Film thickness dependence on morphology of Fe films on self-organized SrTiO$_3$ (001) substrates with inclined angles

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Abstract

The morphologies of Fe films between 1 and 10 nm thick grown on inclined SrTiO$_3$ (001) substrates were investigated by scanning tunneling microscopy. Three-dimensional Fe clusters were formed in all the films and the shape of the clusters changed with the film thickness. As the film thickness increased, the average cluster height and size obtained from their distributions increased, but the surface roughness decreased. The values of the average cluster size and height, and the surface roughness in the films with different film thickness clarified the growth of the Fe films. In the 1 nm-thick Fe film, isolated clusters were observed, but when the film is greater than 3 nm thick, the films consisted of two parts. The lower part was a continuous film and the upper part was a cluster coagulation.

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1. Introduction

The magnetic properties of nanostructures grown on inclined substrates are of great interest for their applications such as high-density storage devices. Proper substrates are required to fabricate nanostructures and investigate their magnetic properties. The inclined SrTiO$_3$ (001) substrates made by the self-organization phenomena have been clarified as an ideal template to fabricate regularly arranged magnetic nanostructures [1,2]. Moreover, the dependence of the inclined angle on the morphology and magnetic properties of a 5 nm-thick Fe film grown on the inclined SrTiO$_3$ (001) substrates were reported in our previous papers [2,3].

Films grown on inclined substrates, have surface features that are significantly treated because stepped structures with many dangle bonds exist. Some studies have researched the relationship between the surface energy and the magnetic properties [4–9]. It has been reported that the surface energy induced by the step density influences the magnetic anisotropy of the magnetic film. In a thinner film on an inclined substrate, the effect of the step structure is also expected to be more significant in terms of their morphologies and magnetic properties. Thus, it is worthwhile to research thickness dependence on the morphology and to find the transition region between the ferromagnetic state and the superparamagnetic one. According to the changes in the film thickness, the crystalline structure and the morphology are also affected by the step structures on the inclined substrates. Since the structure and the morphology play important roles in determining the magnetic properties of thin films, a careful investigation of the morphology in the magnetic film is necessary.

In this study, the film thickness dependence on the morphology of Fe films on the inclined SrTiO$_3$ substrate was investigated by scanning tunneling microscopy (STM). The growth behavior of Fe films on the stepped surface as a function of film thickness was also examined.

2. Experimental procedure

Single-crystallized SrTiO$_3$ (001) substrates were cut at nominal inclined angles ($\alpha$) of $2^\circ$ and its inclined direction was in the [100] direction. Since an inclined substrate with $\alpha = 2^\circ$ is expected to form relatively wide terrace widths on
the surface, it allows the growth procedure of Fe films grown on the inclined substrate to be observed.

The inclined SrTiO₃ substrates were ultrasonically cleaned in an ethanol and acetone solution before the annealing procedure. In order to form the self-organized step structures on the surface, the substrates were annealed for 1 h at 1273 K in O₂ with the flow rate of 1 l/min. The annealed substrates were moved into a UHV chamber and were annealed for 0.5 h at 1073 K in a UHV chamber below 1 × 10⁻⁶ Pa to achieve surface conductivity. These conditions were similar to previous studies using the self-organized SrTiO₃ substrates [2,3].

Fe films were grown at 473 K on the inclined SrTiO₃ (001) substrates in a vacuum below 7 × 10⁻⁸ Pa by molecular beam epitaxy (MBE) using a VG Semicon V-80M multichamber MBE system. Four hundred and seventy three Kelvin was chosen as an annealing temperature to induce the appropriate surface diffusion of Fe atom on the substrate. The Fe films were 1, 3, 5 and 10 nm thick. The deposition time while maintaining a constant deposition rate of 0.5 ML/min (1 ML = 0.1433 nm), determined the thickness of each film. A quartz film thickness monitor attached in the MBE system controlled the Fe deposition rate and the deposition rate was clarified by analyzing the X-ray diffraction profiles in a [Fe/Au]₁₀ multilayer prepared for calibrating Fe film thickness. The reflection high-energy electron diffraction (RHEED) continuously observed the structure of the Fe film during the deposition process and the RHEED observations determined the crystalline orientation relationship between Fe film and SrTiO₃ substrate.

The surfaces of the self-organized SrTiO₃ substrates and the Fe films were observed by STM in UHV. A tip for the STM observation was made by electro-polishing a W wire and then its surface was cleaned by Ar⁺ sputtering. The STM observations were performed in the constant current mode, with a tip bias of 3.3–4.3 V for a substrate and 0.8–1.0 V for Fe film. The tunneling current was 7.3–10.7 nA.

