Molecular Dynamics Study of Epitaxial Growth and Cluster Formation on MgO(001)

Momoji Kubo, Yasunori Oumi, Ryuji Miura, Andras Stirling, and Akira Miyamoto
Dept. of Molecular Chemistry & Engineering, Tohoku University, Sendai 980-77, Japan

A new molecular dynamics (MD) code was developed to simulate the continuous deposition process of various molecules on substrates. In this methodology, various molecules can be deposited over substrate surfaces one by one in regular time intervals with definite velocity. It enables us to simulate homo- and hetero-epitaxial growth process as well as cluster formation process on various substrates. This new MD code has been already applied to study the homoepitaxial growth and Au cluster formation processes on MgO(001). This article compares such phenomena as growth mode, temperature effect, and influence of surface defects in the homoepitaxial growth and Au cluster formation processes on the MgO(001), based on previous results. The continuous deposition process of MgO molecules was simulated. A 2-D smooth MgO layer without any defects was formed at 1,000 K, which was not realized at 300 K. High temperature is favorable for the complete layer-by-layer homoepitaxial growth. The deposition process of Au atoms on the MgO(001) differed significantly from that of MgO molecules. A 3-D hemispherical Au cluster was constructed on the MgO(001) and low temperature was found to be desired for the fabrication of highly active supported Au catalysts. The origin of the different growth mode and the different temperature effect in the homo-epitaxial growth and Au cluster formation on the MgO(001) was clarified.

Introduction

Fabrication of atomically controlled metal oxide layers has been desired in relation to electronic devices including high-temperature superconducting oxide films, magnetic and optical devices (Kawai and Kishio, 1991; Tachiki et al., 1991; Tsukada and Kawazu, 1992; Koinuma, 1994). On the other hand, artificial construction of ultrafine metal particles on metal oxide substrate has also attracted much attention due to its significance as supported metal catalysts and other advanced materials (Anderson, 1975; Iwasawa, 1986). Hence, atomistic understanding of the homo- and heteroepitaxial growth process of metal oxide surfaces, as well as formation process of metal clusters on metal oxide substrates, is required to realize atomically defined structure that exhibits unexplored and interesting properties.

A lot of significant information has been experimentally accumulated for the epitaxial growth and cluster formation on metal oxide substrates. In addition, theoretical approaches, such as molecular dynamics (MD), quantum chemistry, Monte Carlo simulation, and computer graphics (CG) are also effective tools to understand the mechanism of the epitaxial growth and the cluster formation, as well as to realize atomically controlled structures.

The MD technique has been extensively applied to calculating various properties of organic molecules, polymers, metal oxides, glasses, and zeolites (Yonezawa, 1992; Doyama et al., 1993; Miyamoto et al., 1995). However, a small number of MD simulations was devoted to the crystal growth processes. Since it enables us to understand the atomistic mechanism of different crystal growth modes and the effects of several factors, such as substrate species, substrate temperature, and surface defects on the fabricated materials, the interest and attraction on the crystal growth MD simulation has been gradually stimulated. Schneider, Rahman and Schuller (1985) developed the first MD algorithm to simulate the vapor-phase crystal growth process and applied it to Lennard-Jones system. After their development, the growth process of silicon films was actively studied (Schneider et al., 1987; Gawlinski and Gunton, 1987; Biswas et al., 1988). Furthermore, Luedtke and Landman (1991) employed an em-

---

Correspondence concerning this article should be addressed to Akira Miyamoto.
A. Stirling is on leave from: Institute of Isotopes, Hungarian Academy of Sciences, H-1525 Budapest, P.O.B. 77, Hungary.

bedded-atom method to investigate the continuous deposition process of metals on metal surfaces. Athanasopoulos and Garofalini (1992) studied the continuous deposition process of Pt particles on silica surface. We also investigated the deposition process of a single Au or Pd cluster on MgO(001) surface (Miyamoto et al., 1992a, 1994).

Although the deposition process of monoatomic molecules and metal clusters has been simulated, the continuous deposition process of metal oxide molecules and whole epitaxial growth process on metal oxide substrates have not been calculated to the best of our knowledge. Hence, we have developed a new MD code to simulate the continuous deposition process of metal oxide molecules on various substrates. It is an essential and desired simulator for the design of the atomically defined metal oxide superlattice. The above simulator is also available to simulate the continuous deposition of monoatomic molecules and metal clusters on metal oxide substrates. We have already applied our new MD code to investigate the homoepitaxial growth (Kubo et al., 1997) and Au cluster formation (Kubo et al., 1994, 1995a,b) on the MgO (001) surface. However, since those two processes are individually investigated and presented, the difference between the above two processes was not discussed in the previous articles.

We believed that the knowledge on the origins of the differences of the above two processes is helpful and effective to clarify the mechanism of the crystal growth process and to design the atomically controlled structures. Hence, in the present article, we compared and discussed the different phenomena, such as the growth mode, temperature effect, and influence of the surface defects, in the homoepitaxial growth and Au cluster formation processes, on the basis of the summary of the previous results. The origin of the different growth mode and the different temperature effect in the homoepitaxial growth and Au cluster formation on the MgO (001) was clarified in the present article.

Methodology of New MD Code

We developed a new MD code to investigate the epitaxial growth and cluster formation processes on substrates. This MD code was based on MXDORTO code of Kawamura (1992). Our main modifications concern the total number of species in the system which is not fixed but increases with time. Figure 1 shows the model system of these MD simulations which consists of two parts, namely a substrate and a source of emitting molecules. The number of molecules deposited over the substrate surface is increased one-by-one. The molecules are shot to the surface in regular time intervals with definite velocity. Since various kinds of depositing molecules and substrates are capable of being simulated in our MD code, the effect of depositing species and substrate on the structure of constructed materials and the interface structures of various heterojunctions can be clarified. Moreover, the temperature of substrate, the distance between the substrate and the source of emitting molecules, the number of the incident molecules, the incident rates of the molecules, and the time interval between the incidence of the molecules are variable parameters. Hence, various experimental conditions are expected to be optimized by the new MD code. The horizontal positions of the emerged molecules over substrates were randomly selected.

