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Growth of honeycomb-symmetrical Mn nanodots arrays on Si(111)- 7×7 surfaces

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Abstract

The growth of well-ordered Mn nanodots arrays on Si(111)- 7×7 reconstructed surfaces was investigated by means of scanning tunnelling microscopy (STM) as well as kinetic Monte Carlo (KMC) simulation. Mn atoms deposited slowly onto elevated substrates were observed to occupy preferentially on the faulted half unit cells (FHUCs) of the Si(111)- 7×7 surface. The preference occupancy in the FHUCs, P_F , defined as the ratio of the number of FHUCs occupied by Mn nanodots to the number of all occupied in the two halves, decreases with increasing deposition rate as well as decreasing substrate temperature. The KMC simulations, which are in good agreement with the experimental results, were employed to optimize the growth conditions, including deposition rate and substrate temperature, for the self-organized growth of Mn nanodots arrays on Si(111)- 7×7 reconstructed surfaces. By adjusting the deposition rate, one can control the growth of well-ordered and uniform Mn nanodots arrays to form either a triangular symmetry or a honeycomb one.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

It is well known that the nanoscaled materials can exhibit significantly different physical and chemical properties from those of corresponding bulk materials due to the quantum size effects. Some properties of the nanodots can alter drastically even when the cluster size varies by a single atom, and the inhomogeneity in an ensemble of clusters will smear their special properties [1]. Therefore, artificially controllable fabrication of nanostructures on supporting substrates with atomic precision is the ultimate goal in fundamental and applied

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science research [2–6]. Recently, identical indium, gallium, aluminium and cobalt nanodots have been fabricated by self-assembly on Si(111)- 7×7 reconstructed surfaces [7–10]. The Si(111)- 7×7 reconstructed surface consists of two different types of triangular half-unit cell, faulted half-unit cells (FHUCs) and unfaulted half-unit cells (UFHUCs), which are divided by dimer rows and corner holes. Metal nanodots can occupy either of the two halves of unit cells (HUCs) of Si(111)- 7×7 reconstructed surfaces equally to form a honeycomb symmetry, or the FHUCs only with a triangular symmetry. In our previous work, well-ordered and uniform Mn nanodots were fabricated on a Si(111)- 7×7 surface [11]. Due to the relatively large energy difference between adsorption on the UFHUCs and the FHUCs, Mn nanodots deposited onto the elevated substrates were observed to occupy preferentially on the FHUCs of a Si(111)- 7×7 surface to form a triangular symmetry, and the honeycomb structure was not formed even at high coverages. In order to increase the density of Mn nanodots and modify the interdot distance, which may be helpful for investigating the dot–dot interaction, we present the controllable growth of uniform Mn nanodots arrays on Si(111)- 7×7 surfaces from triangular symmetry to honeycomb symmetry. Furthermore, kinetic Monte Carlo (KMC) simulation was employed to optimize the self-organized growth conditions for Mn nanodot arrays on Si(111)- 7×7 reconstructed surfaces.

2. Experimental details

Our experiments were performed with a combined molecular beam epitaxy (MBE)/scanning tunnelling microscope system in ultrahigh vacuum (UHV) at a base pressure of about 1×10^{-10} mbar. A Si(111) substrate was cleaned by resistive flashing in UHV until a high quality 7×7 reconstruction was observed by an OMICRON variable temperature STM. High purity Mn (purity 99.99%) was heated at temperatures ranging from 923 to 1053 K in a boron nitride crucible, and then deposited onto the surface with deposition rates ranging from 0.17 monolayer (ML) min^{-1} ($1 \text{ ML} = 7.88 \times 10^{14} \text{ atoms cm}^{-2}$) to 13.4 ML min^{-1} . All room-temperature scanning tunnelling microscopy (STM) images reported here were recorded with tunnelling current ranging from 200 to 400 pA. A chemically etched tungsten tip was used as the scanning tunnelling microscope probe.

