Growth and Characterization of Ultrathin Fe Films on Molecule-Adsorbed MgO Surfaces

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Effects of the molecule-adsorption on a MgO surface on the growth of ultra-thin Fe films were investigated. Surface observation by reflection high-energy electron diffraction and ultra-high vacuum atomic force microscopy showed that the adsorption of molecules on the MgO surface affected the morphology of Fe films grown on the MgO surface. It was revealed that the surface flatness of Fe films was improved by appropriate molecule-adsorption on the MgO surface. [doi:10.2320/matertrans.M2009218]

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1. Introduction

Ultra-thin films have attracted a lot of attention from the viewpoint of understanding physics in two-dimensional electron systems. Two-dimensional electron systems are formed typically in semiconductor hetero-junctions. These systems show interesting phenomena, such as Shubnikov-de Haas oscillation¹⁾ and quantum Hall effect²⁾ at very low temperatures. Two-dimensional electron systems formed in ultra-thin metallic films also show quantum effects due to interference and confinement of electrons. It is known that thin films of ferromagnetic metals clearly exhibit spin-dependent quantum well states.^{3,4)} In previous studies, ultra-thin metallic films were grown on metallic buffer layers. If the ultra-thin metallic films are grown on an insulator, the further development of spintronic devices such as spin-polarized resonant tunneling transistors which are demonstrated by gate-control of spin-dependent quantum well states is expected.^{5,6)} However, the growth of extremely thin metallic films with a few atomic layers on insulators is quite difficult because three-dimensional island structures are easily formed. Hence, only a few studies on the growth of ultra-thin metallic films on insulators have been reported to date.^{7–11)}

In the first stage of film growth, the metals that have higher surface energies than that of an insulating substrate are generally grown three-dimensionally. The migration of metallic atoms promotes the formation of island structure, and not continuous structure. The growth of metallic thin films using adsorption of atoms or molecules on surfaces of insulators was reported.^{12,13)} M. Lust *et al.* reported that crystalline qualities of ultrathin Fe and Cu films grown on MgO were improved by adsorption of CO molecules on a MgO surface.¹⁴⁾ In addition, it was reported that dissociative adsorption of H₂O molecules occurs on defect sites of the MgO surface, which seems to be an advantage to grow epitaxial films on MgO substrates.^{15,16)} Therefore, the

adsorption of CO or H₂O molecules on the MgO surface has a possibility of realizing the growth of ultra-thin continuous ferromagnetic films.

In this paper, we have investigated the effects of molecule-adsorption on a MgO surface on the growth of ultra-thin Fe films. CO and H₂O were adsorbed on MgO surfaces, and Fe films with a nominal thickness of a few atomic layers were grown on the molecule-adsorbed surfaces. Surface morphology was observed by reflection high-energy electron diffraction and atomic force microscopy. A characteristic feature in this study is that these direct methods were used to investigate effects of adsorption.

2. Experimental Procedure

All the growth processes were performed at room temperature by molecular beam epitaxy with evaporation sources in an ultra-high vacuum (UHV) chamber (base pressure: 1×10^{-7} Pa). MgO(001) substrates were annealed at 600°C for 30 min in UHV, and MgO buffer layers (5 nm) were grown at room temperature. Then, clean MgO(001) surfaces were obtained by annealing at 600°C for 30 min. Ultra-thin Fe films with a nominal thickness of 0.3 nm were grown on the MgO surfaces. The CO-adsorbed MgO surface was prepared by introduction of CO gas with a partial pressure of 4×10^{-6} Pa from a variable leak valve for 10 min equipped in the UHV chamber. The growth of Fe film was started after closing the valve, and the partial pressure of CO gas decreased to the order of 10^{-8} Pa. To adsorb H₂O molecules on a MgO surface, the MgO substrate with the buffer layer mentioned above was unloaded from the UHV chamber, and dipped into ultra-pure water for 5 s. The H₂O-adsorbed MgO surface was annealed again at 100°C for 3 min in the UHV chamber, and an Fe film was grown on the surface. The surface morphology was monitored by reflection high-energy electron diffraction (RHEED) just after deposition and after annealing. The surface profile and the surface flatness of the Fe films were investigated by ultra-high vacuum atomic force microscopy (AFM) using a contact mode.

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Fig. 1 RHEED patterns for (a) a MgO(001) buffer layer after annealing, and for Fe layers with a thickness of 0.3 nm just after deposition on (b) non-adsorbed MgO, (c) CO-adsorbed MgO, (d) H₂O-adsorbed MgO. The incident beam was directed along the azimuth of MgO(110).

3. Results and Discussion

Figure 1(a) shows a RHEED pattern from a MgO buffer layer after annealing. A streaky pattern, which indicates the formation of a flat surface, was observed. An Fe film grown on the non-adsorbed MgO surface shows a spotty pattern as shown in Fig. 1(b). This RHEED pattern indicates the epitaxial island growth of bcc-Fe with an epitaxial orientation relationship of Fe(110) // MgO(100). A similar spotty pattern was also observed for the Fe layer grown on the CO-adsorbed MgO surface (Fig. 1(c)). A RHEED pattern from the Fe layer grown on the H₂O-adsorbed MgO surface revealed a slightly fuzzy pattern as shown in Fig. 1(d). This implies a poor crystalline quality of the Fe film.

