Surface characteristics of epitaxially grown Ni layers on Al surfaces: Molecular dynamics simulation

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Soon-Gun Lee and Yong-Chae Chung

Surface characteristics of epitaxially grown Ni layers on Al surfaces: Molecular dynamics simulation

Soon-Gun Lee and Yong-Chae Chung[a)]
Department of Materials Science and Engineering, Hanyang University, Seoul 133-791, Korea

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The deposition behavior for Ni thin film growth on Al substrates of various orientations according to the incident energy of adatoms was investigated by molecular dynamics simulation. In spite of the low adatom incident energy of 0.1 eV, Ni–Al intermixing occurred actively at the surfaces of Al(001), Al(011), and Al(111) at 80 K and Ni atoms apparently favored the island growth mode irrespective of the Al surface orientation. The highest surface roughness was shown for the case of Al(111) surface. The steering effect, which results in rougher surface, was significantly observed at low incident energy. The steering effect was quantitatively investigated through the extensive measurement of the trajectory and deposition flux of atoms with the variation of incident energy near the artificially structured Ni step positioned on Al surfaces. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2355440]

I. INTRODUCTION

Understanding and controlling the evolving surface morphology of epitaxially grown thin films have been the essential issues of surface science.[1,2] It has been generally known that the interface between metals or metal and insulator had a critical role in the spin polarization of the junction, and the spin polarization for the tunneling characteristics is strongly dependent on the structural quality of the tunnel junction.[3] For applications in magnetic devices, ordered and uniform thin film is especially required for the coherency of the devices.[4] The film morphology and corresponding properties are mainly dependent on the substrate temperature and the adatom incident energy.[5,6] Interestingly, it has been recently reported that the growth mode could be changed from island growth mode to layer-by-layer growth mode with the adatom incident energy of 10 eV.[7]

The surface energy difference and the degree of lattice mismatch of the substrate and adsorbate material could result in different deposition behaviors of adatoms.[8] One particular important factor for determining the evolution of the surface morphology is the behavior of incoming atoms near the substrate surface. Specifically, surface alloying could be caused by the local acceleration[9,10] coming from the attractive forces between the incoming adatom and surface atoms, and island growth mode is generally known to be occurred by the steering effect[11–13] due to atomic attraction near the substrate.

For Ni/Al system, it is relatively difficult to achieve layer-by-layer growth [Frank–van der Merwe (FM)] since the surface free energies of Ni is significantly larger than the value of Al. The local acceleration near the substrate and the steering effect has not been yet substantially investigated for the various surface orientations. It is interesting to note that various intermetallic compounds are known to be thermodynamically stable between Ni and Al, and experimentally NiAl($B2$) and Ni_3Al($L1_2$) intermetallic compounds were found in the interface region of Ni/Al(001) system for the early stage of thin film.[14,15] The surface morphology would be affected with mixing characteristics at the interface according to surface orientation.

In this study, in order to investigate the growth morphology in atomic level, the deposition process of Ni atoms on Al(001), (011), and (111) substrates with various adatom incident energies was extensively investigated by using molecular dynamics (MD) simulation, which is known to be capable of simulating accurate atomic nature of interface characteristics and growth behavior.

II. CALCULATION METHOD

Interatomic potentials were based on the embedded-atom method (EAM).[16] In the present work, the EAM potential developed by Voter and Chen for Al–Al and Ni–Ni (Ref. 17) and Farkas *et al.* for Ni–Al (Ref. 18) were adapted. The cutoff distances of Ni, Al, and Ni–Al were set to 4.79, 5.55, and 5.58 Å, respectively. The XMD 2.5.32 code[19] was utilized for the molecular dynamics simulation and the employed Ni–Ni, Al–Al, and Ni–Al EAM potentials showed good agreement with the experimental values of pure elements as well as those for intermetallic compounds. The size of fcc-Al(001) substrate was set to $20a_0 \times 4a_0 \times 4a_0$, where a_0 is the bulk lattice parameter of Al, with the surface perpendicular to z axis. The periodic boundary conditions were applied in both x and y directions and the position of the bottommost two layers was fixed at 0 K. The temperature of the other layers was maintained at 80 K, a process condition normally adapted to exclude the temperature effect. The substrates of (011) and (111) orientations were similarly prepared for the simulation experiments.