3. Results and discussion

Since the FHUCs of the Si(111)- 7×7 reconstructed surface are more reactive than the UFHUCs and the intrinsic attractive potential wells on the FHUCs effectively trap diffusing metal atoms [12], Mn atoms are expected to occupy preferentially on the FHUCs. In order to control Mn atoms to occupy equally on the FHUCs and UFHUCs and form the honeycomb structure, one must restrict them to diffuse into the FHUCs. The atomic diffusion can be significantly reduced when they are deposited onto a cooled substrate. Figure 1(a) is the STM images of Mn nanodots deposited at 145 K with the coverage of 0.21 ML. Although Mn nanodots distribute randomly on both the FHUCs and UFHUCs of the Si(111)- 7×7 surface, the diameter and height distributions are very broad (figures 1(b) and (c)). The dispersions, $\Delta d = (\langle d^2 \rangle - \langle d \rangle^2)^{1/2}$ and $\Delta h = (\langle h^2 \rangle - \langle h \rangle^2)^{1/2}$ are 0.31 and 0.028 nm, respectively, which are about 23.9% and 12.4% of the average diameter and height values. The broad size distribution of Mn nanodots is undesirable for future applications.

The size distribution of Mn nanodots can be significantly narrowed when the substrate is heated up to 450 K. Figures 2(a)–(c) show the STM images of Mn nanodots deposited onto the Si(111) substrate heated to 450 K with three different deposition rates: 0.17, 0.64 and

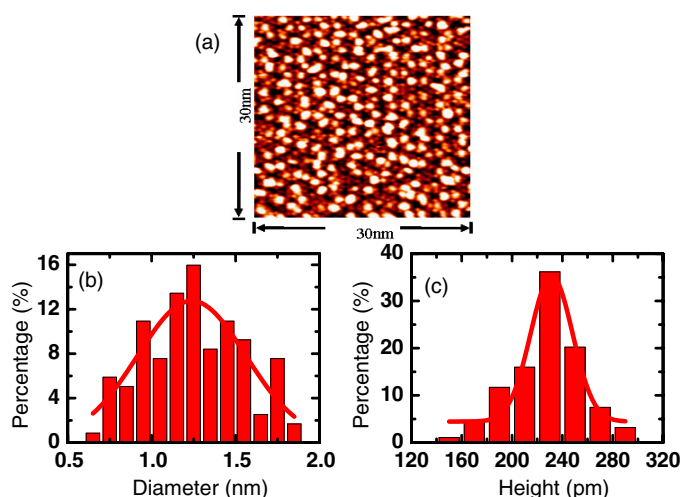


Figure 1. STM images of Mn dots deposited at temperatures of about 145 K with the coverage of 0.21 ML (a) and the corresponding histograms of diameter (b) and height (c). The STM image was obtained at $V_s = -2.093$ V and with a scanned area of 30×30 nm².