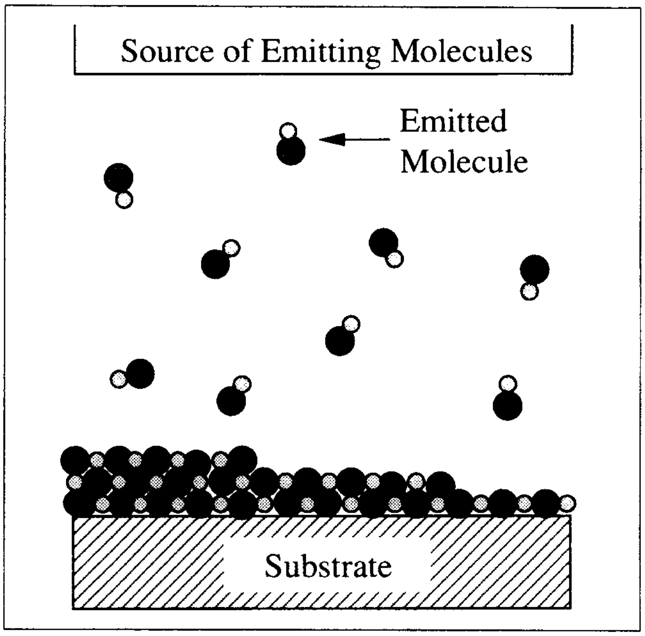

Figure 1. Simulation model for the epitaxial growth process and cluster formation on substrates.

The Verlet algorithm was used for the calculation of the atomic motions, while the Ewald method was applied for the calculation of the electrostatic interactions. Temperature was controlled by means of scaling the atom velocities. The calculations were performed for 50,000–80,000 steps with a time step of 2.0×10^{-15} s at 300–1,000 K. The two-body, central force interatomic potential, as shown in Eq. 1, was used for all calculations. In Eq. 1, the first, second, and third terms refer to Coulomb, exchange repulsion, and Morse interactions, respectively.

$$u(r_{ij}) = Z_iZ_je^2/r_{ij} + f_o(b_i + b_j)\times\exp[(a_i + a_j - r_{ij})/(b_i + b_j)] + D_{ij}\{\exp[-2\beta_{ij}(r_{ij} - r_{ij}^*)] - 2\exp[-\beta_{ij}(r_{ij} - r_{ij}^*)]\} \quad (1)$$

where Z_i is the atomic charge, e is the elementary electric charge, r_{ij} is the interatomic distance, and f_o is a constant. The parameters a and b in Eq. 1 represent the size and stiffness, respectively, in the exchange repulsion interaction, while D_{ij}, r_{ij}^*, and β_{ij} represent bond energy, equilibrium bond distance, and stiffness, respectively, in the Morse function. The parameters of Eq. 1 were determined to reproduce various properties of metal oxides and metal crystals, such as lattice constant, expansion coefficient, melting point, and so on (Miyamoto et al., 1992a; Kubo et al., 1994; Kubo et al., 1997) by using density functional calculations and MD simulations.

Calculations were performed on Hewlett Packard Apollo 9000 Model 710 workstation, while the visualization was made with a Silicon Graphics IRIS Indigo2 workstation and MSI Insight II software. Dynamic features in the epitaxial growth and cluster formation processes were also investigated by using real-time CG visualization with the MOMOVIE code and RYUGA code (Miura et al., 1995) developed in our laboratory on OMRON LUNA-88K and Hewlett-Packard Apollo 9000 Model 715/33 workstations, respectively.

Homoepitaxial Growth Process of MgO(001) at 300 K

Much attention has been given to the MgO thin film fabrication due to its importance as high-temperature material, unreactive substrate, secondary electron emission material, wide-gap insulator, model catalyst, and other practical materials. Hence, a number of interesting experimental works on the epitaxial growth of MgO thin films on various substrates, such as MgO (Booth et al., 1975; Yadavalli et al., 1990; Chambers et al., 1994), silicon (Huang and Kitai, 1992), quartz (Kwak et al., 1989), sapphire (DeSisto and Henry, 1990) and so on, have been done already by using various techniques, such as the molecular beam epitaxy (MBE), chemical vapor deposition (CVD), sputtering, X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), Auger electron spectroscopy (AES), reflection high-energy electron diffraction (RHEED), low-energy electron diffraction (LEED), transmission electron microscopy (TEM), atomic force microscope (AFM), and so on. Recently, MgO thin film has been actively examined as a buffer layer for high *Tc* superconducting metal oxide film grown on metal oxide substrate (Berezin et al., 1990; Fork et al., 1991). Since the atomically flat, smooth and dense MgO buffer layer is required to enhance the growth of uniform superconducting oxides, the studies on the atomistic control of the surface, interface, and growth process of MgO has been rapidly stimulated.

