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Subsurface growth of Ni atoms deposited on a Cu(001) surface

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The growth of Ni deposited at room temperature on a Cu(001) surface was studied by low energy electron diffraction (LEED). For ultrathin Ni films, ranging from 1 monolayer (ML) to 3 ML, subsurface Ni layers below a 1 ML thick Cu surface layer were found to give the best fit to LEED I/V (intensity versus beam voltage) characteristics. For 1 ML thick Ni film, first-principles calculation also predicted that subsurface growth of Ni film was favored energetically to overlayer growth. As the film thickness increased, however, the reliability factor became worse. This indicated that the structure gradually deviated from the ideal subsurface Ni layer below 1 ML surface Cu. The deviation was possibly caused by the increasing disorder in the atomic structure and/or in the chemical homogeneity near the surface. [S0163-1829(97)03112-3]

I. INTRODUCTION

Ultrathin films, especially ferromagnetic films on nonmagnetic substrates, have drawn enormous attention because they have shown magnetic properties quite different from those of bulk.[1] The magnetic properties of ultrathin films have been reported to be very sensitive to the atomic structure of both the thin film and the interface. So we must study the growth modes of thin films. The growth modes of ultrathin films depend both on the energetics and on various factors affecting the kinetics of the deposited atoms. The kinetic variables encompass the relative size of the deposited atoms and the substrate atoms, the deposition rate, the substrate temperature, the structural order of the substrate, the degree of contamination, and so on. The dependence on kinetic factors is due to nonequilibrium processes usually involved in the growth of thin films. This means that depending on the growth conditions of the thin films, various metastable atomic structures can be realized for the same system, possibly with different magnetic properties.[2]

Ni forms bulk alloys with Cu.[3] And for the (001) surface, the surface free energy of Ni (1.94 J/m^2) is higher than that of Cu (1.52 J/m^2).[4] So, it is expected that Cu atoms may form a layer on top of the deposited Ni film on a Cu(001) surface, or the copper atoms may form a surface alloy with Ni atoms. A recent molecular dynamics simulation[5] predicted that the diffusion barrier at the surface can be overcome at elevated temperature or via defect sites. Nevertheless, most experiments on this system have reported direct and indirect evidence[6,7] for pseudomorphic, overlayer growth of Ni films on Cu(001) surfaces. However, a recent x-ray photoelectron diffraction (XPD) study on the Ni/Cu(001) system reported the commencement of surface alloying in the temperature range of 260–300 K, and subsurface alloying between 300 and 450 K.[8] This result suggests the possibility of the interdiffusion of the Ni and Cu atoms near the surface during Ni film growth at room temperature. However, the experimental variables which dictate different growth behaviors for Ni atoms on the Cu(001) surface have not yet been clearly identified.

We therefore performed an extensive surface crystallographic study of ultrathin Ni films deposited on Cu(001) surfaces by the analysis of the I-V (intensity vs electron beam voltage) characteristics of low energy electron diffraction (LEED) spots.[9] In this work, the possibility of interdiffusion of the Ni and the Cu atoms was taken into account. The previous I/V analysis of the Ni/Cu(001) system by Montano *et al.*[10] did not include the possibility of interdiffusion. In addition, the atomic structure of an ultrathin Ni film was calculated by first-principles total energy calculation[11] and then compared with results of dynamic LEED crystallography.

In the following section, our experimental procedure is described. In Sec. III, first-principles calculation and its predictions on the atomic structures of Ni films are described. Then, in Sec. IV, the results from our LEED I-V analysis are compared with theoretical results. In the final section, a summary and conclusion are given.

