Ab Initio Molecular Dynamics Simulation on Structure and Strength of Si/Al Interface

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ABSTRACT

It is of great importance to clarify the atomic structure and strength of interfaces between metal and Si substrate in an electronic device. While interaction between atoms near the interface is complicated, *ab initio* molecular dynamics makes it possible to analyze such properties of interface. In this study, in order to evaluate structure and strength of Si/Al interface, evaporation process of Al atoms onto Si substrate and its tensile deformation are examined by *ab initio* molecular dynamics simulation. Al atoms are precipitated from above the Si-(001) surface. The adhesive strength of the Al atom in the 1st layer on the surface is evaluated as 1.3nN by the tensile simulation. The 2nd layer of Al atoms are formed about 0.1nm above the 1st layer. The interval between the layers are about half of that between (111) planes in the Al single crystal lattice.

Keywords: *Ab initio*, Interface, Structure, Mechanical strength

1 INTRODUCTION

In recent years, manufacturing of devices with nanostructure has been attempted with the advent of the nano-processing technique. Since such devices with scale of nanometers have fine and complex structure composed of different materials, characteristic mechanical property of interfaces has a great influence on the function of the devices. In order to understand the mechanical property of interfaces, it is essential to clarify their atomic and electronic structure. Because of large stress induced by the mismatch between different materials, it is necessary to evaluate precisely the interface.

In this study, molecular dynamics simulation of evaporation of Al atoms onto Si substrate based on *ab initio* method with plane wave basis set is conducted to examine the structure of Al/Si interface and its adhesive strength.

2 SIMULATION PROCEDURE

Figure 1 shows the simulation cell used in this calculation and the initial configuration of Si atoms. Si atoms are arranged in the middle of the cell so that the z axis corresponds to (001). The periodic boundary condition is applied to all directions. Since the region at the top and the bottom is vacuum, the cell includes (001) surfaces. The size of the cell is $L_x \times L_y \times L_z = 4a/\sqrt{2} \times 4a/\sqrt{2} \times 3a$ (=1.54×1.54×1.63nm), where the lattice constant, a, is 0.543nm.

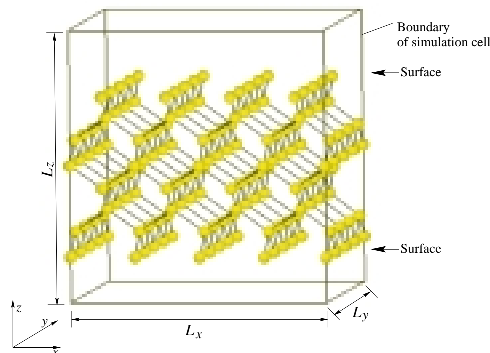

Figure 1: Simulation cell.

The structure of the surfaces is relaxed by molecular dynamics (MD) simulation. First, the surface structure is roughly relaxed using Tersoff potential function[1] for 10000 MD steps, where time interval, Δt, is 1fs. During the MD calculation, temperature is controlled to be 300K by velocity scaling.

Second, the relaxation is continued using *ab initio* MD calculation based on the norm-conserving pseudopotential plane wave method[2]. GGA (Generalized Gradient Approximation) method by Perdew *et al*[3] is adopted for evaluation of exchange-correlation energy. Because of the large cell size, summations over the Brillouin zone were approximated by the Γ point. The TM type pseudopotential[4] is used and the kinetic energy cut-off is 8.0Ry. The wave functions are converged by RMM-DIIS algorithm[5] under parallel computing. The structure is relaxed for 300 steps by MD where the time step is 10a.u.(=0.24fs). The temperature of atomic system is controlled to be 100K and kinetic energy is removed every 10 steps.

Al atoms are precipitated onto the Si surface structure. An Al atom is arranged above the surface ($L_x/2$, $L_y/2$, $0.9L_z$) and the structure is relaxed by 1000 MD

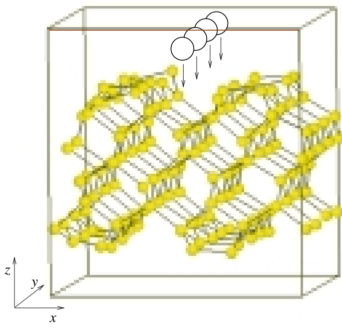

Figure 2: Process of precipitation of 1st layer Al.