Hence, the new MD code and CG technique were applied to the investigation of the homoepitaxial growth process of MgO(001) surface (Kubo et al., 1997). A total of 32 MgO molecules were deposited on the MgO(001) plane one by one with the incident rate of 900 m/s. Here, this amount is equal to the number of the Mg and O atoms which constitute a single MgO layer in the present MD unit cell. Figure 2 shows the homoepitaxial growth process of the MgO(001) plane at 300 K. The MgO substrate is represented by atomic bonds, while the deposited MgO molecules are represented by spheres. After some MgO molecules migrated on the MgO(001) plane, a 2-D and epitaxial growth of the MgO thin layer was constructed at 300 K. Moreover, the deposited MgO molecules were keeping the NaCl-type structure and the (001) oriented configuration during the MD simulation. It indicates that the epitaxial growth of MgO(001) plane follows the Frank-van der Merwe growth mode. Experimentally, Yadavalli, Yang and Flynn (1990) employed a beam of evaporated MgO for the homoepitaxial growth of MgO(001) plane, and observed epitaxial growth of MgO(001) plane with NaCl-type structure in the wide temperature range of 140 K-1,300 K. The present simulation at 300 K reproduced well their experiments on an atomic scale.

However, after all the 32 MgO molecules were deposited on the MgO(001) plane, some defects in the first constructed MgO layer were observed, and some MgO molecules already constructed a second MgO layer. Thus, it was found that the complete layer-by-layer homoepitaxial growth of MgO thin film without defects was not achieved at a low temperature of 300 K.

Temperature Effect on the Homoepitaxial Growth Process of MgO(001)

Experimentally, the substrate temperature is one of the important parameters to construct well-defined MgO thin layers. Therefore, the understanding of the effect of the substrate temperature on the homoepitaxial growth process is essential to optimize the fabrication conditions of the atomically controlled MgO thin films. Hence, the homoepitaxial growth process of the MgO(001) surface at the high temperature of 1,000 K was simulated (Kubo et al., 1997) and the result is shown in Figure 3. The MgO grew epitaxially keeping NaCl-type structure and (001) oriented configuration even at this high temperature. Moreover, it was surprising to see the formation of a single 2-D uniform and flat layer of MgO

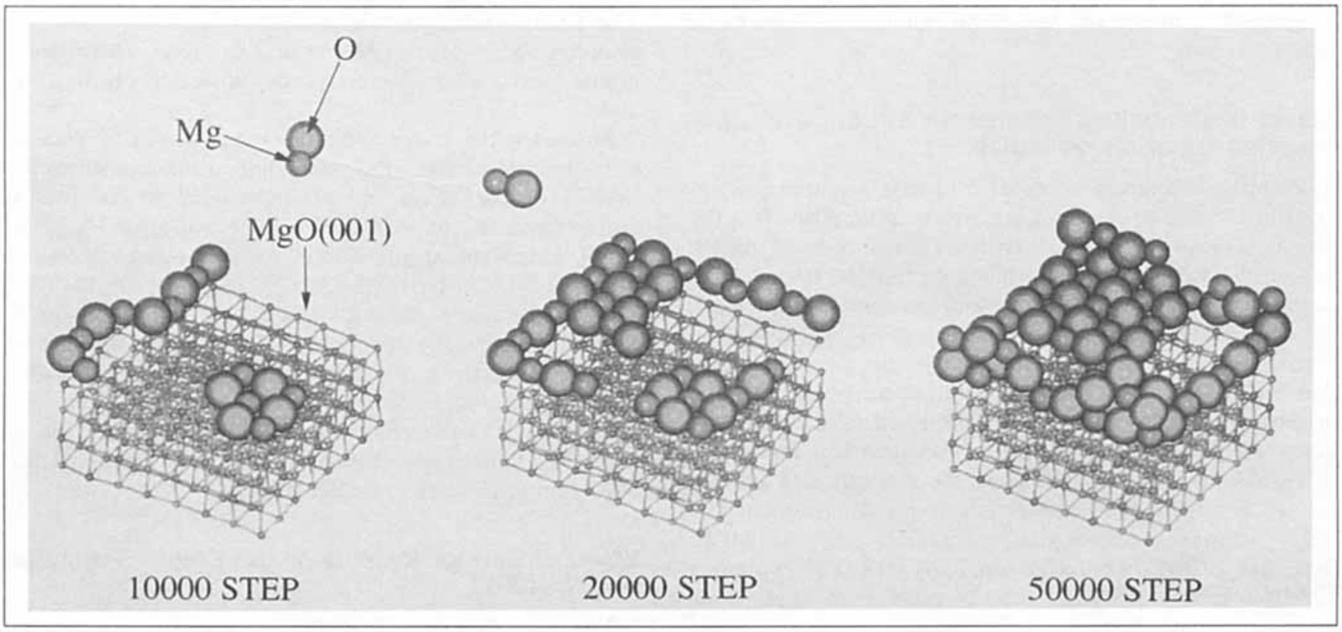

Figure 2. Homoepitaxial growth process of MgO(001) plane at 300 K.