II. EXPERIMENT

The Cu(001) surfaces were prepared through mechanical polishing with alumina powder of size down to 0.05 μm followed by cycles of annealing (up to 1100 K) and Ar ion sputtering. To remove residual carbon atoms, the sample was often annealed at around 500 K in an oxygen environment of

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10^{-8} Torr for about 20 min. The Ni thin film was deposited by evaporating Ni from a Ni wire (99.999% pure) wound around a tungsten wire (99.99% pure). During the Ni deposition, the substrate temperature was in the range of 300–335 K. The thickness of the deposited Ni film was determined from the relative intensities $I_{\mathrm{Ni}}/I_{\mathrm{Cu}}$ of Auger electron peaks, Ni (LMM, 848 eV), and Cu (LMM, 920 eV).[12] For a Ni layer of thickness Δx lying between x and $x+\Delta x$ from the surface,

$$
\begin{aligned}
I_{\mathrm{Ni}}/I_{\mathrm{Ni}}^0 =& [1-\exp(-\Delta x/\lambda_{\mathrm{Ni}}\cos\theta)]\exp(-x/\lambda_{\mathrm{Cu}}\cos\theta)\\
&\times\exp(-x/\lambda_I),\\
I_{\mathrm{Cu}}/I_{\mathrm{Cu}}^0 =& [1-\exp(-x/\lambda_{\mathrm{Ni}}\cos\theta)]\exp(-x/2\lambda_I)\\
&+I_{\mathrm{Cu}}^0\exp(-x/\lambda_{\mathrm{Cu}}\cos\theta)\exp(-dx/\lambda_{\mathrm{Ni}}\cos\theta)\\
&\times\exp[-(x+\Delta x)/\lambda_I]. \qquad (1)
\end{aligned}
$$

Here, $I_{\mathrm{Ni(Cu)}}^0$ is the Auger intensity for LMM transition of Ni (Cu) if the target is pure bulk Ni (Cu), and $\lambda_{\mathrm{Cu\,(Ni)}}$ is the escape depth of the Cu (Ni) Auger electron. λ_I is the mean free path of the incident electron of energy (3 KeV), θ is the CMA acceptance angle, 42.3°. The escape depths were obtained from the universal curve. The relative sensitivity factor $I_{\mathrm{Ni}}^0/I_{\mathrm{Cu}}^0$ was obtained from the database from Physical Electronics Industries, Inc.[13]

The cleanliness of the surface was determined from the AES (Auger electron spectroscopy) data, which showed no definite peaks related to likely contaminants such as carbon and oxygen. Pseudomorphic growth of the film was assumed from the $p(1\times1)$ LEED pattern. The LEED spot size after Ni deposition was a little broader, indicating a smaller terrace size than that of a clean Cu surface, but still the background intensity was very low, displaying a sharp LEED pattern up to an incident electron energy of 400 eV. The base pressure of the chamber was in the low 10^{-10} Torr range, and the chamber pressure during Ni evaporation rose up to 5×10^{-10} Torr. The deposition rate was about 1 ML per 3 min. The residual magnetic field was kept below 0.05 G in all three perpendicular directions by employing three perpendicular Helmholtz coils. The symmetry of our experimental geometry and the effective removal of the residual magnetic field was confirmed from the virtual identity of the I/V characteristics of symmetrically equivalent spots. To obtain the LEED I/V characteristics from the LEED patterns, a video LEED system was employed, which proved to give reliable results for clean Cu(001) and Ag(001) surfaces.[14]

III. FIRST-PRINCIPLES CALCULATION

To determine the stable structure for this Ni on the Cu(001) system, the total energies are calculated as functions of all of the Ni-Cu interlayer spacings for two cases. In one case, the Ni 1 ML remains as an overlayer, and in the other case, the Ni atoms diffuse into the Cu substrate to form a subsurface layer. We employ the single slab approximation. The single slab for the overlayer Ni system is composed of seven layers of Cu(001) with a Ni monolayer on each side of the slab [Ni/Cu(001)]. The subsurface Ni system is composed of five layers of Cu(001) with a Ni monolayer and a Cu layer on each side of the slab, [Cu/Ni/Cu(001)]. The Kohn-Sham equations,[15] established in the local spin density (LSD) approximation, are solved self-consistently employing the full potential linearized augmented plane wave (FLAPW) method.[16] The explicit form of von Barth and Hedin[17] is used for the exchange-correlation potential for spin-polarized calculations. The core electrons are treated fully relativistically, whereas the valence electrons are treated semirelativistically, i.e., including all relativistic terms but spin-orbit coupling.[18]