The incident energy of adatoms was varied in the range of 0.1–7 eV. The range of adatom incident energy considered in this simulation study represents the working pressures practically employed in the sputtering process of thin films in deposition experiments. The adatom was added at a distance of 20 Å from the substrate surface, which was far-

a)Electronic mail: yongchae@hanyang.ac.kr

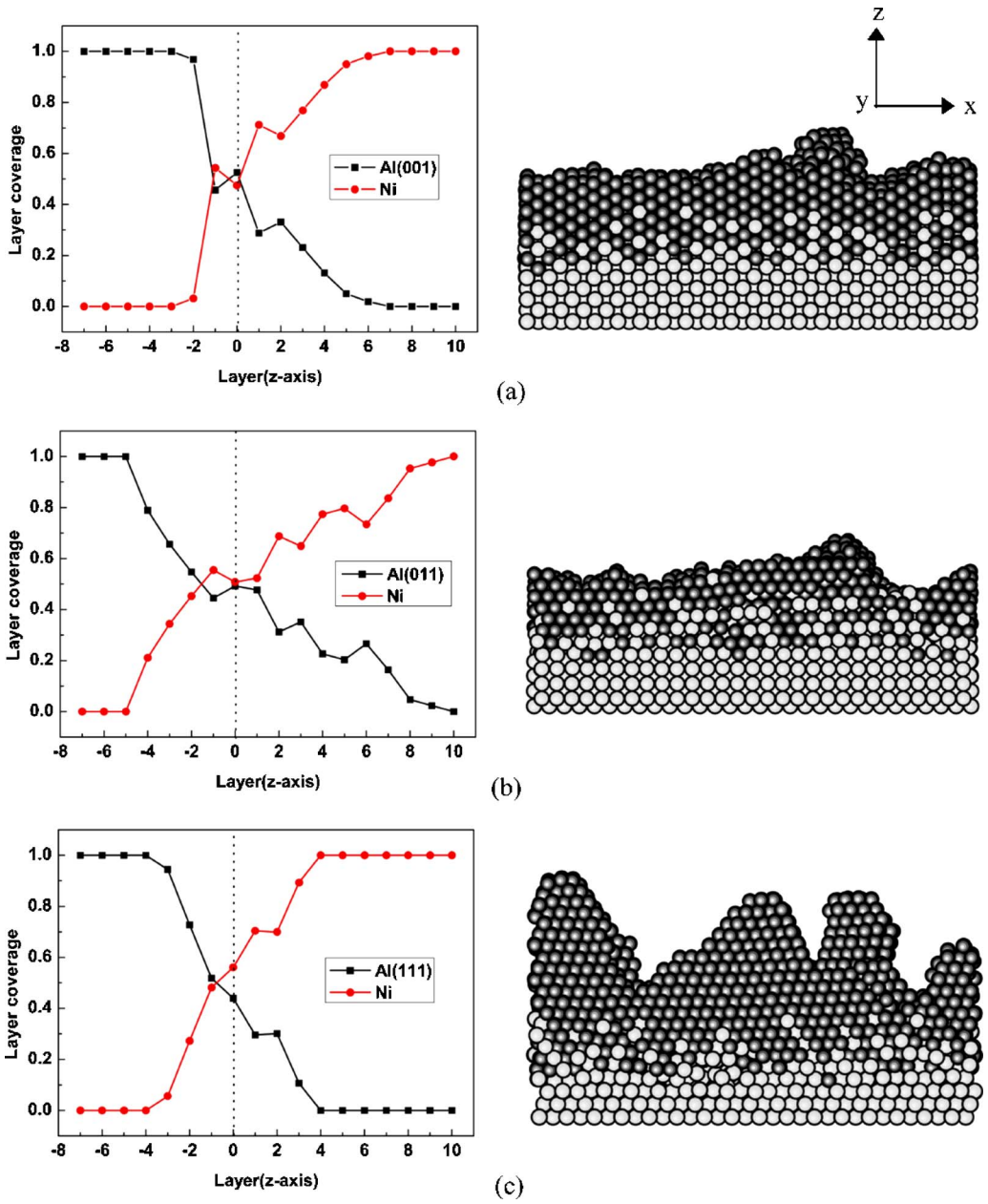

FIG. 1. Layer coverage fraction of Ni atoms with 0.1 eV on (a) Al(001), (b) Al(011), and (c) Al(111) surfaces at 80 K along the z-axis direction. Totally, 10 ML of Ni atoms were deposited on Al surface. The black circles represent the deposited Ni adatoms and the white circles indicate Al substrate atoms.

ther than the cutoff distance of corresponding simulation. The initial positions of the adatoms were randomly selected in the xy plane and incident angle was set to be normal to the surface. The MD time step was set to 1 fs and the total evolution time for the deposition process of single atom was limited to 5 ps.

