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Atomistic Crystal Growth Process of Metal Oxide Electronics Materials: Theoretical Simulation Studies

Momoji Kubo, Toshiyuki Yokosuka, Hitoshi Kurokawa, Ken Suzuki, Seiichi Takami, Akira Miyamoto, Masashi Kawasaki[1], Mamoru Yoshimoto[2], and Hideomi Koinuma[3]

Department of Materials Chemistry, Graduate School of Engineering, Tohoku University, Aoba-yama 07, Sendai 980-8579, Japan.
Phone: +81-22-217-7233, Fax: +81-22-217-7235, E-mail: momoji@aki.che.tohoku.ac.jp

[1]Institute for Materials Research, Tohoku University, 2-2-1 Katahira, Aoba-ku, Sendai 980-8577, Japan.

[2]Materials and Structures Laboratory, Tokyo Institute of Technology, Midori-ku, Yokohama 226-8503, Japan.

[3]CREST-JST and Materials and Structures Laboratory, Tokyo Institute of Technology, Midori-ku, Yokohama 226-8503, Japan.

1. Introduction

Artificial construction of atomically defined metal oxide layers is important in making electronics devices including high temperature superconducting oxide films, magnetic and optical devices. Hence, the atomistic understanding of the epitaxial growth process of metal oxide surfaces is desired to fabricate atomically controlled structure that exhibits unexplored and interesting physical properties.

MgO thin film fabrication has attracted much attention, due to its significance as high temperature material, wide-gap insulator, unreactive insulator, secondary electron emission material etc. Hence, the epitaxial growth of MgO thin films on various substrates, such as MgO, silicon, quartz, sapphire, GaAs, TiO_2, etc., has been well investigated by using various experimental techniques. Although important information has been obtained from the epitaxial growth experiments, theoretical approaches such as molecular dynamics (MD) and quantum chemistry would be also desirable for the atomistic understanding of the epitaxial growth process of MgO thin films. However, no simulation studies were so far devoted to the epitaxial growth process of metal oxide surfaces except our previous works [1-6]. Hence, in the present study we applied our crystal growth MD simulation code MOMODY to investigate the epitaxial growth process of MgO thin films on various metal oxide substrates.

2. Method

The methodology of the crystal growth MD is slightly different from regular MD simulations. The total number of species in the system is not fixed but increases with time. The number of MgO molecules deposited over the substrate surface is increased one by one. The MgO molecules are shot to the surface at regular time intervals of 1,000 time steps, with a constant velocity of 900 m/s. The Verlet algorithm was used for the calculation of atomic motions, while the Ewald method was applied for the calculation of electrostatic interactions. Temperature was controlled by means of scaling the atom velocities under periodic boundary conditions. The calculations were performed for 50,000-150,000 steps with a time step of 2.0×10^{-15} seconds.

3. Results and Discussion

We applied our crystal growth simulation code MOMODY to the investigation of the homoepitaxial growth process of MgO(001) surface. Totally 32 MgO molecules are deposited on the MgO(001) surface, this amount is equal to the number of Mg and O atoms which constitute a single MgO layer of the MD unit cell. The simulation was performed at 300 K. After any MgO molecule migrated on the MgO(001) surface, the 2-dimensional and epitaxial growth of the MgO thin layer was observed at 300 K. Moreover, the deposited MgO molecules kept the NaCl type structure and the (001) oriented configuration. However, after all the 32 MgO molecules were deposited on the MgO(001) surface, some defects in the first constructed MgO layer were observed, and some MgO molecules already started to form a second layer. Thus, the layer-by-layer homoepitaxial growth of MgO thin film without defects was not realized at low temperature of 300 K.

Experimentally, the substrate temperature is one of the important parameters for the construction of well-defined MgO thin films.

Therefore, the understanding of the effect of substrate temperature on the homoepitaxial growth process is essential to optimize the fabrication conditions of atomically controlled MgO thin films. Hence, we simulated the homoepitaxial growth process of the MgO(001) surface at 1000 K and the result is shown in Fig. 1. Even at this high temperature, the MgO grows epitaxially keeping NaCl type structure and (001) oriented configuration. Moreover, the formation of a single 2-dimensional uniform and flat layer of MgO without any defects was observed at 50,000 time step, which is significantly different from the result at 300 K. Thus, high temperature was found to be favorable for a complete layer-by-layer homoepitaxial growth.

Experimentally, the effect of substrate on the structure of constructed MgO thin films is an interesting topic. Therefore, we also simulated the epitaxial growth of MgO thin films on α-Al_2O_3(0001) surface, and the result is shown in Fig. 2. MgO thin film epitaxially grows, keeping (111) oriented configuration during the crystal growth process. Finally, we observed the formation of a MgO quantum dot on α-Al_2O_3(0001) surface, which is significantly different from the results on MgO(001) surface. Moreover, the surface of the MgO quantum dot mainly constitutes of the (001) and (011) planes. It indicates that the formation of the quantum dot is due to the instability of the MgO(111) surface compared to the MgO(001) and MgO(011) surfaces.