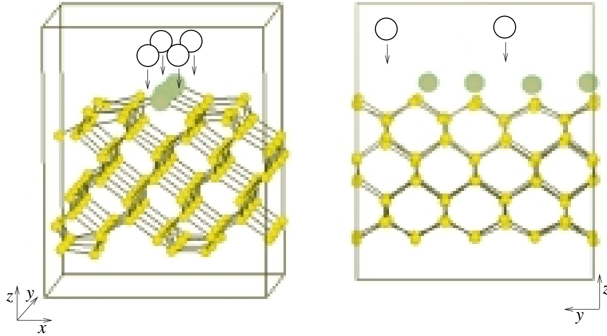

Figure 3: Process of precipitation of 2nd layer Al.

steps, while the site of all the atoms except the added Al is fixed. Then, another Al atom is added and the configuration is relaxed in the same way except that the position of newly added Al is about 7a.u.(0.37nm) away from the others in the y direction (see Fig.2). After 4 Al atoms are placed on the Si surface, the whole configuration of the atoms is relaxed (3000 MD steps at 100K $\rightarrow$ 1000 steps at 10K (in both, kinetic energy is removed every 10steps)). These Al atoms construct "the 1st layer" on the surface.

From this state, adhesive strength of the Al atom on the Si surface is evaluated by tensile simulation. In this process, atoms at the lower surface of Si is fixed and one Al atom in the 1st layer is pulled up in the direction perpendicular to the surface. After displacement of 1.1a.u.(0.059nm) in the z direction is imposed to one Al atom, atom configuration is relaxed. This procedure is repeated until all bonds to the Al atom are broken.

From the structure after the precipitation of 4 Al atoms on the surface, the size of the cell in the z direction is stretched by 1.1 times ($L_z = 1.79$nm) and 4 more Al atoms are precipitated onto the surface structure constructed above in the same manner (see Fig.3). These Al atoms construct "the 2nd layer".

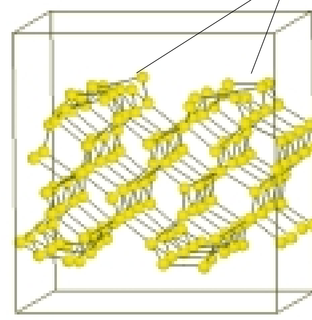

Figure 4: Atom configuration of Si (100) surface after relaxation.

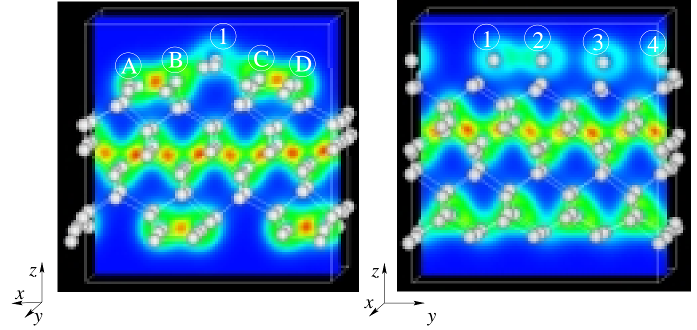

Figure 5: Snapshots after precipitation of 4 Al atoms.

3 RESULT AND DISCUSSION

3.1 Al atoms in the 1st layer

Figure 4 shows atom configuration of the Si-(001) surface after relaxation. Dimers are formed in 2 rows, which has a ditch between them.

Figure 5 shows structure after 4 Al atoms (the 1st layer) are precipitated on the Si surface (before relaxation). Spheres indicate atom position in the figure. Distribution of charge density on sections is also drawn to depict bond structure between the atoms. The Al atoms are placed with almost the same interval distance on the ditch between the dimers of the Si surface. The structure after relaxation is shown in Fig.6. By this relaxation, ①-② and ③-④ become closer to each other. A bond between Al atoms (①-② or ③-④) is formed in each pair, and bonds between Al(①) and Si atoms(Ⓑ and Ⓒ) are also formed. Because charge density which constructs the bond is localized, it can be said that the bond structure is similar to that of covalent ones. Figure 7 illustrates the formation of the bond structure schematically.