III. RESULTS AND DISCUSSION

When Ni atoms are deposited on each Al substrate at 80 K up to 10 ML (monolayers) with low incident energy of 0.1 eV, active surface alloying at the interface could be observed in the early stage of thin film deposition irrespective of substrate orientation, as shown in snapshots of Fig. 1. However, the different degrees of mixing at the interface were observed according to substrate orientation. The layer coverage fraction of Al substrate atoms and deposited Ni atoms with distance along the z direction has been investigated to quantitatively analyze compositional variation in the interface region with respect to the Al substrate orientation (Fig. 1). The zeroth layer corresponds to the surface of Al substrate before deposition. For Ni films grown on Al(001), (011), and (111) substrate, 1.0, 2.0, and 1.3 ML of deposited Ni atoms were incorporated with the substrate Al atoms. The case of Ni films grown on Al(011) substrate has about twice wider mixing region at the interface in comparison with the case of Al(001) and Al(111) substrates. This result would be explained by the structural differences among the corresponding surfaces. The planar density is relatively higher for the (111) and (001) surfaces than the case of (011) surface.

In addition, the measured values of the locally accelerated energy for Ni adatom on Al(001), Al(011), and Al(111) surfaces were measured to be in the range of 2–4 eV (Fig.

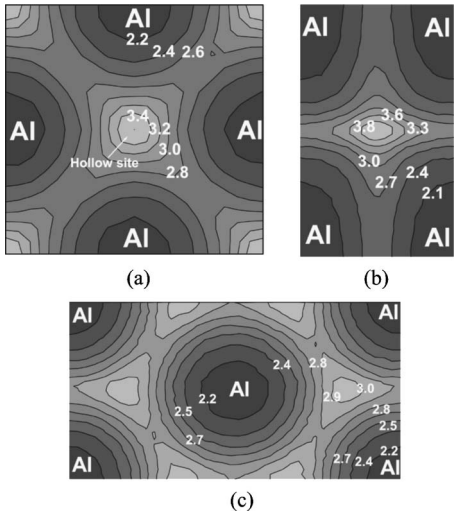

FIG. 2. The locally accelerated energy contour of Ni adatom on each Al surface for 0.1 eV incident energy (a) Al(001), (b) Al(011), and (c) Al(111).

2). In particular, the hollow site, which is the most stable position for adatom deposition, represents the highest degree of locally accelerated energy among all possible sites. The highest values of the locally accelerated energy for Al(001), Al(011), and Al(111) surface were 3.68, 4.12, and 3.15 eV, respectively. It can be reasonably expected that the locally accelerated energy on hollow site would be sufficiently high to overcome the binding energy of Al (3.36 eV), considering the decrement of the cohesive energies for the exposed Al substrate on the surface. Consequently, the Ni adatom could be mixed with the Al substrate in spite of the low adatom incident energy of 0.1 eV.

The surface roughness W^2, after 7–20 ML deposition of Ni atoms on Al substrate, was measured against substrate orientation and adatom incident energy to quantitatively analyze the influence of the variation of substrate orientation and adatom incident energy on the morphology of grown Ni films (Fig. 3). The surface roughness[20] could be calculated as follows:

$$W^2 = \sum_{j=0}^{\infty} (j - \Theta)^2 (\theta_j - \theta_{j+1}), \tag{1}$$

where Θ is the total coverage and θ_j is the coverage of layer j. The tendency of the growth behavior of Ni films on the Al substrate was turned out to be varied with substrate orientation and adatom incident energy, as illustrated in Fig. 3

For the coverage of a few monolayers, irrespective of surface orientation, low surface roughness could be observed. However, the film roughness clearly increases according to increment of Ni deposition and the highest surface roughness was appeared for Ni thin film on Al(111) substrate with low incident energy of 0.1 eV, as illustrated in Fig. 3(a). The growth behavior seemed to have changed from island growth mode to layer-by-layer growth mode for adatom incident energy of 3–7 eV, in which the upper Ni layers began to fill after piling up the lower Ni layer, consequently, resulting in the low surface roughness, as shown in Fig. 3(b).