Figure 2 shows AFM images of the surfaces of a MgO buffer layer and Fe films. In Fig. 2(a) which shows the surface of the MgO buffer layer, where bright particle-like regions and dark voids dispersed over the entire surface are observed. On the surface of an Fe film grown on non-adsorbed MgO as shown in Fig. 2(b), a similar image to Fig. 2(a) is observed. On the surface of an Fe film grown on CO-adsorbed MgO as shown in Fig. 2(c), a number of distinct voids are observed. On the surface of an Fe film grown on H₂O-adsorbed MgO as shown in Fig. 2(d), the numbers of bright regions and voids have been reduced. No apparent difference in morphology is seen in this view-scale between the MgO buffer layer (Fig. 2(a)) and the Fe film grown on non-adsorbed MgO (Fig. 2(b)). The number of bright regions in Fig. 2(c) seems to be smaller than that in Fig. 2(b). The bright regions are not observed in Fig. 2(d). These results reveal that the adsorption of molecules such as CO or H₂O on the MgO surface apparently affects the morphology of the Fe thin film. Magnified images of Figs. 2(a), (b), (c) and (d) are shown in Figs. 2(e), (f), (g) and (h), respectively. Figure 2(f) indicates that small Fe particles were grown on non-adsorbed MgO. Fe particles are also observed in the magnified images of the Fe films grown on CO-adsorbed MgO (Fig. 2(g)) and on H₂O-adsorbed MgO (Fig. 2(h)). It is found that bright regions in Fig. 2(b), (c) and (d) consist of a number of Fe particles. The sizes of Fe

Table 1 Surface roughness (Ra) of a MgO buffer layer and Fe films grown on various MgO surfaces.

Ra/nm	MgO buffer	Ultra-thin Fe films		
		Non-ad.	CO-ad.	H ₂ O-ad.
All area	0.79	1.0	0.89	0.72
Excl. voids region	0.66	0.71	0.49	0.43

particles seem to be almost the same for all the Fe films. The existence of Fe particles shows that even Fe thin films grown molecular-adsorbed MgO surfaces form three-dimensional islands.

Surface roughness (Ra) of the MgO buffer layer and the Fe films grown on respective MgO surfaces is summarized in Table 1. Here, Ra is defined as an arithmetic average of the absolute values of heights. Ra is expressed as

$$Ra = \frac{1}{x_{\max}y_{\max}} \int_0^{x_{\max}} \int_0^{y_{\max}} |z(x,y) - z_0| dx dy, \quad (1)$$

where $x_{\max}$ and $y_{\max}$ are side lengths of an arbitrary rectangle region, and $z(x,y)$ and z_0 are the height at (x,y) and the average height in the whole region, respectively. Ra of the Fe film grown on non-adsorbed MgO is larger than that of the MgO buffer layer. On the other hand, Ra 's of Fe films grown on CO-adsorbed and H₂O-adsorbed MgO are smaller than that on non-adsorbed MgO. Particularly, Ra 's of both the Fe films estimated by excluding void regions are considerably smaller than those of the MgO buffer layer and of the Fe film on non-adsorbed MgO. This indicates that the surface flatness is improved by molecule-adsorption on the MgO surface.

The results shown above reveal the improvement of the surface flatness of Fe films by the adsorption of CO and H₂O on MgO. A possible mechanism for this effect is considered as follows. Fe islands are three-dimensionally grown on MgO, because the surface energy of Fe is higher than that of MgO. Therefore, the migration of Fe atoms promotes the aggregation of Fe particles. In the case of CO-adsorption and H₂O-adsorption, the migration of Fe atoms is possibly blocked by CO or H₂O molecules. As a result, the formation of rough islands is suppressed, and the surface flatness is improved. In order to elucidate more details on the mechanism, however, all the experiments should be performed *in-situ*, and precisely controllable introduction of H₂O molecules is needed.

4. Summary

The effect of molecule-adsorption on the MgO surface on the growth of ultra-thin Fe films have been investigated. It has been revealed that the adsorption of CO and H₂O on the MgO surface apparently influences the morphology of Fe thin films. Although small Fe particles are grown, the surface flatness of Fe films is improved by the adsorption of CO or H₂O. This result indicates a possibility of the growth of an ultra-thin continuous Fe film with an atomically flat surface by optimizing a suitable condition of the molecule-adsorption.

Fig. 2 AFM images of the surface of (a) a MgO buffer layer and Fe layers with a thickness of 0.3 nm grown on (b) non-adsorbed MgO, (c) CO-adsorbed MgO, (d) H₂O-adsorbed MgO. (e), (f), (g) and (h) are magnified images of (a), (b), (c) and (d), respectively.

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