3.2 Adhesive strength of Al atom on Si surface

Snapshots during the tensile simulation is shown in Fig.8. It clarifies the change in bond structure. Bonds of Ⓑ'-③, Ⓒ'-③ and ③-④ are cut as the atom③ is pulled

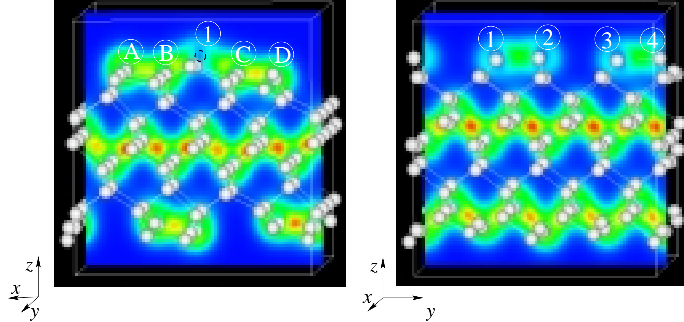

Figure 6: Snapshots of relaxed configuration after precipitation of 4 Al atoms.

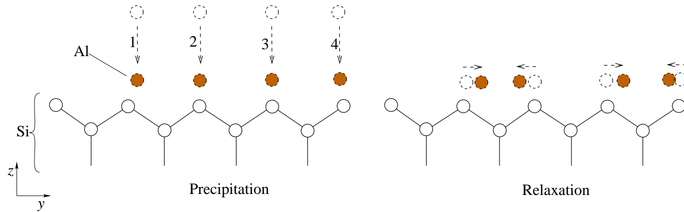

Figure 7: Schematic illustration of motion of Al atoms.

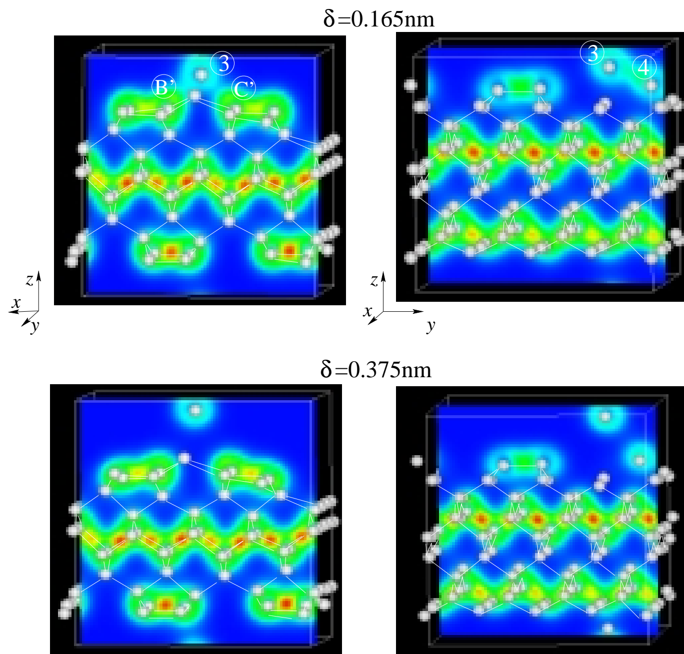

Figure 8: Snapshots during tensile simulation.

upwards, and only the Al③ is detached from the surface. Figure 9 shows displacement of the atoms during the tensile deformation. In the figure, the displacement of the atoms are indicated by vectors, where the length is multiplied by 5 times. Motion of the Si atoms(Ⓑ and Ⓒ) and the Al atom(②) which are bonded with Al① pulled is conspicuous. Range in which atoms affected by the tension is within 3∼4 layers from the surface.

