

Surface Diffusion Coefficients of Adatoms on Strained Overlayers

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Adatom kinetics on the surfaces of Co overlayers, prepared on the W(110) surface, was studied with scanning tunneling microscopy. By counting the number-density of the adatom-islands, we estimated the ratio of adatom diffusion coefficients. The ratio $D_{W(110)} : D_{1ML\ Co} : D_{2ML\ Co}$ was measured to be 1 : 125 : 33000 at room temperature, where $D_{W(110)}$, $D_{1ML\ Co}$, and $D_{2ML\ Co}$ are the diffusion coefficients on bare W(110) surface, on one-monolayer Co overlayer, and on two-monolayers Co overlayers, respectively. An increased diffusion coefficient on two-ML Co overlayers, relative to that on one-ML Co overlayers, was explained with the heteroepitaxial strain effect.

Keywords : Scanning tunneling microscopy, Epitaxy, Nanostructures, Surface structure

I. Introduction

For nano-scale science and electronics, the atomic-level epitaxial growth is an essential technique. Artificial structures, such as superlattices, magnetic alloys, and nano-islands, have been grown with various materials, exhibiting novel electronic and optical property. It was demonstrated, for example, that electrons can be confined into two, one, and zero-dimensional quantum structures, absorbing or emitting selective photons, whose energy levels are controlled by epitaxial growth condition [1–3]. The growth behavior of heteroepitaxial growth is classified into three modes layer-by-layer growth, three-dimensional island growth, and layer-by-layer followed by three-dimensional island growth. This classification is made by thermodynamic energetics, where the surface tension and free energy is minimized. In many cases, however, the growth process is not under thermodynamic equilibrium, thus kinetic factors may

do the leading role.

Surface diffusion is a key factor in atomic-level growth process. It was reported that surface diffusion anisotropy leads to one-dimensional or asymmetric adatom-islands [4], and that increased attempt frequency enhances the surface diffusion of adatoms to move off two-dimensional islands, resulting in layer-by-layer growth [5]. It was suggested there are elastic or electronic effects on the surface diffusion of adatoms [6,7]. On elastically distorted surfaces, for example, the diffusion coefficient of surface adatoms can be modified and controlled by strain. It was reported that the diffusion coefficient of adatoms is layer-dependent, regarding the strained Cu surfaces prepared on the Ru(0001) substrate [6]. On pseudomorphic surfaces, an *ab initio* density functional calculation showed that surface diffusion coefficient increases with a compressive surface strain, while it decreases with a tensile strain [7].

In this paper, we report the ratio of diffusion co-

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efficients of Cr adatoms on Co overlayers, prepared on the W(110) substrate. We observed that the diffusion coefficient of Cr-adatoms on a two-monolayers (ML) Co surface is two orders of magnitude higher than that on a one-ML Co surface. The adatom kinetics will be discussed on the basis of our observation.

II. Experiment

The detailed design of the STM used in this study can be found elsewhere [8]. The W(110) substrate was cleaned by repeated cycles of Ar^+ ion sputtering and annealing. Repeated annealing was performed at 2200 °C with an O_2 pressure of 1×10^{-7} torr and in 1×10^{-10} torr to remove carbon impurity. Co was deposited in the substrate by heating a high purity Co wire (99.99%). Cr was deposited by using a Knudsen cell. The deposition rates of Co and Cr could be controlled within $0.1 \sim 1.1$ ML/min. The deposition rates were estimated with a quartz crystal thickness monitor. The deposition rates were cross-checked with STM

images, with a coverage of less than 1 ML. All the STM images were taken at room temperature with a sample bias voltage of between -2 V and 2 V.