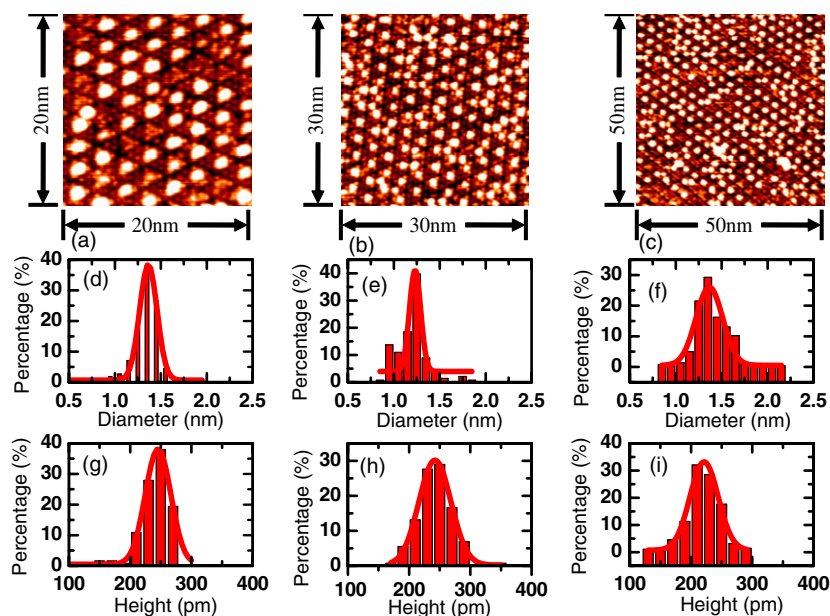


Figure 2. (a)–(c) are STM images of Mn nanoclusters deposited on the Si(111)- 7×7 surface at about 450 K with three different deposition rates: 0.17, 0.64 and 1.8 ML/min, respectively. The coverage of the Mn dots in (a)–(c) is 0.21, 0.18 and 0.18 ML, respectively. Images were obtained with the sample bias of -2.2 V and tunnelling current of 0.25 nA. Below the STM images are the corresponding histograms of cluster diameter and height, (d)–(i), respectively.

1.8 ML min⁻¹, respectively. The coverage of the Mn dots in figures 2(a)–(c) is 0.21, 0.18 and 0.18 ML, respectively. The diameter and height histograms of the nanodots are illustrated in figures 3(d)–(i). The dispersions, Δd and Δh , are found to decrease to about 10% of the

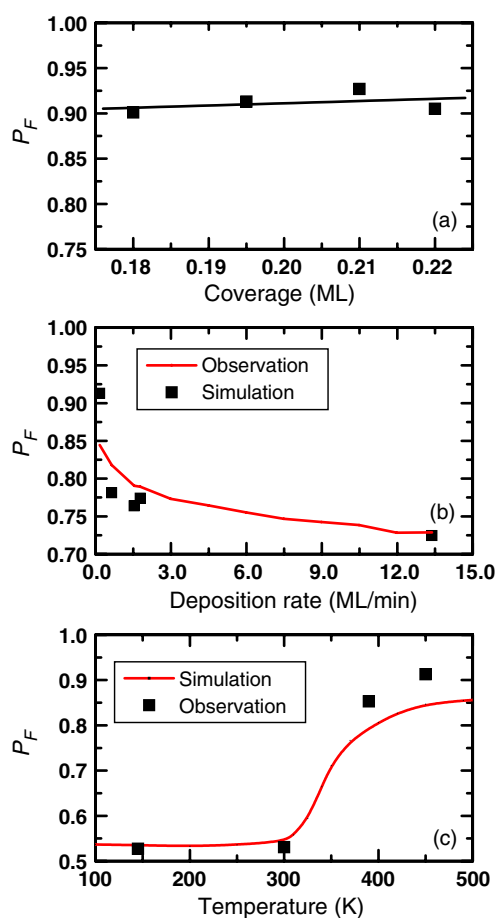


Figure 3. Preference factor P_F as a function of coverage for Mn nanodots deposited onto the substrate at a temperature of 450 K (a), deposition rates (b), and substrate temperatures (c).

average diameter and height values when the substrate temperature increases up to 450 K. With increasing deposition rate, the size and shape of Mn nanodots remain essentially unchanged. At elevated substrate temperature, the deviation in size distribution can be eliminated by atoms hopping quickly in half unit cells (HUCs). In the case of Mn nanodots on Si(111)- 7×7 surfaces, the attractive potential wells formed by the triangular HUCs (2.7 nm) restrict Mn nanodots to overgrow the HUC boundaries at low coverage, and consequently result in the narrow size distribution.