Figure 10 shows change in tensile load, P, acting on the Al atom pulled. The transverse axis indicates the displacement of the Al atom, δ, from the position after

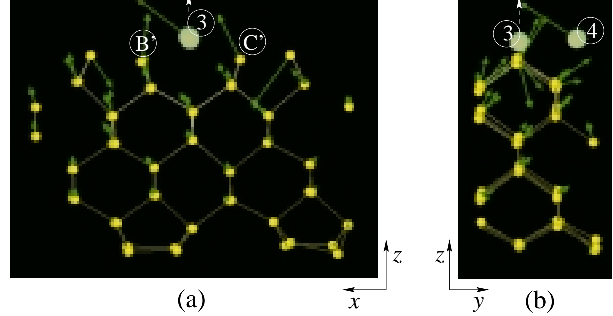

Figure 9: Displacement of atoms during tension.

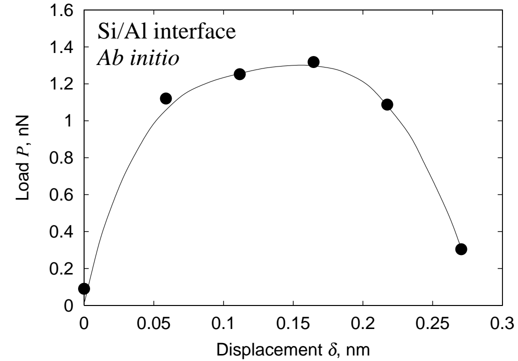

Figure 10: Relationship between load and displacement.

the relaxation of the 1st layer. With an increase of the displacement the load increases largely until reaching 0.05nm. Then, the load shows little increase and reaches a maximum value of 1.3nN at $\delta \simeq$0.17nm, before it drops sharply after δ excesses 0.2nm. That is, the adhesive strength of Al atom on Si surface is evaluated as 1.3nN, which is higher than the tensile strength of atomic chain of Al, 1.1nN, estimated by *ab initio* analysis[6].

3.3 Al atoms in the 2nd layer

Figure 11 is snapshots during the precipitation of Al atom ⑤. From the initial position, Al⑤ descends straightly onto the "mound" of Si-Al-Si structure on the surface, and then falls down on the mound (see Fig.12). Figure 13 shows the final configuration after the precipitation of Al atoms ⑥∼⑧ and the relaxation. After the precipitation and the relaxation, those Al atoms are placed above the mound of Si-Al-Si structure (the 2nd layer of Al). Distance between the 1st layer and the 2nd one is about 0.1nm, which is about half of distance between (111) planes of single crystal in aluminum (0.2nm). This implies that the atomic structure of Al up to 2 layers on the Si surface is far from that in bulk.

From charge distribution in the final state (Fig.13), bond structure of the Al atoms on the Si surface can be evaluated. Al atoms in the 2nd layer have bonds with Si ones on the surface as well as Al ones in the 1st layer. Charge density constructing those bonds is slightly spread and is not localized, which means the

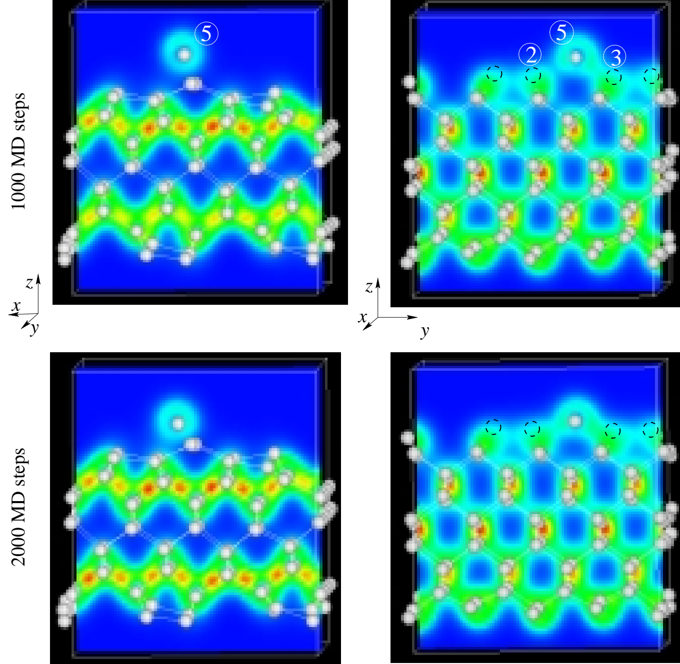

Figure 11: Snapshots of precipitation of Al atom.