III. Results and discussion

The growth of Co on W(110) has been extensively studied [9–11] with diffraction low energy electron diffraction and STM. At submonolayer coverages, the pseudomorphic Co islands are formed with bcc(110) structure even though there is a significant lattice mismatch. At the coverage around 1 ML, the pseudomorphic structure transforms to a closed-packed structure with hexagonal symmetry. The closed-packed structure shows a Nishiyama–Wassermann orientation ($[\bar{1}\bar{1}00]$ Co || $[\bar{1}\bar{1}0]$ W), as determined by diffraction experiments. As coverage is increased, two-ML and three-ML height islands are formed, with strain relieving dislocations. We performed the experiments in two-step processes. As a first-step, we prepared one- and two-ML Co layers on a W(110) substrate. About 1ML Co was deposited at room-temperature and

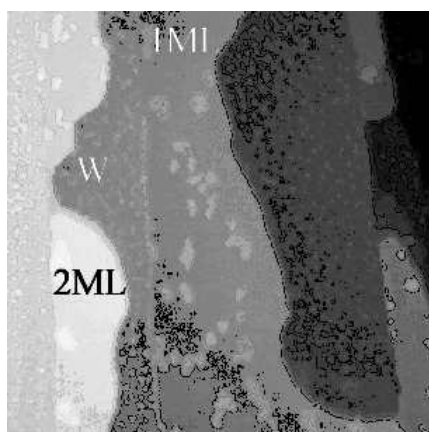


Fig. 1. (a)

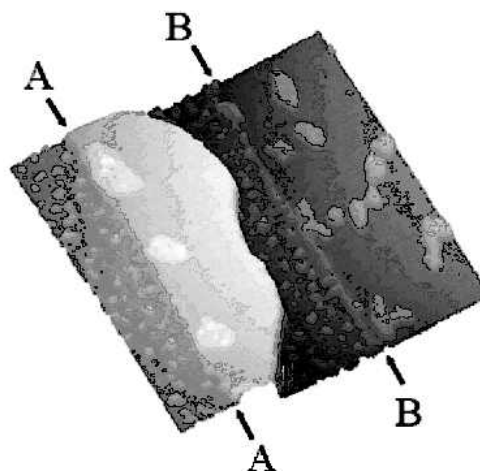


Fig. 1. (b)

Fig. 1. (a) STM image of the surface prepared with two step processes described in the text. Annealing temperature for the Co was 190 °C. The bare W(110) surface, one-monolayer Co region, and two-monolayer Co region, were labeled as W, 1ML, and 2ML, respectively. The image size is $1700 \times 1700 \text{ \AA}^2$. (b) Three dimensional view for the part of (a). The two kinds of boundaries are labeled as "A" and "B".

subsequently annealed at 600 K. We were able to prepare the samples with three kinds of regions, a one-ML Co region, a two-ML Co region, and a bare W(110) region. As a second-step, we deposited 0.1 ML Cr on the surfaces prepared in the first-step. The substrate was held at room temperature when Cr was deposited, and was not annealed afterward. Figure 1 shows a representative STM image of the surfaces prepared with the two-step processes. Cr islands are nucleated on the three regions. The three surface regions are labeled as 1ML, and 2ML, and W in Fig. 1(a), signifying a one-ML Co region, a two-ML Co region, and a bare W region, respectively. A field emission electron microscopy experiment showed that Cr atoms are mobile on the W(110) surface, even at room temperature [13]. Randomly distributed islands are formed on the three regions with a homogeneous nucleation scheme: Newly deposited adatoms randomly diffuse on the surfaces until they meet other mobile adatoms or preexisting two-dimensional islands.

One-ML and two-ML Co regions are easily distinguished by their apparent heights, in Fig. 2(b), a three-dimensional view for the part of Fig. 2(a).

