A Comparative Study on the Differences in the Evolutions of Thin Film Morphologies of Co-Al Binary System: Molecular Dynamics Study

Sang-Pil Kim^{1,2}, Seung-Cheol Lee¹, Kwang-Ryeol Lee¹, Kyu-Hwan Lee¹, and Yong-Chae Chung²

¹Future Technology Research Division, Korea Institute of Science and Technology, Seoul 136-790, KOREA

²CPRC, Department of Ceramics, Hanyang University, Seoul 133-791, KOREA

ABSTRACT

The morphology evolution of thin films was studied by molecular dynamics simulation. In this simulation four deposition and substrate elements combinations were used: Al on Al(001), Al on Co(001), Co on Co(001), Co on Al(001). The Al thin film was always grown by layer-by-layer mode regardless of substrates used. On the other hand, thin films formed by Co deposition depended on substrates used. While Co thin films on the Co substrates were grown by the island mode, a 3 monolayer (ML) thickness of CoAl surface compound was initially formed on Al substrate, before pure Co thin film growth occurred. In addition to the study on morphologies, the degrees of mixing of atoms in the interface were studied quantitatively. No surface mixing and a sharp interface were observed when Co was used as a substrate regardless of deposited atoms. On the contrary, a large amount of surface mixing or compound formation was observed when Al was used as a substrate.

INTRODUCTION

Recently, devices fabricated by magnetic ultra-thin-film multilayers have attracted great attention due to their unique properties such as GMR (Giant Magneto-Resistance) [1,2] or TMR (Tunneling Magneto-Resistance) [3-5] phenomena. Magnetic random access memory (MRAM) [6] that utilizes the spin dependent tunneling of electrons between two ferromagnets through the insulating barrier, is expected to replace the conventional Si-based random access memory. Since, the spin dependent tunneling of the device is highly sensitive to the interface structures [7], understanding the interface evolution is of decisive importance to improve the device performances.

In conventional thin film growth models, substrates have been treated as an inert template

[8] on which atoms are deposited, diffused, and nucleated. Recent studies, however, have shown that substrates are no longer an inert medium and monolayer-confined surface alloying between the substrate and deposited atoms is commonly observed [9-11]. Considering the thickness of the insulating film used for MRAM device approaches to the nanometer scale (corresponding to 4 ~ 5 monolayers), this means that about a half of atoms in insulating thin films belongs to the interface region.

This study was focused on the morphology evolution of thin films for the cases of Co or Al atoms, using molecular dynamics simulations. Cobalt is used for a ferromagnet TMR junction and aluminum is used for making an insulating barrier after oxidation. To understand the effects of a substrate and an adatom interaction, various combinations of the Co and Al substrate was used.

COMPUTATIONAL METHOD

In many metallic systems, interatomic potentials based on the embedded atom method (EAM) [12] are in good agreement with experimental results. In the present work, we used the EAM potential developed by Pasianot and Savino [13] for Co-Co, Voter and Chen potential [14] for Al-Al, and Vailhé and Farkas [15] for Co-Al. The cut-off distance for Co, Al, and Co-Al was set to 5.26 Å, 5.55 Å, and 5.6 Å, respectively. XMD 2.5.30 [16] was used for the simulation.

In this simulation, four deposition conditions were studied: Al on Al(001), Co on Al(001), Co on Co(001), and Al on Co(001), respectively. The slab model was adapted to simulate the surface. The fcc-Al and fcc-Co (001) slab was set to $6a_0 \times 6a_0 \times 4a_0$, where a_0 is the bulk lattice parameter of Al and Co, respectively, with the surface perpendicular to z-axis. Periodic boundary conditions were applied in both x and y directions. Both the Al and Co slabs have three active layers and one fixed layer along the z-axis at the start of the simulation.

The substrate temperature was set to 300 K. Before depositing thin film atoms, substrates were equilibrated for 3 ps with a time step (Δt) of 0.2 fs. After the equilibration, Δt was reduced to 0.1 fs to obtain more accurate results. The kinetic energy of incident atoms is 0.1 eV. The positions of the deposited atom were randomly selected in the x-y plane. The vertical position of the deposited atoms was 16.8 Å from the substrate surface, which was farther than the cut off distances. The atoms were incident normal to the surface. Total evolution time for single atom deposition was set to 5 ps.