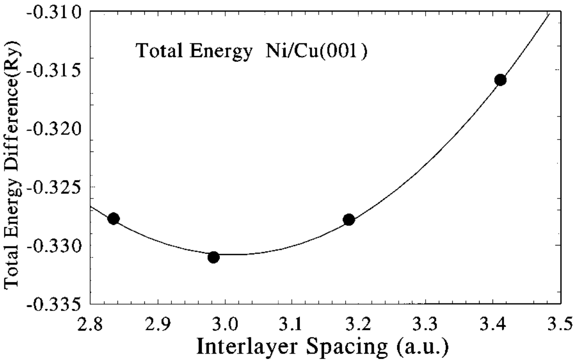

FIG. 1. A plot of ε_1 (solid line), the total energy difference per unit cell of the Ni/Cu(001) system as a function of the Ni-Cu interlayer spacing. The solid circles are calculated data points, which are fitted to a parabola. The total-energy minimum is found at an Ni-Cu spacing of 3.01 atomic units.

Figure 1 shows the total energy difference of the Ni/Cu(001) system as a function of the Ni-Cu interlayer spacing. The solid circles are our calculated data points, which are fitted to a parabola. The total energy minimum is found to be at a Ni-Cu spacing of 1.592 Å, which is 10.5% smaller than from the average of bulk Cu-Cu and Ni-Ni bond lengths. In this geometry, the magnetic moment per Ni atom is $0.23\mu_{\mathrm{B}}$. The large relaxation of the Ni-Cu bond length possibly has its origin in the large reduction of Ni magnetic moment on the surface.[19] Figure 2(a) shows the total energy of the Cu/Ni/Cu(001) system as a function of the interlayer spacing of Cu(S)-Ni while the interlayer spacing of Ni-Cu(S-2) is fixed at the interlayer spacing of the bulk Ni. The total energy minimum is found at a Cu(S)-Ni spacing of 1.577 Å. In Fig. 2(b), the calculated total energy is presented as a function of the interlayer spacing of the Ni-Cu(S-2) with the Cu(S)-Ni layer spacing fixed at the optimum spacing found in Fig. 2(a). The total energy minimum is found to be at an Ni-Cu(S-2) interlayer spacing of 1.713 Å. The total energy of the subsurface Ni system is smaller by 30 mRy per atom than that of the overlayer Ni system. In the subsurface system, the magnetic moment per Ni atom is very small, $0.18\mu_{\mathrm{B}}$, compared to that of the Ni atom in its bulk state, $0.57\mu_{\mathrm{B}}$, and smaller than that of the Ni on the Cu(001) surface, $0.23\mu_{\mathrm{B}}$. Further details of the magnetic and electronic properties will be published elsewhere.[20]

IV. RESULTS AND DISCUSSION

In Fig. 3 are shown I/V characteristic curves of (10) LEED spot from a Cu(001) surface on which 1 to 3 ML (monolayers) of Ni has been deposited.[21] When 1 ML of Ni film has been deposited, the peak associated with a clean