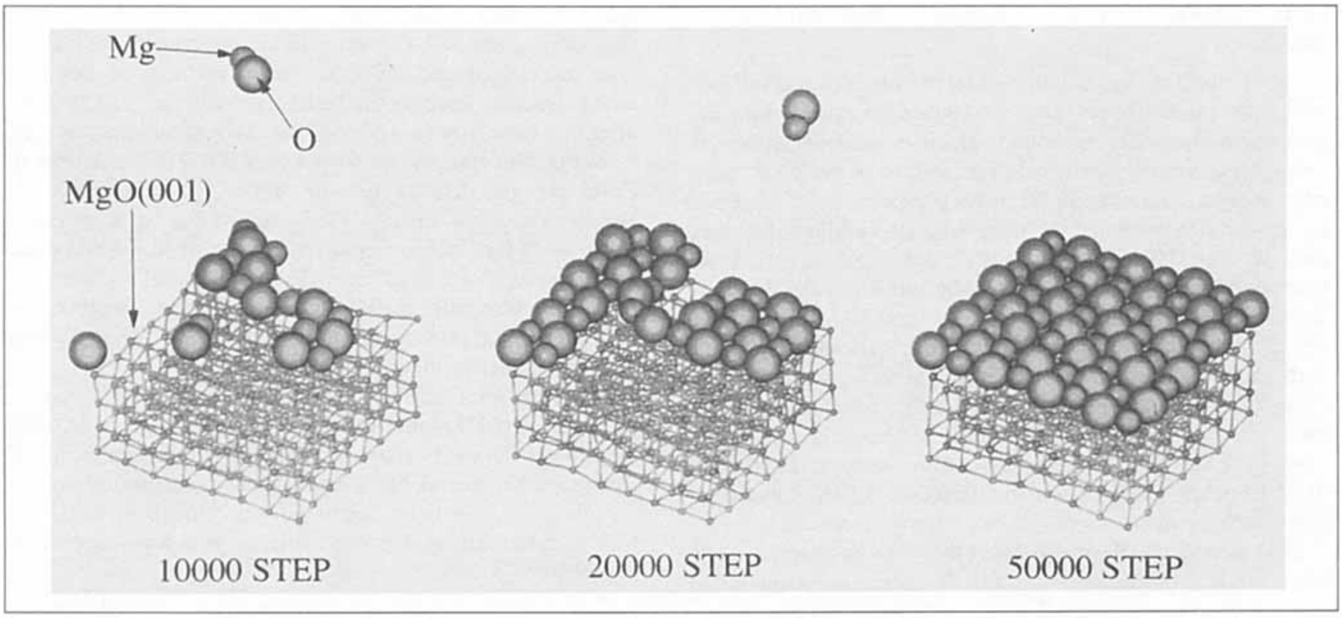

Figure 3. Homoepitaxial growth process of MgO(001) plane at 1,000 K.

without any defects at the 50,000 time step, which is significantly different from the result at 300 K. Therefore, high temperature is desired for the complete layer-by-layer homoepitaxial growth. Experimentally, Chambers, Tran and Hileman (1994) applied the MBE technique to the homoepitaxial growth of the MgO(001) plane and investigated the surface morphology of the grown MgO film at various temperatures by using RHEED, LEED, and AFM measurements. They reported that the surfaces of the MgO films grown on MgO(001) plane at 923 K and 1,023 K are much smoother than those obtained at lower temperature such as 298 K, which agreed with the present MD simulations. Since the 2-D growth mode, the structure of constructed material, and the effect of substrate temperature were reproduced by the MD simulations, the efficiency of our new MD code was confirmed to predict and design the atomically defined metal oxide thin layer.

Effect of Deposited Species on Structure of Constructed Materials on MgO(001)

Computer chemistry is expected to have a role in predicting the detailed structure of undeveloped materials. It is the key technology to realize the artificial design of novel materials which exhibit unexplored functions. Hence, the effect of deposited species on the structure of the constructed materials on MgO(001) surface should be accurately reproduced by the MD simulations. On the other hand, supported gold catalyst has high activity for low-temperature oxidation of carbon monoxide (Haruta et al., 1989, 1993; Knell et al., 1992) and many other interesting reactions (Galvagno and Parravano, 1978; Sakurai et al., 1993). Hence, the structure and location of Au clusters on MgO surface has been well investigated by using various experimental techniques, such as MBE, RHEED, LEED, high resolution TEM (HRTEM), scanning electron microscope (SEM), and so on (Robins et al., 1966; Cowley and Neumann, 1984; Duriez et al., 1990; Giorgio et al., 1991). It was expected that the structure of fabricated materials by the deposition of Au atoms is significantly different from that by the deposition of MgO molecules. Hence, we applied our new MD code to simulate the continuous deposition process of Au atoms on the MgO(001) plane (Kubo et al., 1994). Figure 4 shows the continuous deposition process of 20 Au atoms on the MgO(001) plane with the incident rate of 180 m/s at 300 K. When any Au atom approached the MgO(001) plane, it initially migrated on the surface with high mobility. After the Au atoms aggregated gradually, a 3-D and hemispherical Au_{19} cluster was formed on the MgO(001) plane. It indicates that the deposition process of Au atoms follows the Volmer-Weber growth mode which agrees well with the experimental results (Giorgio et al., 1991). However, this structure is in significant contrast to that obtained by the deposition of MgO molecules as shown in Figures 2 and 3. The homoepitaxial growth process of MgO(001) plane follows the Frank-van der Merwe growth mode. The different crystal growth modes were reproduced well by our new MD code.

Moreover, the above different crystal growth mechanisms were interpreted from the standpoint of the atomic interactions. The larger atomic interaction between the Au atom-Au atom, compared to that between the Au atom-MgO substrate, causes the aggregation of Au atoms and the fabrication of a 3-D cluster on the MgO(001) plane. On the other hand, in the case of the homoepitaxial growth process of the MgO(001) plane, the deposited MgO molecules are inclined to adhere directly to the substrate instead of the aggregation, due to the stronger atomic interaction between the MgO molecule-MgO substrate compared to that between the Au atom–MgO substrate. Hence, it leads a 2-D and epitaxial MgO thin layer on the MgO(001) plane.

Effect of Surface Defects on the Cluster Formation on MgO(001)

The effect of defects in MgO surface on the structure and location of the Au cluster on the MgO surface would be interesting and has been investigated by using various experimental techniques (Robins et al., 1966; Cowley and Neumann, 1984; Duriez et al. 1990). Therefore, MD simulations were performed to understand the effect of surface defects in the MgO(001) plane on an atomic scale (Kubo et al., 1995a). Figure 5 shows the formation process of an Au cluster on the MgO(001) plane with a point defect which is one Mg^{2+} and one O^{2-} vacancies at 300 K. After 80,000 time steps, the formation of a hemispherical Au cluster was observed just over the point defect in the MgO(001) plane at 300 K. The favorable location for the Au cluster, the site on top of the point defect in the MgO(001) plane, was detected at 300 K. In other words, the surface defects on the MgO(001) plane significantly affect the location of Au clusters on the surface at the low temperature of 300 K in agreement with the experimental results (Robins et al., 1966; Cowley and Neumann, 1984; Duriez et al., 1990).