RESULTS AND DISCUSSION

Morphology evolutions by atom deposition

Figure 1(a), (b), (c), and (d) show the morphologies of thin films after 3 ML of atoms are deposited: 1(a) for Al on Al(001), 1(b) for Al on Co(001), 1(c) for Co on Co(001) and 1(d) for Co on Al(001), respectively. When the deposited elements are the same as the substrate elements (figures 1(a) and 1(c)), the black and gray spheres were used to merely distinguish the deposited film and the substrate. On the other hand, the black and gray spheres in figures 1(b) and 1(d) were used to distinguish the Co and Al, respectively. In the cases of Al deposition on Al and Co substrates, which are shown in figures 1(a) and 1(b), the layer-by-layer growth of an Al thin film was observed in both substrates. The layer-by-layer growth of an Al deposited film on Al and Co substrates could be explained the surface energies of the substrate, the deposited film, and the interface [8].

When Co atoms are deposited on Al and Co substrates, morphologies of the thin films are different from the case of Al deposition. In homogeneous Co deposition (figure 1(c)), the Co film exhibits an island growth mode, which should be grown by the layer-by-layer. The island growth of the Co film on the Co surface might result from the ‘steering effect’ [17], which bends the trajectory of the deposited atom to a Co island formed on surface, due to the strong attractive forces between the deposited atoms and the island.

Figure 1. Morphology evolution of various adatom/substrate combinations after 3 ML of atoms were deposited. (a) Al/Al (001), (b) Al/Co (001), (c) Co/Co (001), and (d) Co/Al (001)

In the case of Co deposited on Al (001), the morphology of thin film drastically changed. Most of the deposited Co atoms mixed with the Al substrate atoms. The structure of the mixed phase was the “B2” structure of CoAl, as determined by radial distribution function (RDF) analysis [18]. This is the most stable structure in the Co-Al binary system. Considering the low incident energy (0.1 eV) and the low temperature of the substrate (300 K), the formation of the B2 structure is quite surprising.

Mixing Behavior

To analyze the mixing quantitatively, the normalized number densities of the deposited atoms and the substrate atoms perpendicular to the surface normal were plotted in figure 2(a), (b), (c), and (d), respectively.

Figure 2. Normalized atom number densities of deposited atoms and substrate atoms plotted as a function of position normal to the specimen surface. Solid lines indicate the number density for the deposited atoms and dashed lines corresponds to the substrate atoms. The shaded area indicates the position of the surface before deposition. (a) Al/Al (001), (b) Al/Co (001), (c) Co/Co (001), and (d) Co/Al (001).

The dotted and dashed lines indicate the normalized density of the substrate and deposited atoms, respectively. The shaded areas correspond to the surfaces before deposition. In the case of Al on an Al surface (figure 2(a)), 14 % of substrate atoms were exchanged with the deposited Al atoms. Snapshot analyses showed that the mixing of Al on an Al substrate mostly occurred at the moment of deposition, by the exchange mechanism [19]. In cases of Al on Co(001) and Co on Co(001), which are shown in figure 2(b) and 2(c) respectively, interlayer mixing was not observed and sharp interfaces evolved on the surface. Small differences in the number densities of deposited atom over the surface in figure 2(b) and 2(c) are due the fact that the thin films are grown by different modes: layer-by-layer growth for figure 2(b) and island mode for figure 2(c). The layer number density of Co on an Al surface (figure 2(d)) is substantially different from the above mentioned cases. A zigzag pattern of the number densities of deposited and substrate atoms was observed. This is caused by the formation of B2 structure of CoAl on the Al(001) surface. The zigzag pattern spans up to 3 layers, which agrees well with the experimental results [20].

CONCLUSION

Thin film morphology evolution was studied using molecular dynamics simulations. In these simulations, four combinations of substrates and deposited atoms were used: Al on Al(001), Al on Co(001), Co on Co(001), and Co on Al(001). The layer-by-layer growth of Al thin films could be observed regardless of the substrate elements chosen. Thin film morphologies of Co deposited films were largely different from Al thin films. While Co thin films were grown by the island mode on Co(001) substrate, the B2 structure of CoAl was first formed on the Al(001) surface, before the Co phase was stabilized in the thin films. When Co or Al atoms are deposited on a Co surface, no interlayer mixing was observed. Substantial mixing or the formation of a surface compound was found when Al or Co atoms deposited on an Al (?) surface, respectively. Considering that only two elements were used in this simulation, it is interesting that three different morphologies could be evolved. The origin of the differences in thin film morphologies should be studied more detail in near future.

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