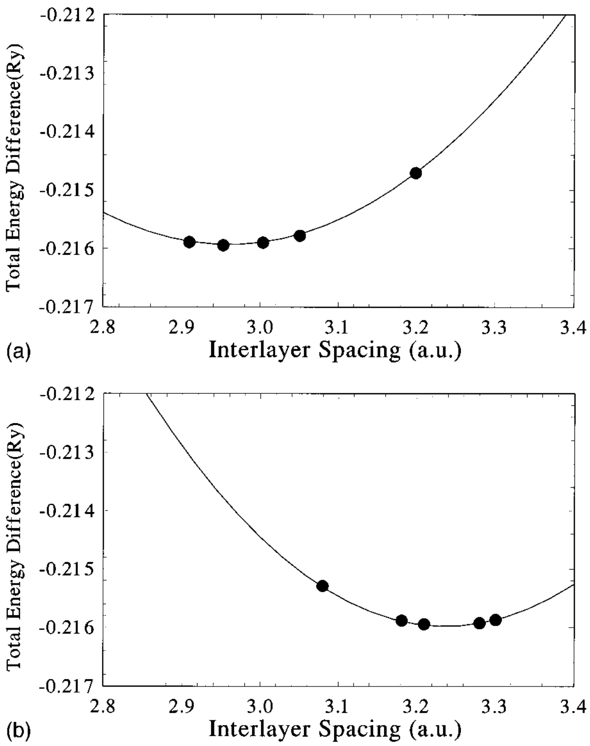

FIG. 2. The total energy difference of the Cu/1 ML-Ni/Cu(001) system as a function of the interlayer spacing of (a) Cu(S)-Ni and (b) Ni-Cu(S-2). In (a), the spacing of the Ni-Cu(S-2) is fixed at the bulk Ni-Ni spacing, and in (b), the Cu(S)-Ni spacing is fixed at the optimum distance determined from (a). The total-energy minimum is found at Cu(S)-Ni and Ni-Cu(S-2) interlayer spacings of 2.963 and 3.094 a.u., respectively.

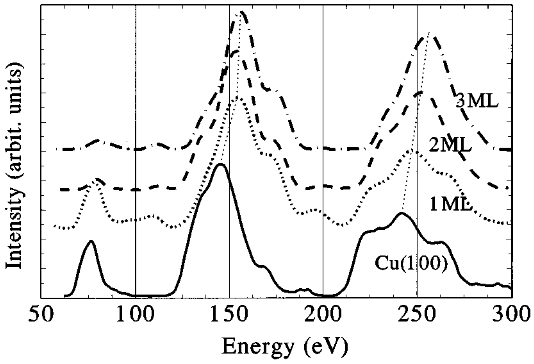

FIG. 3. LEED I/V characteristic curves of a (10) beam for various thicknesses as of the Ni films deposited on a Cu(001) surface: (a) a clean Cu(001) surface, (b) a 1-ML-thick Ni film, (c) a 2-ML-thick Ni film, and (d) a 3-ML-thick Ni film. The overall shapes are similar except for the shifts of peak positions as indicated by the dotted lines.

copper surface is still found as a shoulder in the low energy region around 80 eV. When more Ni atoms are deposited, this intact feature gets smaller due to the gradual wetting of the surface by the Ni layer. The peak positions are shifted toward higher energies as the film gets thicker. This suggests that the interlayer distances near the surface decrease because the peak positions are associated, in the single-scattering approximation, with the Bragg condition in the z direction.[22] Hence, the blueshifts of the peak positions imply that the atomic structure near the surface is influenced more and more by the deposited Ni, which has a smaller interlayer spacing than that of Cu in its bulk state. These shifts of the peak positions do not seem to originate from the inner potential shift or the work-function change, since the peak positions associated with clean Cu remain intact, and the amounts of the peak-position shifts are too large to be attributed to a possible work-function change caused by the deposition of the Ni thin film. This qualitative information on the atomic structure gives a clue in the search for the atomic structure by dynamic LEED analysis, i.e., the residence of the deposited Ni atoms near the surface.

Quantitative information on the atomic structure near the surface was acquired from the analysis of the I/V characteristics of the LEED spot intensities using a recently developed automated tensor LEED package.[23] In Table I we show the summary of all the model structures employed to fit the experimental LEED I/V characteristics for a system with 1 ML Ni film[21] and their respective optimum R (reliability) factors (Pendry)[24]. The optimum structure was found to be the 1 ML thick subsurface Ni film below a 1 ML thick Cu layer, which was *consistent* with the model according to which the AES calibration of the thickness of the Ni film was carried.[21] In Fig. 4(a) are shown the experimental I-V curves with the best-fit theoretical curves for this 1 ML thick Ni film. For the thin film calibrated by AES as 1 ML thick Ni *overlayer* system, we have also taken LEED I/V data. But we could

TABLE I. The optimum atomic structures reached from various reference structures when a 1-ML equivalent of Ni is deposited. d_{ij} means the difference in the relative interlayer spacing between the ith layer and the jth layer from the interlayer spacing of bulk Cu(001). The Pendry R factor was employed. The ''theory'' shows the results from our first-principles calculation for Cu/Ni/Cu(001) structure.