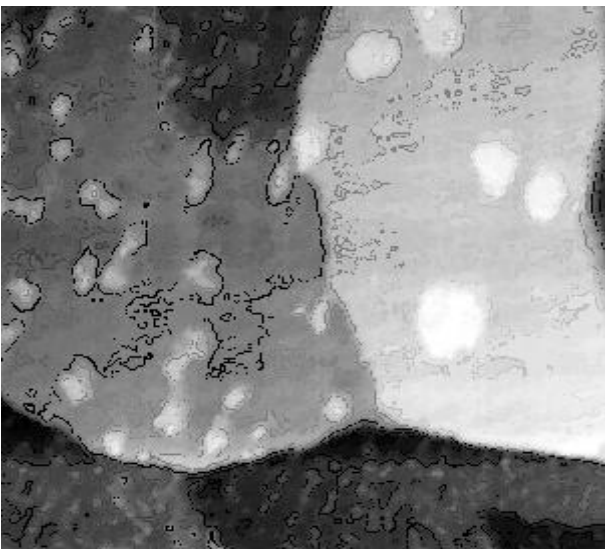


Fig. 2. STM image of the surface prepared with the same method as Fig. 2. The Co coverage for the first step was 1 ML. Image size is $1500 \times 1500 \text{ \AA}^2$.

There are two boundaries A-A and B-B. Both boundaries are formed at the original step edges of bare W(110). Around the boundary A-A, the right region is apparently higher than the left region. The right is a two-ML Co region, while the left is bare W(110). Around the boundary B-B, the right and left regions show apparently similar height. The right is a one-ML Co region and the left is bare W(110). The shapes of the Cr islands at the two boundaries are different. After the growth of Cr, the boundary A-A is still smooth, while the boundary B-B has small Cr islands forming chain-like structure.

In Fig. 1(a), the number density of the Cr islands on a two-ML Co are smaller than that on a one-ML Co, which is again much smaller than that on bare W. The same behavior is clearly visible in Fig. 2, with a larger two-ML Co than in Fig. 1. The number densities were counted to be 2 ± 1 , 10 ± 2 , 64 ± 10 per 10^4 \AA^2 on a bare W surface, on a one-ML Co, and on a two-ML Co, respectively. It is known that there is a power-law dependence between the island number density and the diffusion coefficient of adatoms. In a two-dimensional random walk model without diffusion anisotropy, the number density of an island is proportional to D^{-i+1} , where D is the diffusion coefficient of adatoms and i is the critical island size [14]. In various metal surfaces the critical island size is reported to be two, meaning dimer nucleation [4]. Assuming dimer nucleation on these three surfaces, the ratio $D_{W(110)} : D_{1ML Co} : D_{2ML Co}$ is estimated to be $1 \pm 1 : 125 \pm 8 : 33000 \pm 1000$, where $D_{W(110)}$, $D_{1ML Co}$, and $D_{2ML Co}$ are diffusion coefficients of Cr adatoms on the bare W(110) surface, on a one-ML Co and on a two-ML Co, respectively. This estimation holds at room temperature. It is noted that some islands are nucleated even on the Cr islands, which are on a two-ML Co (Fig. 3). The number density looks higher than 2 per 100 \AA^2 . But, it is too small number to correlate with these diffusion coefficients.

It is inappropriate to compare the diffusion co-

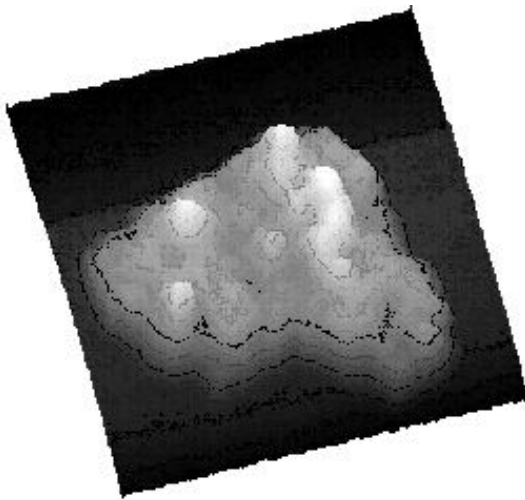


Fig. 3. STM image of Cr islands on the two-monolayer Co region. Image size is $150 \times 150 \text{ \AA}^2$.