Moreover, it is interesting to compare the lattice constants of the MgO(111) and α-Al_2O_3(0001) surfaces. The lattice mismatch is approximately 8.4 %. Experimentally, it is well known that the epitaxial growth takes place when the lattice mismatch of the substrate and thin film is less than 5 %. Hence, we concluded that the epitaxial growth of the MgO thin film on α-Al_2O_3(0001) surface is unusual phenomenon. It is designated as a high-order epitaxy. We also analyzed why such an unusual epitaxial growth takes place on α-Al_2O_3(0001) surface.

Finally, we concluded that our crystal growth simulator is effective to elucidate the epitaxial growth process of metal oxides and to design new metal oxide structures which exhibits unexplored and interesting properties.

References

[1] M. Kubo, Y. Oumi, R. Miura, A. Fahmi, A. Stirling, A. Miyamoto, M. Kawasaki, M. Yoshimoto, and H. Koinuma, J. Chem. Phys. **107**, 4416 (1997).

[2] M. Kubo, Y. Oumi, R. Miura, A. Stirling, A. Miyamoto, M. Kawasaki, M. Yoshimoto, and H. Koinuma, Phys. Rev. B **56**, 13535 (1997).

[3] M. Kubo, Y. Oumi, R. Miura, A. Stirling, A. Miyamoto, M. Kawasaki, M. Yoshimoto, and H. Koinuma, J. Chem. Phys. **109**, 8601 (1998).

[4] M. Kubo, Y. Oumi, R. Miura, A. Stirling, A. Miyamoto, M. Kawasaki, M. Yoshimoto, and H. Koinuma, J. Chem. Phys. **109**, 9148 (1998).

[5] M. Kubo, Y. Oumi, H. Takaba, A. Chatterjee, and A. Miyamoto, J. Phys. Chem. B **103**, 1876 (1999).

[6] M. Kubo, Y. Oumi, H. Takaba, A. Chatterjee, A. Miyamoto, M. Kawasaki, M. Yoshimoto, and H. Koinuma, Phys. Rev. B **61**, 16187 (2000).

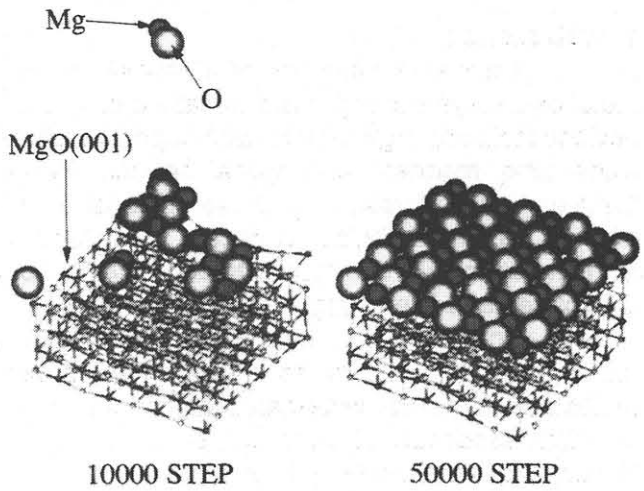

Fig. 1 Homoepitaxial growth process of MgO(001) surface at 1000 K

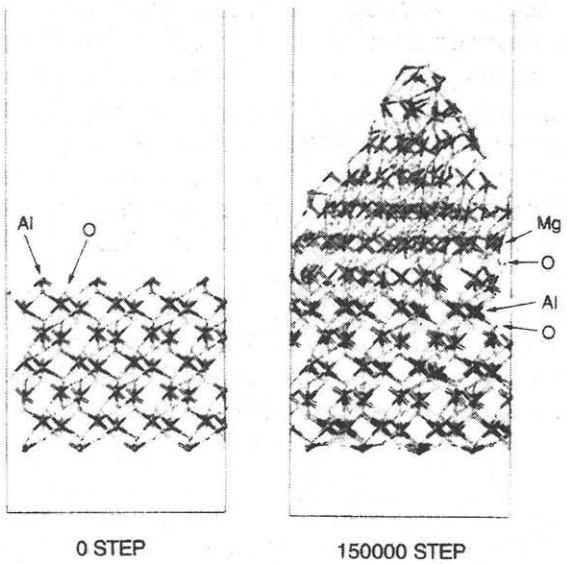

Fig. 2 Crystal growth process of MgO quantum dot on α-Al_2O_3(0001) at 1000 K