Reference structure	d_{12} (%)	d_{23} (%)	d_{34} (%)	d_{45} (%)	d_{56} (%)	R factor
Ni/Cu	−7.0	1.4	−1.1	3.9	4.3	0.311
Ni/Ni/Cu	−2.4	−1.0	−0.5	2.2	0.1	0.249
Cu/Ni/Cu	−1.8	−1.9	0.6	1.8	−0.6	0.204
Cu/Ni/Ni/Cu	−1.6	−2.2	−1.3	−1.5	3.1	0.292
Cu/Cu/Ni/Cu	−2.3	−0.6	−0.4	−2.9	1.4	0.305
Cu/Cu/Ni/Ni/Cu	−2.3	−0.7	−0.4	−2.25	0.3	0.294
Ni/Cu/Ni/Cu	−4.6	2.5	0.3	0.9	1.0	0.298
Theory	−12.6	−5.1				

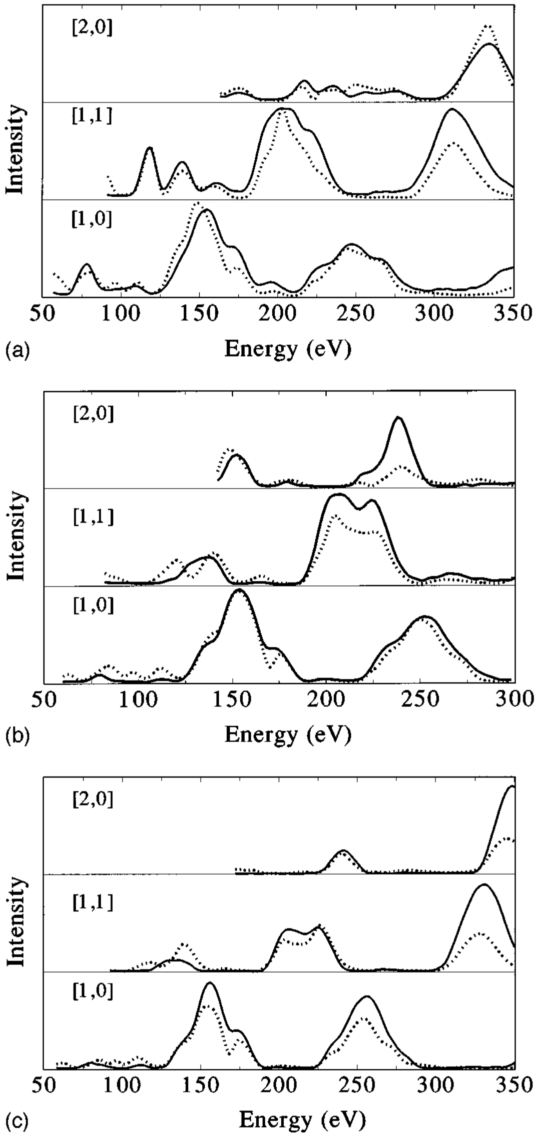

FIG. 4. The experimental LEED I/V curves (solid lines) of (1,0), (1,1), and (2,0) beams for systems with (a) 1-ML-, (b) 2-ML-, and (c) 3-ML-thick Ni deposited on a Cu(001) surface. The dotted lines are the theoretical curves which give the best fit to the experimental data. In all cases, the best fit was obtained with subsurface Ni layers below a 1-ML-thick Cu capping layer.

not fit the I/V data, when we modeled the film as a Ni overlayer system with the reliability factor, R_{Pendry} less than 0.4. These results support subsurface growth of Ni rather than overlayer Ni growth under the current experimental condition, as anticipated from the results of the first-principles calculation.

