment (FSM) band structure calculations for fcc ordered Fe_3Ni,[8] and 8.55 electrons for Ni, as experimentally obtained from Ni/Cu(100) thin films.[25]

In the following, $I^{+(-)}$ corresponds to the photon intensity measured for a propagating direction (anti)parallel to the sample magnetization. In Fig. 4, we show, as an example, the typical absorption spectra obtained at 10 K at the Ni and Fe $L_{2,3}$ edges for a 10-ML-thick $Fe_{62}Ni_{38}$ sample. Following the same method as in Ref. 22, we can define the fit parameters

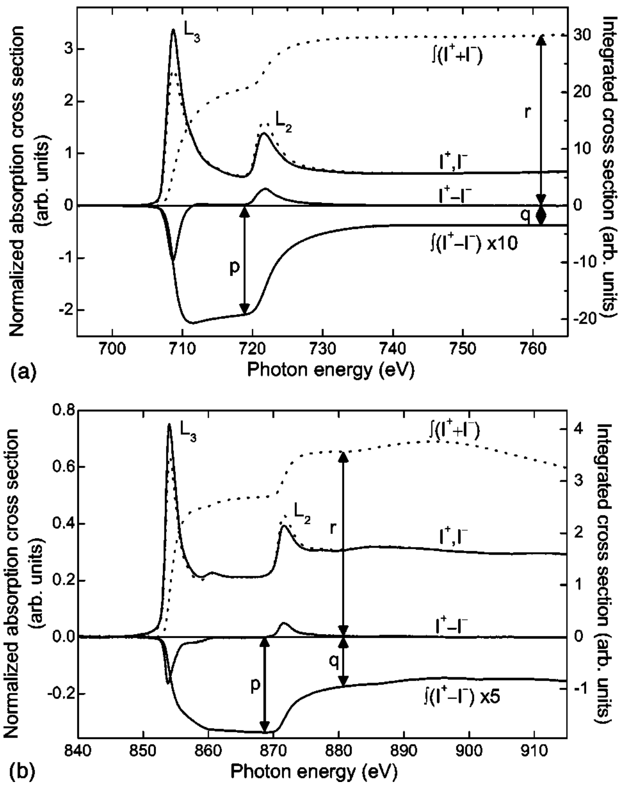

FIG. 4. XAS spectra at (a) Fe and (b) Ni $L_{2,3}$ edges of ten $Fe_{62}Ni_{38}$ ML/Cu(100) at 10 K. I^+ (I^-) are the normalized XAS spectra for (anti)parallel magnetic field light helicity. I^+-I^- indicates the XMCD spectrum. The values p, q, and r are the parameters needed in the sum rules analysis (see text).

(p, q, and r parameters), which allow the determination of both the orbital (m_l) and spin (m_s) magnetic moments.

Using the sum rules for the example shown in Fig. 4, the following values may be deduced for the magnetic moments (as soon as we normalize the XMCD intensities by the experimental ratio we determine between the magnetization at saturation M_S and at remanence M_R which is roughly equal to 2.2): $m_{\mathrm{XMCD}}^{\mathrm{Fe}} = 2.42 \pm 0.24 \mu_B$/at. ($m_l = 0.11 \pm 0.02 \mu_B$/at. and $m_s = 2.31 \pm 0.22 \mu_B$/at.) and $m_{\mathrm{XMCD}}^{\mathrm{Ni}} = 1.61 \pm 0.15 \mu_B$/at. ($m_l = 0.20 \pm 0.02 \mu_B$/at. and $m_s = 1.41 \pm 0.13 \mu_B$/at.). One can notice that the value for the Fe $3d$ magnetic moment agree with those expected from the calculations[26,27,28] and measured on bulk Invar alloy, while on the contrary the Ni $3d$ moment is considerably higher than the value measured in bulk Invar alloy.[26] The m_l/m_s ratio between the orbital and spin contributions to the magnetic moment does not depend either on the occupation number of the $3d$ states or by the reduction of the sample magnetization at remanence. This ratio can be obtained quite accurately [within 10% (Ref. 24)] by applying the sum rules to the experimental data. For the $Fe_{62}Ni_{38}$ sample (Fig. 4), m_l/m_s is equal to 0.05 ± 0.02 for Fe, which is identical to the value obtained in bulklike iron thin films,[22] and to 0.14 ± 0.02 for Ni which is close to the value obtained for bulklike nickel thin films.[29] The m_l/m_s ratio do not seem to vary as a function of the concentration (not shown). This observation reinforces the conclusion that there is no charge transfer between Fe and Ni atoms, as suggested from theory.[8,10] The variation of the Fe and Ni magnetic moment (corrected to the M_S/M_R ratio) with the alloy composition is reported in Fig. 5. In the whole range of concentration, where the Fe_xNi_{1-x} alloy films are in the fcc structure, Fe and Ni atoms have a net magnetic moment. The most striking result lies in the fact that, whereas we observe a 44% decrease of the Fe magnetic moment, this decrease is only 29% on the Ni atoms. Lets us note that there is no sudden change in the magnetic moments.