In an attempt to quantitatively analyze the roughness characteristics, the artificially engineered four layer high

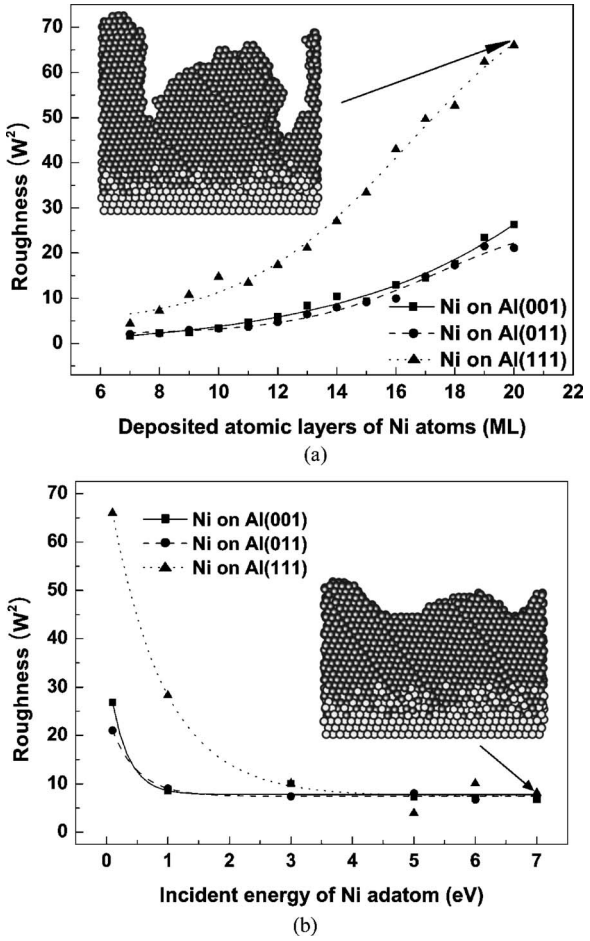

FIG. 3. Surface roughness at 80 K after deposition of Ni films on each Al surface (a) after 7 ~ 20 ML deposition with 0.1 eV incident energy, (b) after 20 ML deposition with 0.1–7 eV incident energy. The black circles represent the deposited Ni adatoms and the white circles indicate Al substrate atoms

steps of Ni atoms were situated on the deposited Ni film/Ni–Al mixing region/Al substrate and extensive trajectory calculations were performed in the direction perpendicular to the plane of incidence. The trajectories of the adatoms of the low incident energy of 0.1 eV clearly show that the steering effect, bending of trajectories of the incident atoms, occurs most notably near the step due to the interaction between the approaching adatom and surface atoms [Fig. 4(a)]. Subsequently, the two-dimensional distribution of the deposition flux at the surface normalized to the homogenous flux far above the surface was derived at a incident energy of 0.1 eV to analyze the steering effect quantitatively near Ni step on Al(001), Al(011), and Al(111) substrates [Fig. 4(b)]. In fact, all Ni adatoms whose trajectories pass through substantially distorted areas of the attractive potential near the Ni step did contribute to enhanced flux. For the case of Ni adatom on Al(111), substrate, it appears to be the existence of the largest enhancement of the deposition flux near Ni step in comparison with Al(001) and Al(011), as shown in Fig. 4(b). This result is qualitatively consistent with the roughness observation of Fig. 3(a). Consequently, it can be reasonably inferred that steering effect is a critical factor in determining the morphology of the growing films.