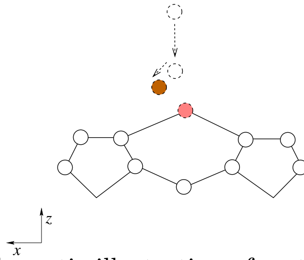

Figure 12: Schematic illustration of motion of Al atom.

bond property is similar to that of metallic one. Bond length of the Al-Al and Si-Al is about 0.27nm, which is almost the same as that in Al single crystal (0.28nm). This implies that the bond structure is similar to that in bulk.

4 CONCLUSION

As the first step for clarifying the structure of interface between different materials, *ab initio* molecular dynamics simulation on evaporation process of Al atoms onto Si substrate is conducted. The results are summarized as follows:

(1) Structure of Si-(001) surface is reproduced. Dimers are constructed in rows and between them ditch-like structure is formed.

(2) Al atoms precipitated from above the surface is relaxed on the ditch of Si surface in a row (the 1st layer of Al). Pairs of Al atoms are formed and each pair is connected by a covalent-like bond. Bonds between Al and Si are also formed and the structure of Si-Al-Si has a shape of a mound.

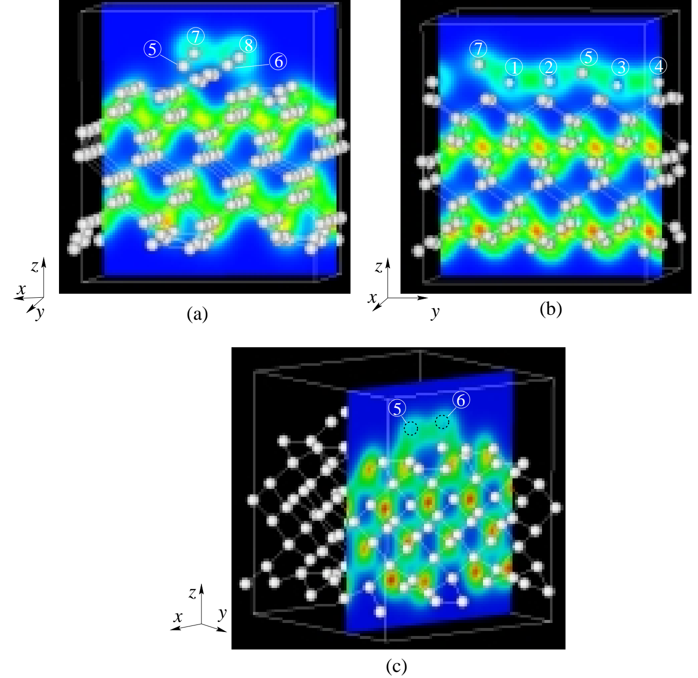

Figure 13: Final configuration of atoms and charge distribution.

(3) Adhesive strength of the Al on the surface is evaluated as 1.3nN by the tensile simulation. Range in which atoms are affected by the tension is within 3∼4 layers from the surface.

(4) More Al atoms are precipitated from above the mound. They are relaxed above the mound structure (the 2nd layer of Al). Distance between the 1st and the 2nd layers is about 0.1nm, which is about half of that in Al single crystal. This indicates the atomic structure is far from that in bulk.

(5) The Al atoms in the 2nd layer are bonded with Si ones on the surface as well as Al ones in the 1st layer. Charge constructing the bonds is not localized and their length is about 0.27nm, which is almost the same as that in Al single crystal. This implies that the bond structure is similar to that in bulk.

REFERENCES

[1] Tersoff,J., *Phys. Rev.*, B, **37**(1988), 6991–7000.
[2] Car,R. and Parrinello,M., *Phys. Rev. Lett.*, **55**(1985), 2471–2474.
[3] Perdew,J.P., *Phys. Rev. Lett.*, **55**(1985), 1665–1668.
[4] Troullier,N. and Martins,J.L., *Phys. Rev.*, B, **43**(1991), 1993–2006.
[5] Kresse,G. and Furthmüller,J., *Phys. Rev.*, B, **54**(1996), 11169–11186.
[6] Kitamura,T., Umeno,Y. and Nishida,M., *Transactions of JSME* (*in Japanese*), A, **67**(2001), 203–208.