

Atomic Scale Understanding of Intermixing Behavior of Thin Metal Multilayer

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ABSTRACT

We have investigated the atomic scale intermixing behavior of Co-Al system in both theoretical and experimental methods. Asymmetric intermixing was observed by molecular dynamics simulation: severe intermixing occurs when Co was deposited on Al surface resulting in 3 atomic layers of CoAl-B2 compound. In contrast, an atomically sharp interface forms between Al film and Co substrate. The predicted asymmetry in atomic scale intermixing was experimentally confirmed by a coaxial impact collision ion scattering spectroscopy (CAICISS). The adsorbed Al atoms on Co (0001) surface were placed on either fcc or hcp hollow sites with approximately the same probability. On the other hand, the adsorbed 0.5 monolayer (ML) Co ad-atoms were mixed with Al atoms of substrate resulting in the thin surface layer of ordered CoAl-B2 phase. Magneto-optical Kerr effect (MOKE) measurement of Co/Al multilayer also agrees very well with the simulation results in a quantitative manner.

Keywords: atomic scale intermixing, metal multilayer, provide up to five comma separated, keywords for indexing, don't capitalize

1 INTRODUCTION

Thin multilayer structures are widely used in current devices and sensors, such as spintronics devices utilizing the tunneling magneto resistance (TMR) or the giant magneto resistance (GMR) phenomena [1-6]. For these devices, the electro-magnetic properties are largely dependent on the interface structure between thin ferromagnetic and nonmagnetic layers [7]. Because of the short coherence length of spin polarization, electric spin phenomena can only occur across a few nm thick ordered thin films. It must be also noted that spin injection from ferromagnetic electrode to semiconductor channel is known to be strongly dependent on the interfacial structure [8,9]. Atomic scale control of the interface is thus the prerequisite for these devices, which requires well understanding of the surface intermixing behavior during deposition in both theoretical and experimental ways.

In the present work, we investigated the intermixing behavior of Co-Al system by a molecular dynamic simulation using semi-empirical EAM potentials. This

nonmagnetic/ferromagnetic system has drawn much attention in developing tunneling magnetoresistance (TMR) or magnetic random access memory (MRAM) devices [9]. Asymmetric intermixing in the Co/Al interface was observed. The interesting behavior was definitely confirmed by a coaxial impact collision ion scattering spectroscopy (CAISS) when 0.5ML of atoms were deposited. Magnetic properties of Co/Al multilayer using MOKE measurement

2 METHODS

2.1 Molecular Dynamics Simulation

We used the EAM potential developed by Pasianot and Savino for Co-Co [10], Voter and Chen potential for Al-Al [11], and Vailhe' and Farkas for Co-Al [12]. The Al (001) or Co (0001) slab were set to $6a_0 \times 6a_0 \times 4a_0$, where a_0 is the bulk lattice parameter of substrate. Periodic boundary conditions were applied in lateral directions. The Al slab has three active layers and one fixed layer at the start of the simulation along the z axis. The substrate temperature was set to 300 K. Before deposition, the substrate was equilibrated for 3 ps with time step of 0.2 fs. After the equilibration, the time step was reduced to 0.1 fs to obtain more accurate results. The incident atoms had a kinetic energy of 0.1 eV. The positions of the deposited atom were random on the surface. The deposit atoms were added at the distance of 16.8 Å from the substrate surface, which was farther than the cutoff distances. Incident angle was normal to the surface. Total evolution time for single atom deposition was set to 5 ps.

2.2 CAISS and MOKE Measurement

Deposition and CAICISS experiments were performed at room temperature in an ultra-high vacuum chamber of base pressure less than 1×10^{-10} Torr. One side mirror-polished Co (0001) and Al (001) single crystals were used as the substrate. Prior to the deposition, the surface residues were removed by sputtering with 2.0 keV Ar⁺ ions for several tens of minutes. The specimens were then annealed at 650K for 1-2 hours to relax the surface structure. A half monolayer of Al (3.4×10^{14} cm⁻²) was deposited by evaporation using a heated tungsten filament. For CAICISS

measurement, 3 KeV He⁺ ions of beam current 10 pA were irradiated on the surface at a specific angle. Yields of He⁺ ions scattered by the substrate Co were measured with polar scan.