On the other hand, the approaching Ni adatoms with

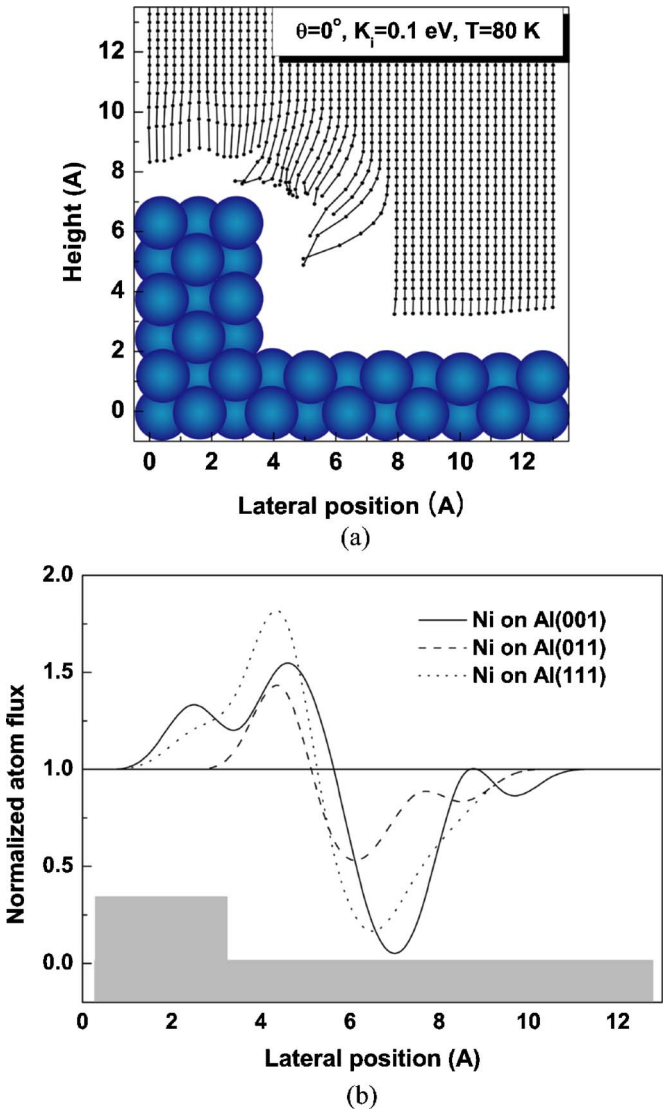

FIG. 4. (a) (projected view onto the y, z plane) trajectories for Ni adatoms with 0.1 eV near surface of Ni step formed on Al(001) surface. (b) Calculated Ni atom flux at the various surfaces, normalized to homogeneous atom flux far above the surface.

high incident energy of 3–7 eV experience much smaller steering force than the case of 0.1 eV. Consequently, the adatoms could be distributed homogeneously on the Al surface with the initial incident direction irrespective of the substrate orientation due to greatly reduced the steering effect. The measured value of the locally accelerated energy for Ni adatom on Ni surface is in the range of 2–4 eV, which is similar to the value for the locally accelerated energy for the Ni adatom on Al surface. The total kinetic energy of adatoms which can be represented as the sum of initial incident energy and locally accelerated energy was, apparently, found to be sufficient to overcome the attractive forces between the adatoms and the local geometry of the substrate near Ni island. In addition, the adatom incident energy of 3–7 eV certainly has sufficient kinetic energy to break the Ni–Ni bond (4.45 eV) of the island structure, resulting in smoother surface by funneling down the Ni atoms on the surface of Ni island after the impact between Ni adatom and the island, which could successfully explain the roughness characteristics with the incident energy of 3–7 eV in Fig. 3(b).

IV. CONCLUSION

The characteristics of Ni thin film growth on Al(001), (011), and (111) surfaces at 80 K with respect to incident energy were investigated by using molecular dynamics simulation. Active surface alloying occurred at the early stage of Ni thin film deposition on each Al substrate with 0.1 eV incident energy, and after around 7 ML deposition of Ni, only Ni film growth without alloying was observed. The surface roughness became progressively rougher according to increment of Ni deposition at low incident energy of 0.1 eV. The remarkable roughening observed for Ni adatom on Al(111) surface could be rationalized in terms of steering effect representing the enhanced flux of Ni adatom through the distorted areas of the attractive potential near the Ni island structure. For the Ni deposition of 20 ML, subsequently, the tendency of island growth markedly observed in case of Ni film grown on Al(111) surface. In spite of the locally acquired accelerated energy above 2 eV near Ni surface, Ni adatoms with initial kinetic energy of 0.1 eV turned out to be insufficient to collapse down and overcome the steering force near the Ni island structure grown on Al substrate and, consequently, followed the island growth mode. However, in the range of 3–7 eV incident energy, the surface became smoother, since the atom could use its extra energy to funnel down the corresponding atoms after the impact and also experiences much smaller steering force irrespective of Al substrate orientation. It could be verified that the adatom incident energy, which could control the degree of steering effect, is a critical factor in determining the morphology for Ni thin film growth on Al substrate.

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