Figure 3(a) illustrates the preference in the FHUCs' occupancy, P_F , (ratio of the number of FHUCs occupied by Mn nanodots to the number of all occupied FHUCs in the two halves) as a function of coverage. Mn atoms occupy the FHUCs, which is energetically favourable in the case of low coverage. It is surprising that P_F is almost invariable with increasing coverage even up to 0.22 ML. With further increasing the coverage, Mn nanodots will aggregate into big islands across the HUC boundaries. Since the preference P_F is kinetically determined by the ratio between hopping rate and deposition rate, the maximum value of P_F can be achieved when the Mn adatoms have enough time for rearranging on the surface during growth. When the deposition rate is too high, they form sizeable clusters prior to diffusing into the more stable

position. Therefore, P_F is expected to decrease with increasing the deposition rate, as plotted in figure 3(b).

Detailed behaviours for atoms on surfaces are highly desirable to understand the underlying formation mechanism of these nanodots. However, the *in situ* observation of the atoms' behaviours is very difficult at current experimental conditions. A deeper understanding of the kinetics of the dots' growth on Si(111) surfaces has been obtained with the help of kinetic Monte Carlo (KMC) simulations. The coarse grained KMC model implements have been described in detail elsewhere [13, 14]. Mn atoms are assumed to fall initially in the two half-unit cells (HUCs) randomly. Once an atom falls into an HUC, it immediately moves to its adjacent occupied HUCs if they exist. In this model, an isolated cluster cannot overgrow HUC boundaries, and the capacity of an HUC to accommodate atoms cannot exceed n_s . When the number of atoms in a cluster $n \leq n^*$ (n^* is the critical size), the cluster can decay by atoms hopping out one by one. The activation energy for one atom hopping out of an HUC containing n atoms is given by $E_n = E_{F,U} + (n - 1)E_a$, where $E_{F,U}$ is the barrier for a single Mn atom hopping out of FHUCs and UFHUCs, respectively, and E_a is the effective binding energy between adatoms inside HUCs. The attempt frequency ν_0 is assumed to be the same for the FHUCs and UFHUCs, and attempts by Mn atoms to hop out of an HUC are considered to be independent, so that the hopping rate of a Mn atom can be written as $\nu_n^{F,U} = n\nu_0^{F,U} \exp(-E_n/kT)$. Our simulation consists of growth at 450 K and a subsequent annealing process at room temperature for an hour. By fixing $\nu_0^{F,U} = 5 \times 10^9 \text{ s}^{-1}$, $n^* = 5$, $n_s = 21$ [14] and changing E_F , E_U , E_a , randomly, we found a least square relationship between simulation and experimental data points. The simulated P_F as a function of deposition rate is shown in figure 3(b). The model parameters which provide the best agreement with experimental data are $E_F = 0.72 \text{ eV}$, $E_U = 0.70 \text{ eV}$, and $E_a = 0.05 \text{ eV}$. The values of E_F , E_U , and E_a are very close to those of Ag islands on Si(111)-7 × 7 surfaces [13]. The low value of E_a indicates a weak interaction of Mn atoms inside an HUC. Theoretical calculations showed that there existed three non-intersecting closed diffusion paths for metal adatoms inside a Si(111)-7 × 7 HUC so that metal adatoms can move independently with minimal interaction [15]. The (observed) qualitative agreement suggests that the model correctly captures the basic features of the growth process for Mn nanodots on Si(111)-7 × 7 surfaces. By fixing the deposition rate to 0.17 ML min^{-1} , the effect of substrate temperature on the preference P_F was also observed by STM and KMC simulation. As is shown in figure 3(c), both KMC simulation and STM image observation demonstrate consistently that a weak preference of Mn nanodots in the FHUCs can be obtained when the substrate is cooled to low temperatures. Therefore, the KMC simulations were employed to optimize the growth conditions, including deposition rates and substrate temperatures, for the self-organized growth of Mn nanodots arrays on Si(111)-7 × 7 reconstructed surfaces, illustrated in figure 4. This reveals that the maximum value of preference in the FHUC occupancy, P_F , can be achieved when Mn is deposited onto an elevated substrate with very low deposition rate. The preference can be weakened either by decreasing the substrate temperature or by increasing the deposition rate and keeping the substrate temperature at a certain temperature.