MOKE measurement of Co/Al multilayers was carried out at room temperature. The Co/Al multilayers were prepared by well controlled electron beam evaporation system. The prepared samples are Co layer on Si (Co(30 Å)/Si), Al layer deposited on the Co layer (Al(30 Å)/Co(30 Å)/Si) and Co layer on thick Al layer (Co(30 Å)/Al(840 Å)/Si). To protect contamination and prevent oxidation, all samples are capped by Cu film.

3 RESULTS AND DISCUSSION

Figure 1 shows the simulated film morphologies. When Al atoms were deposited on Co (0001) surface, atomically sharp interface forms between the film and the substrate (see Fig. 1 (a)). On the other hand, severe intermixing was observed when Co atoms were deposited on Al surface as shown in Fig. 1 (b). Radial distribution function of the intermixing region is compared with those of CoAl-B2 phase and Al fcc phase in Fig. 2. It is evident that CoAl-B2 compound layer was formed at the interface between Co and Al. Thickness of the compound layer was 3MLs or 1 nm as confirmed by the composition analysis. This asymmetric intermixing was discussed in terms of the increase in the kinetic energy of the deposited atom near the substrate surface and the activation barrier for the atomic intermixing [13-14]. We also suggested a novel process for high performance spin valve devices based on this asymmetry [15].

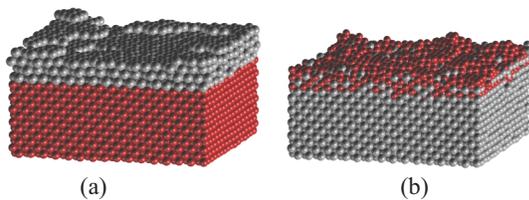


Figure 1: Film Morphologies after 3 monolayers deposition (a) Al on Co (0001) (b) Co on Al (001)

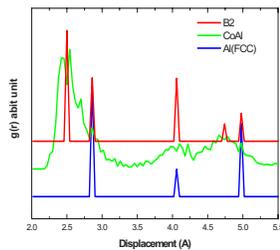


Figure 2: Radial distribution functions of intermixing layer (green), CoAl-B2 and Al fcc phases.

Spin resolved local density of state of the CoAl-B2 phase was calculated by the first principles calculation of spin resolved local density of state (Fig. 3). Symmetric band structure of up-spin and down-spin shows that the B2 structure is non-magnetic. Therefore, the intermixing of Co on Al substrate will degrade the magnetic property of the Co thin film.

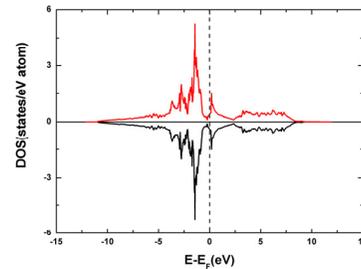


Figure 3: Spin resolved local density of state of CoAl-B2 structure obtained by the first principle calculation.

Figure 4 shows the polar scan spectrum of 3KeV He⁺ particles scattered by substrate Co atoms along the [1 $\bar{1}$ 00] direction when Al was deposited on the Co substrate. The spectrum is almost identical to that of pure (0001) Co surface, except the peak at the incidence angle of about 37°. The observed spectrum agrees very well with the calculated one based on the assumption that the deposited Al atoms are placed at the distance of 1.8 Å from the top Co layer on either fcc or hcp site with the same probability. This result clearly shows that no intermixing occurs between the deposited Al atom and the Co substrate. First principle calculations of the adsorption energy of Al ad-atoms on Co (0001) surface support the experimental observations. The calculated adsorption energy of hcp and fcc hollow site are 3.557 and 3.548 eV, respectively. Al atoms can thus occupy both hcp and fcc hollow sites at room temperature with approximately the same probability. Equilibrium height of the Al ad-atom from the surface Co layer was calculated to be 1.87±0.01 Å, which is comparable to the observed value, 1.8 Å.