In order to clarify the detailed mechanism of the above process, attention was given to the dynamics and behavior of each Au atom during the formation process. It was observed that an Au atom was trapped in the point defect on the MgO(001) plane at the 16,000 time step as shown in Figure 5. After other Au atoms came in contact with the trapped Au atom in the point defect, the gradual formation of the Au cluster occurred over the point defect. Finally, a hemispherical Au cluster was formed just over the point defect in the MgO(001) plane. Hence, the Au atom trapped in the point defect has a role of nucleation center in the Au cluster formation on the MgO(001) plane at 300 K. This is a reason why the Au cluster was formed just over the point defect in the MgO(001) plane.

Moreover, the deposition process of MgO molecules on the MgO(001) plane with a point defect was also simulated. However, the obvious effect was not obtained, which is in marked contrast to the above Au cluster formation process.

Temperature Effect on the Cluster Formation on MgO(001)

The effect of substrate temperature on the formation process of Au cluster on the MgO(001) plane was investigated (Kubo et al., 1995a), and the result was compared with the temperature effect in the homoepitaxial growth process of the MgO(001) plane. The deposition process of 20 Au atoms on the smooth MgO(001) plane was simulated at 1,000 K in order to compare the result at 300 K as shown in Figure 4. Consequently, 5 Au atoms evaporated from the surface and 15 Au atoms were fixed on the MgO(001) plane at 1,000 K, which is in contrast to the result at 300 K. In the latter case, only the 1 Au atom evaporated and the Au_{19} cluster was constructed on the MgO(001) plane. It indicates that the high substrate temperature promotes the evaporation of Au atoms from the MgO(001) plane. Hence, low temperature was found to be suitable to fix more Au atoms efficiently on the MgO(001) plane, which is a different temperature effect in the case of the deposition process of the MgO molecules. This result was also interpreted from the standpoint of the atomic interactions. The larger atomic interaction between the MgO molecule–MgO substrate, compared to that between the Au atom–MgO substrate, causes the low migration ability of the MgO molecules. Hence, the increment of substrate temperature leads to the higher diffusivity of the deposited MgO molecules and the complete layer-by-layer homoepitaxial growth. On the other hand, the Au atoms on the MgO(001) plane have high migration ability even at low temperature because of the small interaction between the Au atom–MgO substrate. Hence, the increment of substrate temperature changes the high migration ability of Au atoms into the evaporation ability.

The effect of substrate temperature on the formation process of Au cluster on the MgO(001) plane with a point defect was also examined. Figure 6 shows the formation process of Au clusters on the MgO(001) plane with a point defect at 1,000 K. A hemispherical Au cluster was formed on an unspecified site at 1,000 K, which is also in remarkable contrast to that at 300 K, as shown in Figure 5. This result indicates that there is no single favorable location of the Au cluster on the MgO(001) plane with a point defect at 1,000 K. This behavior is similar to that on the smooth MgO(001) plane, as shown in Figure 4. It was revealed that the presence of the point defects does not greatly affect the location of the Au cluster deposited over the MgO(001) plane at the high temperature of 1,000 K. The above mechanism was interpreted as follows. After an Au atom was trapped in the point defect on the MgO(001) plane at the 20,000 time step, it was released from the point defect at the 28,000 time step. Moreover, another Au atom was trapped in the point defect at the 36,000 time step again; however, it was also released from the point defect at the 40,000 time step. The higher vibrational mobility of the Mg^{2+} and O^{2-} ions adjacent to the

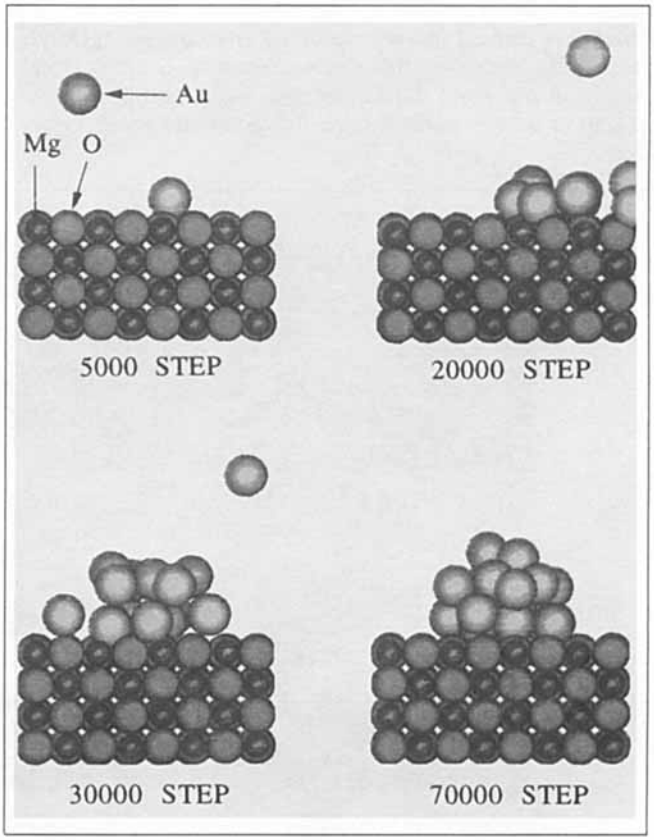

Figure 4. Formation process of an Au cluster on MgO(001) plane at 300 K.