The subsurface growth of Ni was tested by dosing 100 L (Langmuir) of oxygen with the oxygen partial pressure maintained at around 5×10^{-7} Torr after Ni is deposited. During oxygen dosing, the sample temperature was kept at room temperature. Ni is very reactive with oxygen, and when oxygen is exposed to the Ni surface, it forms a $p(2\times2)$ or $c(2\times2)$ adlayer for oxygen dosages much less than 100 L.[7] In the current experiment, however, no superstructure was found in the LEED pattern, and no AES signal associated with oxygen was detected. On the contrary, the Cu(001) surface is very inert to oxygen and it needs an oxygen dosage much more than 100 L to form an oxygen overlayer structure.[25] These measurements suggest that the surface layer is formed by relatively inert Cu, and reinforces the conclusion of subsurface growth of the Ni atoms deposited on the Cu(001) surface.

The formation of clusters and/or a disordered alloy of Ni and Cu near the surface in registry with the bulk terminated Cu(001) surface is a possibility. Those structures also preserve the $p(1\times1)$ LEED patterns. However, we expect a well segregated Ni sublayer growth which gives a reasonably good R-factor value, $R=0.204$, although the possibility of Ni and Cu alloying in small amounts cannot be excluded entirely by the current LEED analysis. For detailed information on the chemical stoichiometry for each layer, the application of chemical tensor LEED (Ref. 26) is highly desirable and is in progress.

The interlayer distances for Cu(S)-Ni and Ni-Cu(S-2) (Table I) from both the first-principles calculation and the LEED I-V analysis show inward relaxation from the average bond length of Cu-Cu and Ni-Ni in their bulk states. This relaxation is attributed not only to the relatively small lattice size of Ni in its bulk state, but to the large reduction of the magnetic moment of Ni as given in the first-principles calculation and the resulting lowered magnetic pressure.

Martensson *et al.*[8] reported a pronounced photoelectron peak (Ni 2p) at an angle 45° from the surface normal

TABLE II. The optimum atomic structures reached from various reference structures when a 2-ML equivalent of Ni is deposited. d_{ij} means the difference in the relative interlayer spacing between the ith layer and the jth layer from the interlayer spacing of the bulk Cu(001). The Pendry's R factor was employed.

Reference structure	d_{12} (%)	d_{23} (%)	d_{34} (%)	d_{45} (%)	d_{56} (%)	R factor
Ni/Cu	−3.1	−4		−4.6	10	0.409
Ni/Ni/Cu	−2.6	−4	−1.5	4.3	−3.5	0.377
Cu/Ni/Cu	−1.9	−4	−3.1	1.6	−0.9	0.331
Cu/Ni/Ni/Cu	−1.6	−3.7	−3.8	−1.2	2.5	0.289
Cu/Ni/Ni/Ni/Cu	−1.8	−3.7	−3	−3.2	2.8	0.303
Cu/Cu/Ni/Ni/Cu	−2.5	−4.8	−6.5	62.7	54	0.408
Cu/Cu/Ni/Cu	−2.8	−4.1	−3.6	2.2	2.7	0.349

TABLE III. The optimum atomic structures reached from various reference structures when a 3-ML equivalent of Ni is deposited. d_{ij} means the difference in the relative interlayer spacing between the ith layer and the jth layer from the interlayer spacing of the bulk Cu(001). The Pendry's R factor was employed.

Reference structure	d_{12} (%)	d_{23} (%)	d_{34} (%)	d_{45} (%)	d_{56} (%)	R factor
Ni/Cu	−3.5	−7.5	0.4	6.6	−3.3	0.561
Ni/Ni/Cu	−4.9	−4.3	−4.1	−1.2	1.5	0.41
Cu/Ni/Cu	−2.1	−5.3	−3.4	0.6	−0.1	0.393
Cu/Ni/Ni/Cu	−3.8	−4.5	−4.8	−4	4.5	0.377
Cu/Ni/Ni/Ni/Cu	−3.4	−4.9	−3.7	−5.6	5.9	0.356
Cu/Cu/Ni/Ni/Cu	−3.9	−3.7	−4.9	23.7	−20	0.432
Cu/Ni/Cu/Ni/Cu	−1.6	−7	−2.5	7.5	−3.5	0.458