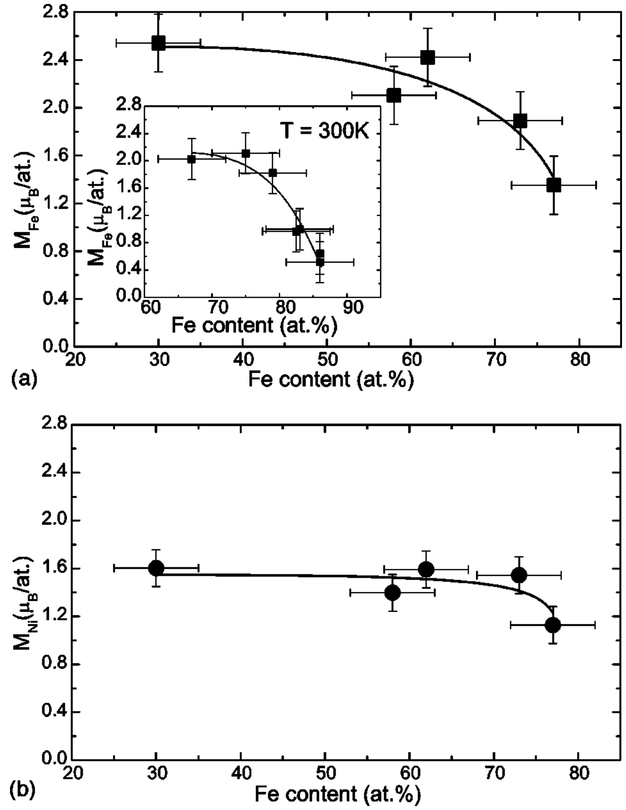

FIG. 5. (a) Fe-$3d$ and (b) Ni-$3d$ total magnetic moments (corrected to the M_S/M_R ratio) variation as a function of Fe concentration at 10 K, as derived by sum rules applied to XMCD spectra (experiments performed in remanence). The inset presents the Fe-$3d$ total magnetic moments versus Fe concentration at 300 K (experiments performed in static field). Lines are guides for the eye.

The room temperature behavior of the total magnetic moment (m_{XMCD}) on Fe as a function of the Fe concentration (x) is shown in the inset of Fig. 5(a). These results show that the Curie temperatures (T_c) of the studied samples are well above room temperature, for instance, even for $x = 0.86$ a net magnetization is measured at 300 K. As a consequence, the behavior of the Fe-$3d$ magnetic moments at 10 K, we observe for $0.6 < x < 0.7$, cannot be attributed to a decrease of the T_c, as it is the case for bulk Fe_xNi_{1-x} alloys.

The total magnetization of the FeNi alloys films were obtained from *ex situ* SQUID measurements performed at IPCMS in Strasbourg (France), applying a magnetic field up to ±5 T in the plane of the surface. SQUID was performed at 50 K on the thick (50-ML) Cu-capped Fe_xNi_{1-x} ($0.53 < x < 0.81$) samples grown on the Cu/MgO buffer. In Fig. 6, we compare the values of the average magnetic moments as de-

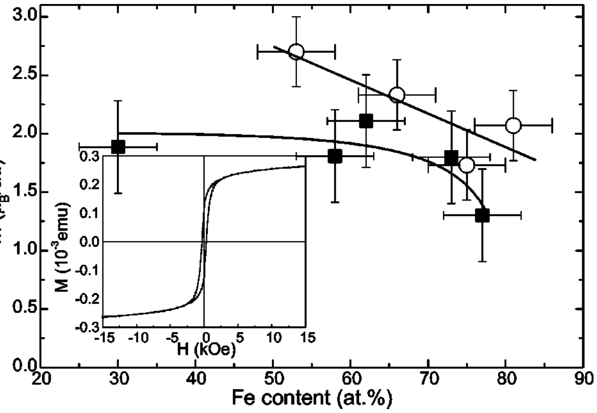

FIG. 6. Comparison between the total magnetic moment determined by XMCD (□) (see Fig. 3 for details) and by SQUID measurements (○) (at saturation) as a function of Fe concentration. SQUID experiments were recorded at 50 K on 50 Fe_xNi_{1-x} ML/ 165 Cu ML/MgO(100) capped by 50-ML-thick Cu layer. Lines are guides for the eye. The inset presents an example of SQUID hysteresis loop measured on 50-ML $Fe_{81}Ni_{19}$, showing the remanence to saturation ratio.

duced from the SQUID experiments performed at saturation (M_S) with those obtained from XMCD. As shown in the inset of Fig. 6, the ratio between the saturation (M_S) and the remanent (M_R) magnetization is exactly 2.2, which is almost constant (within the experimental error) in the whole range of concentration we investigated. When we correct the XMCD results by this factor both set of results become quite consistent. Particularly, the continuous decrease of the total magnetic moment is confirmed. Finally, it should be noted that the quite ''good'' agreement between the XMCD and SQUID results is an indirect validation of the numbers of $3d$ holes we used in the application of the sum rules.