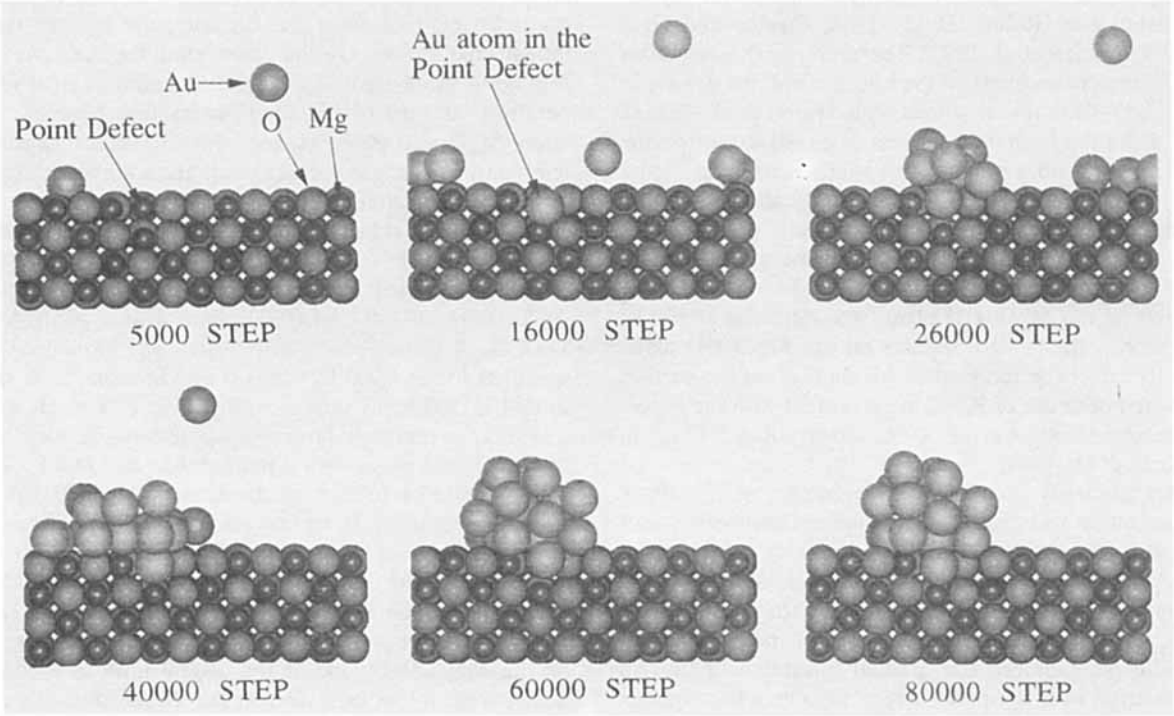

Figure 5. Formation process of an Au cluster on MgO(001) plane with a point defect at 300 K.

defect at higher temperatures was one of the reasons for the ready release of the trapped Au atom, in addition to the higher translational mobility of Au atoms. Finally, an Au cluster was formed in an unspecified site on the MgO(001) plane. This is due to the ready release of the Au atoms trapped in the point defect and the less probability of the nucleation center formation in the point defect at higher temperatures. In other words, the higher mobility, which is the consequence of higher temperature, led to the random location for the ultrafine Au particles on the MgO(001) plane with a point defect. These results suggested that the low temperatures of the MgO substrate lead to atomically controlled ultrafine Au particles on the MgO(001) plane which are desirable for the advanced materials.

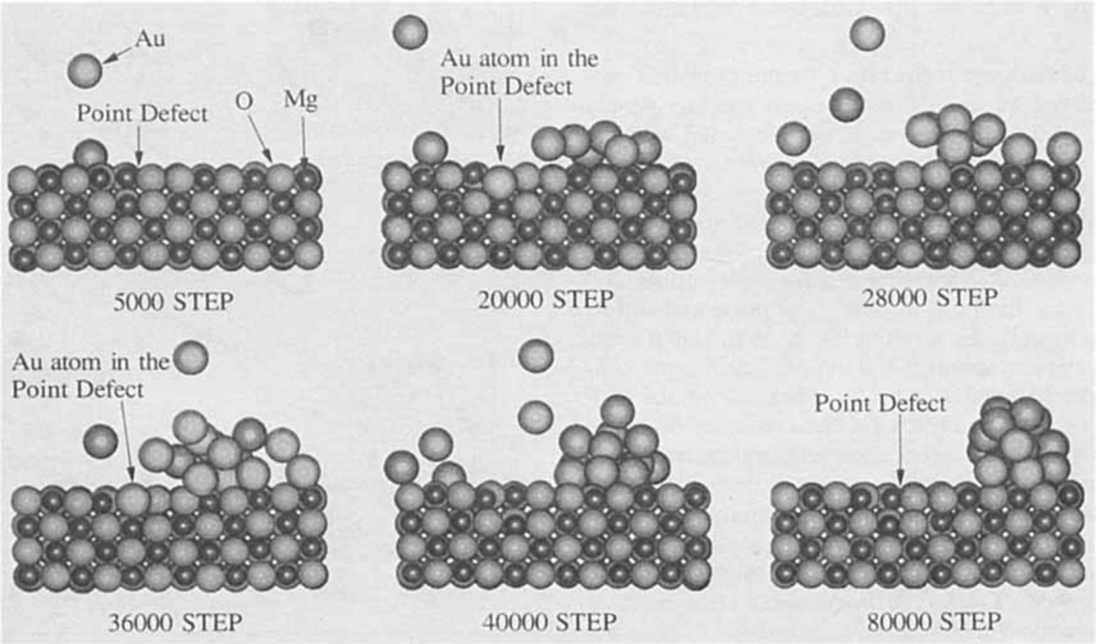

Figure 6. Formation process of an Au cluster on MgO(001) plane with a point defect at 1,000 K.