alongthe [110] direction, when 0.67 ML of Ni is deposited on the Cu(001) surface. This observation was interpreted as evi-dence for the residence of Ni below a 1 ML Cu surface layer. Their XPD curve of a Ni 2p peak showed photoelectron intensities at angles, ranging from $\theta = 6^\circ$ to 60° from the surface normal direction. If there were a substantial amount of Ni atoms in the third layer or deeper from the surface, there should be another peak of photoelectron intensity (Ni 2p) at $\theta = 0^\circ$. Then, we expect to find a noticeable increase of the Ni 2p intensity at $\theta = 6^\circ$, the minimum angle of observation, since the full width at half maximum of the peak at 45° is about 16°. But there was found no increase of the photoelectron intensity near the angle, $\theta = 6^\circ$. This result impies that the Ni atoms did not reach layers deeper than the second layer from the surface. XPD cannot tell whether there are Ni atoms on the surface layer or not. For the comparison with our conclusion, further study, such as oxygen titration experiment, is required. However, we note that the study of Martensson *et al.* is, at the least, not contradictory to our result.

With 2 ML thick Ni layers and 3 ML thick Ni layers[21] deposited on the Cu(001) surface, the same analyses were repeated [see Figs. 4(b) and 4(c)]. Structures made of a one monolayer thick Cu capping layer with 2 ML and 3 ML thick subsurface Ni layers were found to be optimum structures, respectively, as shown in Tables II and Tables III. As the film thickness increases, the R factors get larger. This indicates the growth of disorder in the atomic structure and/or in the chemical composition near the surface. However, for 1 and 2 ML thick Ni films the R factor is still good enough to identify the growth mode of the Ni unambiguously as subsurface growth. Thus, the picture of the initial growth of a Ni on Cu(001) surface supported by the current study is a segregated subsurface Ni layer growth below a 1 ML thick Cu capping layer which behaves as a surfactant. This growth behavior is not unique for a Ni/Cu(001) system, but also found for an Fe/Au(001) system.[27] Recently, a model study on the surfactant effect of the floating substrate layer was also reporteded for a Ni/Ag(001) system.[28]

V. CONCLUSION

The initial growth mode of the Ni atoms on Cu(001) was studied by LEED I/V analysis and first-principles total energy calculation. Both the experimental results and the theoretical predictions are consistent with each other on the initial growth mode of Ni on a Cu(001) surface: the growth of a subsurface Ni film below a 1 ML thick Cu surface layer. The subsurface growth of Ni, instead of the widely reported metastable overlayer growth of Ni, implies that under the current experimental condition there are effective channels for Cu and Ni atoms to interdiffuse, e.g., the structural or the chemical defect sites as found in the recent STM (scanning tunneling microscope) study.[29] The thin film growth depends very sensitively on various energetic and kinetic variables, so the growth mode for the same system may vary widely according to experimental conditions. The discrepancies in the literature on the growth mode of Ni on Cu(001) might have originated for the same reason. To clarify this problem, detailed studies on the growth mode with well controlled experimental variables are required.

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[1] *Ultrathin Magnetic Structure*, edited by J. A. C. Bland and B. Heinrich (Springer-Verlag, Berlin, 1994); *Magnetism and Structure in Systems of Reduced Dimension*, edited by R. F. C. Farrow (Plenum, New York, 1993).

[2] G. A. Prinz, J. Magn. Magn. Mater. **100**, 469 (1991).

[3] R. Hultgren, P. Desai, D. Hawkins, M. Gleiser, and K. Kelley, *Thermodynamic Properties of Binary Alloys* (American Society of Material, Metals Park, 1973); *Cohesion in Metals*, edited by F. R. de Boer and D. G. Perifor (North-Holland, Amsterdam, 1988).

[4] V. K. Kumikov and K. B. Khokonov, J. Appl. Phys. **54**, 1346 (1983).

[5] T. J. Raeker and A. E. DePristo, J. Vac. Sci. Technol. A **10**, 2396 (1992).

[6] Y. Idzerda and G. A. Prinz, Surf. Sci. **284**, L394 (1993); J. Zhang, Z.L. Han, S. Varma, and B. P. Tonner, *ibid.* **298**, 351 (1993); M. Tischer, D. Arvantis, T. Yokoyama, T. Lederer, L. Tröger, and K. Baberschke, *ibid.* **307-309**, 1096 (1994); F. Huang, M. T. Kief, G. J. Mankey, and R. F. Willis, Phys. Rev. B **49**, 3962 (1994).