efficients on bare W(110) with those on the Co overlayers. Diffusion coefficients vary, depending on surface elements and atomic structures. However, it is still possible to compare the diffusion coefficient on one-ML Co region and that on two-ML Co region. There must be some elastic or electronic effect on the overlayer surfaces. Similar effects were reported for the Ni surfaces prepared on an Ru(0001) substrate [6]. On the basis of *ab initio* density functional calculation, it was reported that diffusion coefficient increases with compressive strain and decrease with tensile strain, on pseudomorphic surfaces [7]. There have been experimental reports that indicate the diffusion coefficient of adatoms on a Ag overlayer grown on Pt(111) substrate, is higher than that on Ag(111) surface, due to the compressive strain on the Ag overlayer surface [15]. This behavior regards to pseudomorphic surfaces, but we are tempted to apply the idea to the present system. It is known that submonolayer Co may be pseudomorphic or closed-packed. They can be distinguished by an apparent height difference of $\sim 0.5 \text{ \AA}$ [11]. In our experiment, more than 90 % of the one-ML regions turned out to be closed-packed. The closed-packed layers show a Nishiyama-Wassermann orientation.

On a W(110) surface, the lattice mismatches are -3.2% in $[\bar{1}10]$ and -21% in $[001]$. The lattice constants of Co are smaller than those of W in both directions. Thus, Co atoms try to form relaxed structures, while W atoms try to stretch the Co atoms, resulting in tensile strain to the Co atoms. It is known that the Co overlayer will follow Stranski-Krastanov growth mode. As the thickness of Co increases up to the critical thickness, the tensile strain on the flat overlayer increases. Beyond the critical thickness of $> 5 \text{ ML}$, the tensile strain is relieved by three-dimensional islands and other dislocation structures. Since one- and two-ML are below the critical thickness, they have tensile strain. It was reported that the tensile strain relaxes to a certain extent, and that the two-ML is less strained than the one-ML [11]. Since the tensile strain is stronger on the one-ML Co than on the two-ML Co, the diffusion coefficient is lower on the one-ML Co.

There may be additional effects from the substrate. Atomic configuration of one-ML Co can be quite different from that of two-ML Co. Co atoms in one-ML Co should bind with W atoms, while the two-ML Co does not have any W atoms as its nearest neighbors. For example, the charge transfer between the overlayer and substrate will be largest at one-ML Co, possibly affecting the diffusion coefficient of adatoms.

IV. Conclusion

In conclusion, from the observed island distribution in STM images, we measured the ratio $D_{W(110)} : D_{1ML Co} : D_{2ML Co}$ to be $1 : 125 : 33000$, where $D_{W(110)}$, $D_{1ML Co}$, and $D_{2ML Co}$ are diffusion coefficients of Cr adatoms on the bare W(110) surface, on a one-ML Co and on a two-ML Co, respectively. An increased diffusion coefficient on a two-ML Co, relative to that on a one-ML, can be understood with strain effect.

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스트레인을 받고 있는 표면에서의 원자 확산계수

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W(110) 표면에 성장한 Co 박막에서 원자 정역학을 주사터널링 현미경으로 연구했다. 원자섬의 개수 밀도를 측정하여 원자 확산 계수의 비를 알 수 있었다. W(110) 표면, Co가 1 원자층 성장된 표면, Co가 2 원자층 성장된 표면의 원자 확산 계수의 비는 상온에서 1 : 124 : 33000인 것으로 측정되었다. Co가 2 원자층 성장된 표면의 확산 계수가 Co가 1 원자층 성장된 표면의 확산 계수보다 큰 것은 이중성장의 스트레인 효과로 인한 것으로 해석되었다.

주제어: 주사터널링 현미경, 에피성장, 나노구조, 표면구조

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