Moreover, we simulated the continuous deposition process of Au atoms on the MgO(001) plane with two point defects at 300 K and 1,000 K (Kubo et al., 1995b). Two separated Au cluster was constructed just on the two point defects at 300 K, while only a single large Au cluster was formed on an unspecified site at 1,000 K. These phenomena were readily understood from the discussion made in the Au cluster formation process on the MgO with a single point defect. Two point defects have a role of nucleation centers of two separated Au clusters at a low temperature, while the high diffusivity of Au atoms prohibits the above role of point defects at a high temperature. Industrially, it is essential to develop highly dispersed metal catalysts because high surface area is essential for the high turnover frequencies of catalyst reactions. Hence, low temperature and surface defects were found to be favorable for the fabrication of high active catalysts. This temperature effect was different to that of the homoepitaxial growth process of the MgO(001) plane.

Conclusion

We successfully simulated a 2-D homoepitaxial growth of the MgO(001) plane and a 3-D Au cluster formation on the MgO(001) plane. The above two different crystal growth mechanisms were clarified in the present study from the standpoint of the atomic interactions. The larger atomic interaction between the Au–Au, compared to that between the Au–MgO, causes the aggregation of Au atoms and the fabrication of a 3-D cluster on the MgO(001) plane. On the other hand, the deposited MgO molecules are inclined to adhere directly to the MgO(001) plane instead of the aggregation, due to the stronger atomic interaction between the MgO–MgO compared to that between the Au–MgO. It leads a 2-D and epitaxial MgO thin layer on the MgO(001) plane.

Moreover, the origin of the temperature effect was also discussed. The larger atomic interaction between the MgO-MgO, compared to that between the Au–MgO, causes the low migration ability of the MgO molecules. Hence, the increment of substrate temperature leads to the higher diffusivity of the deposited MgO molecules and the complete layer-by-layer homoepitaxial growth. On the other hand, the Au atoms on the MgO(001) plane have a high migration ability even at a low temperature because of the small interaction between the Au–MgO. Hence, the increment of substrate temperature changes the high migration ability of Au atoms into the evaporation ability. Moreover, the different effects of the surface defects on the above two processes were also clarified.

Finally, the efficiency was confirmed of our new MD code in the clarification of the crystal growth mechanism and in the design of the atomically controlled structure on substrates.

Literature Cited

Anderson, J. R., *Structure of Metallic Catalysts*, Academic Press, New York (1975).

Athanasopoulos, D. C., and S. H. Garofalini, "Molecular Dynamics Simulations of the Effect of Adsorption on SiO_2 Surfaces," *J. Chem. Phys.*, **97**, 3775 (1992).

Berezin, A. B., C. W. Yuan, and A. L. de Lozanne, "$Y_1Ba_2Cu_3O_{7-x}$ Thin Films Grown on Sapphire with Epitaxial MgO Buffer Layers," *Appl. Phys. Lett.*, **57**, 90 (1990).

Biswas, R., G. S. Grest, and C. M. Soukoulis, "Molecular-Dynamics Simulation of Cluster and Atom Deposition on Silicon(111)," *Phys. Rev. B.*, **38**, 8154 (1988).

Booth, J. R., W. D. Kingery, and H. K. Bowen, "Preparation and Properties of MgO Single Crystals Grown by Chemical Vapor Deposition," *J. Cryst. Growth*, **29**, 257 (1975).

Chambers, S. A., T. T. Tran, and T. A. Hileman, "Molecular Beam Homoepitaxial Growth of MgO(001)," *J. Mater. Res.*, **9**, 2944 (1994).

Cowley, J. M., and K. D. Neumann, "The Alignment of Gold Particles on MgO Crystal Faces," *Surf. Sci.*, **145**, 301 (1984).

DeSisto, W. J., and R. L. Henry, "Deposition of (100) Oriented MgO Thin Films on Sapphire by a Spray Pyrolysis Method," *Appl. Phys. Lett.*, **56**, 2522 (1990).

Doyama, M., J. Kihara, M. Tanaka, and R. Yamamoto, eds., *Computer Aided Innovation of New Materials II*, North-Holland, Amsterdam (1993).

Duriez, C., C. Chapon, C. R. Henry and J. M. Rickard, "Structural Characterization of MgO(100) Surfaces," *Surf. Sci.*, **230**, 123 (1990).

Fork, D. K., F. A. Ponce, J. C. Tramontana, and T. H. Geballe, "Epitaxial MgO on Si(001) for Y-Ba-Cu-O Thin-Film Growth by Pulsed Laser Deposition," *Appl. Phys. Lett.*, **58**, 2294 (1991).

Galvagno, S., and G. Parravano, "Chemical Reactivity of Supported Gold IV. Reduction of NO by H_2," *J. Catal.*, **55**, 178 (1978).

Gawlinski, E. T., and J. D. Gunton, "Molecular-Dynamics Simulation of Molecular-Beam Epitaxial Growth of the Silicon (100) Surfaces," *Phys. Rev. B*, **36**, 4774 (1987).

Giorgio, S., C. Chapon, C. R. Henry, G. Nihoul, and J. M. Penisson, "High-Resolution Transmission Electron Microscopy Study of Gold Particles (Greater than 1 nm), Epitaxially Grown on Clean MgO Microcubes," *Phil. Mag. A.*, **64**, 87 (1991).

Haruta, M., N. Yamada, T. Kobayashi, and S. Iijima, "Gold Catalysts Prepared by Coprecipitation for Low-Temperature Oxidation of Hydrogen and of Carbon Monoxide," *J. Catal.*, **115**, 301 (1989).