[7] J.-S. Kim, M. H. Mohamed, and L. L. Kesmodel, Surf. Sci. **260**, 185 (1992).

[8] B. Hernnas, M. Karolewski, H. Tillborg, A. Nilsson, and N. Martensson, Surf. Sci. **302**, 64 (1994).

[9] M. A. van Hove and S. Y. Tong, *Surface Crystallography by LEED* (Springer-Verlag, Berlin, 1979).

[10] M. A. Abu-Joudeh, B. M. Davies, and P. A. Montano, Surf. Sci. **171**, 331 (1986).

[11] A. J. Freeman and R-Q. Wu, J. Magn. Magn. Mater. **100**, 497 (1991).

[12] The thickness calibration of the AES signals is based on a specific growth model of the deposited film. However, in this paper, the growth mode of the thin film is under investigation, so all the possible growth modes should be examined. For the respective growth mode, the calibration was carried out as described in the following references: D. Briggs and N. P. Seah, *Practical Surface Analysis by Auger and X-ray Photo Electron Spectroscopy* (John Wiley, New York, 1990); S. Mroz, Prog. Surf. Sci. **46**, 377 (1994).

[13] L. E. Davis, N. C. MacDonald, P. W. Palmberg, G. E. Riach, and R. E. Weber, *Handbook of Auger Electron Spectroscopy*, 2nd ed. (Perkin Elmer Corporation, Eden Prairie, 1976).

[14] J. H. Kim, D. J. Kim, S. M. Lee, J-S. Kim, D. H. Byun, and H. G. Min, J. Kor. Vac. Soc. **2**, 515 (1993).

[15] W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).

[16] E. Wimmer, H. Krakauer, M. Weinert, and A. J. Freeman, Phys. Rev. B **24**, 864 (1981), and references therein.

[17] U. von Barth and L. Hedin, J. Phys. C **5**, 1629 (1972).

[18] D. D. Koelling and B. N. Harmon, J. Phys. C **10**, 3107 (1977).

[19] V. L. Moruzzi, P. M. Marcus, K. Schwarz, and P. Mohn, Phys. Rev. B **34**, 1784 (1986).

[20] S. C. Hong and R. T. Rho (unpublished).

[21] The thicknesses calibrated from the AES data are in reference to Cu/n-Ni/Cu(001) ($n=$ 1,2,3) structures, respectively.

[22] G. Ertl and J. Küpper, *Low Energy Electrons and Surface Chemistry* (VCH, Weinheim, 1985), p. 230.

[23] P. J. Rous, Prog. Surf. Sci. **39**, 3 (1992); P. J. Rous, M. A. van Hove, and G. A. Somorjai, *ibid.* **226**, 15 (1990); A. Wander, M. A. van Hove, and G. A. Somorjai, Phys. Rev. Lett. **67**, 626 (1991).

[24] The Auger signals corresponding to the 1 ML-Ni/Cu(001) structure can also be calibrated to be Cu/1.64 ML Ni/Cu(001). Considering the rough assumptions in the thickness calibration of the AES signals, wild variations of model structures were taken.

[25] M. Wuttig, R. Franchy, and H. Ibach, Surf. Sci. Lett. **224**, 979 (1989).

[26] R. Döll, M. Kottcke, and K. Heinz, Phys. Rev. B **48**, 1973 (1993); R. Döll, M. Kottcke, and K. Heinz, Surf. Sci. **304**, 309 (1994).

[27] F. J. Himpsel, Phys. Rev. B **44**, 5966 (1991)

[28] J. M. Roussel, A. Saúl, G. Treglia, and B. Legrand, Surf. Sci. **352-354**, 562 (1996).

[29] A. K. Schmid, D. Atlan, H. Itoh, B. Heinrich, T. Ichinokawa, and J. Kirschner, Phys. Rev. **B 48**, 2855 (1993).