Figures 5(a) and (b) illustrate two typical STM images of Mn nanodots with deposition rates of 0.17 ML/min and 13.4 ML/min , respectively. The coverages of the Mn dots in figures 5(a) and (b) are 0.21 ML , and 0.41 ML , respectively. A triangular symmetry of the Mn nanodot array is observed when the deposition rate is very low. On increasing the deposition rate up to 13.4 ML/min , the triangular symmetry of Mn nanodots transforms to a honeycomb structure due to Mn occupying the two halves of the unit cells of the Si(111)-7 × 7 reconstructed surface equally. Therefore, the controllable growth of well-ordered and uniform Mn nanodot arrays with a honeycomb symmetry can be achieved on Si(111)-7 × 7 surfaces.

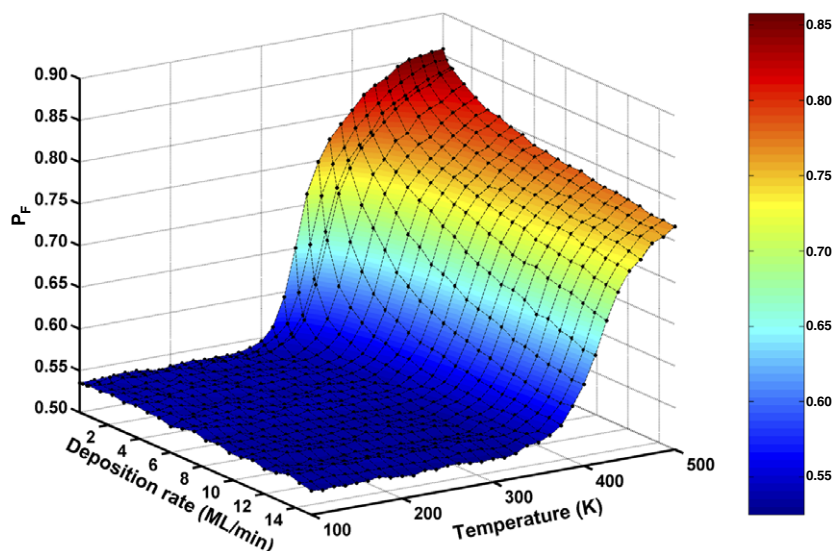


Figure 4. Simulated diagram of growth conditions, including deposition rates and substrate temperatures, for the self-organized growth of Mn nanodot arrays on a Si(111)- 7×7 reconstructed surface.

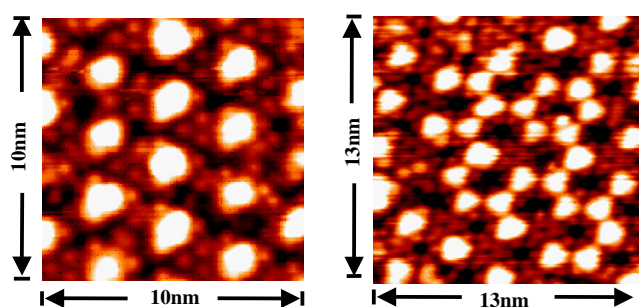


Figure 5. Typical STM images of Mn nanodots with deposition rates of 0.17 ML/min (a) and 13.4 ML/min (b). The coverage is 0.21 ML (a) and 0.41 ML (b). The images were obtained with a sample bias of -2.5 V and tunnelling current of 0.25 nA.

4. Summary

In summary, both STM observation and KMC simulation indicate that the reduction of preference in the FHUC occupancy, P_F , can be achieved either by decreasing the substrate temperature or by increasing the deposition rate and keeping the substrate temperature at a certain temperature. However, the diameter and height of Mn nanodots deposited onto a cooled substrate show a very broad distribution. Due to the attractive potential wells formed by the triangular HUCs (2.7 nm), which restrict the overgrowth at HUCs boundaries, uniform Mn nanodot arrays can be ordered a honeycomb structure on Si(111)- 7×7 surfaces with a substrate temperature of 450 K by increasing the deposition rates.

Acknowledgments

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