Figure 5 shows the polar scan spectrum scattered by substrate Al atoms along [001] direction when 0.5 monolayer of Co was deposited on Al (001) surface. By the Co deposition, the spectrum was significantly changed from that of pure Al (001) surface. However, the calculated spectrum of CoAl-B2 structure fits very well with the observed spectrum, which shows that the deposited Co atom significantly induces the CoAl-B2 layer.

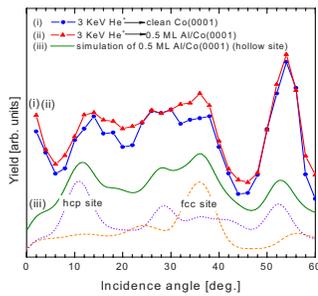


Figure 4: Polar scan curves for clean Co (0001) surface, 0.5ML Al/Co (0001) and calculated curve for 0.5ML Al/Co (0001) without mixing.

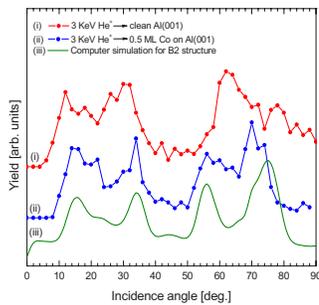


Figure 5. Polar scan curves for clean Al (001) surface, 0.5ML Co/Al (001) and calculated curve for CoAl-B2 structure

Figure 6 shows the hysteresis curves of MOKE measurement of Co/Al multilayers. The shape and width of the hysteresis loops of Co(30 Å)/Si and Al(30 Å)/Co(30 Å)/Si are identical. However, the width of hysteresis loop of Co(30 Å)/Al(840 Å)/Si is much narrower than that of Co(30 Å)/Si sample. These behaviors can be explained by the formation of nonmagnetic CoAl-B2 interlayer resulting in reduced thickness of the magnetic Co layer. We also observed the MOKE hysteresis loops for various thicknesses of Co thin film on Al(840 Å)/Si substrate. Hysteresis loop cannot be observed when the thickness of Co was smaller than 10 Å. It can thus be said that the thickness of the nonmagnetic CoAl-B2 layer is 10 Å or 3 atomic layers, which agrees well with the MD simulation results.

4 CONCLUSIONS

The most significant result of the present work is that the asymmetry in atomic scale intermixing is confirmed by

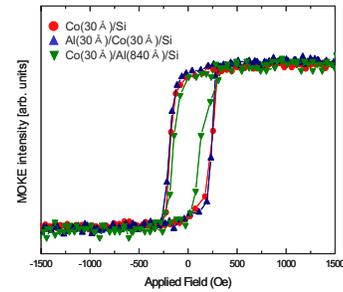


Figure 6: MOKE hysteresis loops of Co-Al multilayers

experimental methods. Molecular dynamics simulation shows that Co atoms deposited on an Al substrate form a 3 ML thick surface layer of B2 structure even at room temperature. On the other hand, Al deposition on Co surface shows a sharp interface between the deposited layer and the substrate. CAISS analysis of the surface and MOKE hysteresis measurement agree very well with the predictions made by the molecular dynamics simulation. The driving force for the intermixing would originate from the stability of the B2 phase in the Co-Al system. However, the mechanism for the ease of the surface alloying that occurs during a very short period at room temperature is yet to be clarified. Thermodynamic analysis is insufficient to understand the asymmetric mixing behavior. Kinetic analysis of the deposition behavior is in progress.

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