Haruta, M., S. Tsubota, T. Kobayashi, H. Kageyama, M. J. Genet, and B. Delmon, "Low-Temperature Oxidation of CO over Gold Supported on TiO_2, α-Fe_2O_3, and Co_3O_4," *J. Catal.*, **144**, 175 (1993).

Huang, R., and A. H. Kitai, "Temperature-Dependence of the Growth Orientation of Atomic Layer Growth MgO," *Appl. Phys. Lett.*, **61**, 1450 (1992).

Iwasawa, Y., ed., *Tailored Metal Catalysts*, Reidel, Dordrecht (1986).

Kawai, M., and K. Kishio, eds., *Chemical Designing and Processing of High-Tc Superconductors*, North-Holland, Amsterdam (1991).

Kawamura, K., "Interatomic Potential Models for Molecular Dynamics Simulations of Multi-Component Oxides," *Molecular Dynamics Simulations*, F. Yonezawa, ed., Springer-Verlag, Berlin, 88 (1992).

Knell, A., P. Barnickel, A. Baiker, and A. Wokaun, "CO Oxidation over Au/ZrO_2 Catalysts: Activity, Deactivation, Behavior, and Reaction Mechanism," *J. Catal.*, **137**, 306 (1992).

Koinuma, H., ed., *Crystal Engineering of High Tc-Related Oxide Films*, MRS, Pittsburgh (1994).

Kubo, M., R. Yamauchi, R. Vetrivel, and A. Miyamoto, "Formation Processes of Ultrafine Metal Particles on MgO(100) as Investigated by Molecular Dynamics and Computer Graphics," *Appl. Surf. Sci.*, **82/83**, 559 (1994).

Kubo, M., R. Miura, R. Yamauchi, R. Vetrivel, and A. Miyamoto, "Mechanism of the Formation of Ultrafine Gold Particles on MgO(100) as Investigated by Molecular Dynamics and Computer Graphics," *Appl. Surf. Sci.*, **89**, 131 (1995a).

Kubo, M., R. Miura, R. Yamauchi, M. Katagiri, R. Vetrivel, E. Broclawik, and A. Miyamoto, "Atomic Control of Ultrafine Gold Particles on MgO(100) as Investigated by Molecular Dynamics and Computer Graphics," *Jpn. J. Appl. Phys.*, **34**, 6873 (1995b).

Kubo, M., Y. Oumi, R. Miura, A. Stirling, A. Miyamoto, M. Kawasaki, M. Yoshimoto, and H. Koinuma, "Layer-by-Layer Homoepitaxial Growth Process of MgO(001) as Investigated by Molecular Dynamics, Density Functional Theory, and Computer Graphics," *J. Chem. Phys.*, in press (1997).

Kwak, B. S., E. P. Boyd, K. Zhang, E. Erbil, and B. Wilkins, "Metal-organic Chemical Deposition of [100] textured MgO Thin Films," *Appl. Phys. Lett.*, **54**, 2542 (1989).

Luedtke, W. D., and U. Landman, "Metal-on-Metal Thin-Film

Growth: Au/Ni(001) and Ni/Au(001)," *Phys. Rev. B*, **44**, 5970 (1991).

Miura, R., H. Yamano, R. Yamauchi, M. Katagiri, M. Kubo, R. Vetrivel, and A. Miyamoto, "Development of RYUGA for Three-Dimensional Dynamic Visualization of Molecular Dynamics Results," *Catal. Today*, **23**, 409 (1995).

Miyamoto, A., T. Hattori, and T. Inui, "Molecular Dynamics Simulation of Deposition Process of Ultrafine Metal Particles on MgO(100) Surface," *Appl. Surf. Sci.*, **60/61**, 660 (1992a).

Miyamoto, A., R. Yamauchi, and M. Kubo, "Atomic Processes in the Deposition and Sintering of Ultrafine Metal Particles on MgO(001) as Investigated by Molecular Dynamics and Computer Graphics," *Appl. Surf. Sci.*, **75**, 51 (1994).

Miyamoto, A., T. Hattori, and M. Misono, eds., *The Impact of Computers on Catalyst Research and Development*, Elsevier, Amsterdam (1995).

Robins, J. L., T. N. Rhodin and R. L. Gerlach, "Dislocation Structures in Cleaved Magnesium Oxide," *J. Appl. Phys.*, **37**, 3893 (1966).

Sakurai, H., S. Tsubota, and M. Haruta, "Hydrogenation of CO_2 over Gold Supported on Metal Oxides," *Appl. Catal. A*, **102**, 125 (1993).

Schneider, M., A. Rahman. and I. K. Schuller, "Role of Relaxation in Epitaxial Growth: A Molecular-Dynamics Study," *Phys. Rev. Lett.*, **55**, 604 (1985).

Schneider, M., I. K. Schuller, and A. Rahman, "Epitaxial Growth of Silicon: A Molecular-Dynamics Simulation," *Phys. Rev. B*, **36**, 1340 (1987).

Tachiki, T., Y. Muto, and Y. Shono, eds., *Proc. of Int. Conf. on Materials and Mechanisms of Superconductivity High Temperature Superconductors III*, Kanazawa, Japan, North-Holland, Amsterdam (1991).

Tsukada, M., and A. Kawazu, eds., *Automatically Controlled Surfaces and Interfaces*, North-Holland, Amsterdam (1992).

Yadavalli, S., M. H. Yang, and C. P. Flynn, "Low-Temperature Growth of MgO by Molecular-Beam Epitaxy," *Phys. Rev. B*, **41**, 7961 (1990).

Yonezawa, F. ed., *Molecular Dynamics Simulation*, Springer-Verlag, Berlin (1992).

Manuscript received Oct. 28, 1996, and revision received Apr. 16, 1997.