III. DISCUSSION

Since by using XMCD we obtained the magnetic moments on both Ni and Fe atoms, our experiments give experimental evidence that the magnetic moment on Ni, in the whole concentration range, is large and even slightly higher than the moment of bulk Ni. There are different mechanisms that could explain the formation of such a stable ferromagnetic ground state even in the concentration and temperature range where, according to theory, a nonmagnetic ground state could be expected. One possibility is that an ordered macroscopic moment might result from fluctuations of the local concentration of nickel and iron, which would result in regions of the sample where x could be small enough for a HM state to be more stable. However, AES measurements made on different regions of the samples with a spatial resolution of ≈ 1 μm allow us to exclude the existence of such heterogeneities. Another possibility might be that an ordered Fe_3Ni phase (which is predicted to be ferromagnetic[10]) coexists with a random Fe_xNi_{1-x} alloy, which would be in a nonmagnetic ground state. However, the existence of such an ordered compound can be discarded due to the fact that the ordered Fe_3Ni should present a characteristic diffraction pattern in XRD that we never observed on any samples. From our RHEED, EXAFS, XRD, and AED (Ref. 15) results, we can also rule out the occurrence of ferromagnetic bcc clusters, even if the formation of such bcc crystallites has indeed been reported for $x > 0.74$.[17] Another hypothesis is that a magnetic ground state could be stabilized by a small tetragonal distortion.[10] Indeed, the layer is in-plane constrained by the substrate, but is free to contract or expand in the direction normal to the surface. In Ref. 30, it is reported that a lattice constant variation of $\pm 0.5\%$ corresponds to a 5% change of the Fe magnetic moment. As a consequence, the 1% variation of the atomic volume observed both in this work and in Ref. 18, cannot explain the 44% variation of the Fe magnetic moments we deduced experimentally.

The comparison between the concentration behaviors of the magnetic moments (on Fe and Ni) (Fig. 5) and the atomic volume [Fig. 3(c)], strongly suggests the existence of magnetic moment-volume instabilities in the concentration range varying from 60 to 80% of Fe which are characteristic of the Invar effect. We actually observe a decrease of the magnetic moment correlated with a small decrease of the atomic volume. Moreover, most of the experimental results we report in our paper, particularly the weak atomic volume variation (1%) correlated to the decrease of the Fe magnetic moments, which still stay in a stable ferromagnetic state, and the stabilization of the Ni magnetic moments, can be explained in the framework of recent theoretical calculations made on random fcc Fe_xNi_{1-x} alloys and based on noncollinear spin alignments[31,32] (the spins are tilted from the overall magnetization direction). Starting from a typical Invar system, i.e., a random fcc FeNi alloy, the authors show in Ref. 31 that a ferromagnetic HS ground state is associated to a high volume while at lower volume the ground-state spin configurations are in a noncollinear disordered state. The main effect is that increasing the Fe concentration (x), or decreasing the atomic volume, continuously reduces the spin correlation between Fe-Fe nearest neighbors until the occurrence at higher x values (or at small volumes) of a random spin distribution. On the contrary, for the Ni-Ni nearest neighbors sites, the spins remain essentially collinear and almost parallel with respect to the sample magnetization. As a result, the average magnetic moment of Fe atoms will decrease continuously to zero, whereas the magnetic moment on the Ni sites should stay more or less constant. This is exactly the situation we experimentally observe for the concentration dependence of the Fe- and Ni-$3d$ magnetic moment in our fcc FeNi alloys.

IV. CONCLUSION

In this paper, we confirm that thin Fe_xNi_{1-x} alloy films grow layer by layer on Cu(100) in the fcc structure. In particular, no formation of clusters with bcc-like structure is evidenced both by RHEED, EXAFS, and XRD. We observe a small variation of the atomic volume in 5 to 50 ML alloy films in a concentration range $0.53 \leqslant x \leqslant 0.81$. Furthermore, we address independently, from XMCD experiments, the behavior of the Fe and Ni magnetic moments as a function of concentration at low temperature in thin fcc Fe_xNi_{1-x} layers on Cu(100). In particular, the Fe moment decreases by 44%, while the reduction of the Ni moment is only 29%. All the results reported here may be explained by recent calculations, which show that the Invar effect may be due to some misalignment of the Fe moments with the Fe_xNi_{1-x} alloy composition. In this framework, the average Fe magnetic moment is expected to decrease when increasing the Ni concentration, whereas the Ni magnetic moment should not vary with the alloy composition.

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