3. Results and discussion
3.1. Crystal growth of Fe film on the inclined SrTiO₃ substrate

Fig. 1(a) shows the RHEED pattern and its schematic diagram of SrTiO₃ substrate with α = 2° and Fig. 1(b) shows those of the 5 nm-thick Fe film grown on the substrate. In the substrate, (001)SrTiO₃ streaks appear, which implies that the SrTiO₃ (001) surface of the substrate is flat. On the other hand, in the Fe film, spots and streaks are observed and the spots coincide with the streaks, which indicates that the Fe film consists of cluster structures formed on the substrate. Furthermore, each streak in the pattern of the substrate is inclined by 2° due to its rotated-reciprocal lattice [10]. The inclined patterns are also observed in the RHEED patterns of the Fe film, which means that the Fe film is grown with the inclined surface. These features are also observed when Fe films are grown on inclined substrates with different inclined angles, 4 and 6° [2]. Analyzing the patterns in Fig. 1(a) and (b) indicates that the crystalline orientational relationship between SrTiO₃ and Fe film is Fe(001)[110]/SrTiO₃(001)[100].

3.2. Morphology of Fe film with different film thickness

Fig. 2 shows STM images of Fe films with film thickness t of 1, 3, 5, and 10 nm corresponding to Fig. 2(a)–(d), respectively. The upper right inset of each figure is a high-magnified image. Three-dimensional Fe clusters are observed in all the films. The Fe clusters are in the [010] direction and maintain the shape of the straight step edge of the substrate for all the films. Details of such surface features have been reported in previous papers [2,3]. In the high-magnified image, the shape of Fe clusters is clearly distinguished: a circular shape for t = 1 nm, circular, square and rectangular shapes for t = 3 nm, square and rectangular shapes for t = 5 nm, and an oblate circular shape for t = 10 nm. It is certain that the change in shape of the Fe cluster depends on film thickness. In respect of the continuity between the Fe clusters, the clusters start to connect with one another in the 3 nm-thick film and the connection between the Fe clusters causes the coexistence of the circular, square, and rectangular clusters. Especially, in the 5 nm-thick Fe film, square clusters are rotated 45° with respect to a lattice structure of the SrTiO₃ (001) substrate, which coincides with the result of RHEED observation shown in Fig. 1.

The change in the strain between Fe and SrTiO₃ can explain the shape of the Fe clusters. Coherent islands are formed in a thinner film such as a 1 nm-thick film and the strain induced by the misfit (3.9%) between Fe and SrTiO₃ causes circular Fe clusters. When the thickness of the Fe film increases, the introduction of the dislocation into the interface between Fe and SrTiO₃ causes the interface becomes incoherent. The strain energy is released in
the interface. Thus, the shape of interface becomes flat and the shape of the Fe clusters changes from circular to square. In a thicker film, adjacent clusters connect and the flat interface in the Fe clusters vanishes. These results are consistent with the results previously reported for a 3.6 nm-thick Fe films grown on the MgO (001) substrate [11–13].

3.3. Dependence of surface feature on film thickness

Fig. 3(a)–(d) shows the distributions of the Fe cluster size obtained from STM images and correspond to a film thickness, \( t \), of 1, 3, 5 and 10 nm, respectively. The shape of the clusters must be ignored in order to compare the cluster size according to film thickness since the shape of

![Fig. 2. STM images of Fe (t nm)/SrTiO₃ (001) substrate with inclined angle of 2°. Film thickness \( t \) is (a) 1 nm, (b) 3 nm, (c) 5 nm and (d) 10 nm. The upper right insets of each figure show high magnification images.](image1)

![Fig. 3. Distributions of the cluster size for the Fe film with a thickness of (a) 1 nm, (b) 3 nm, (c) 5 nm and (d) 10 nm. The inset of (a) shows the method used to measure cluster size.](image2)
the clusters is different for every film thickness, as shown in Fig. 2(a)–(d). The cluster size, $S$, in this paper is determined by the average value of two lengths from the parallel direction $x$ and the perpendicular direction $y$ as shown in the inset of Fig. 3(a). As the film thickness increases, the ranges of size distributions are 6 (1–7), 14 (3–17), 12 (5–17) and 8 (7–15) nm for $t = 1, 3, 5$ and 10 nm, respectively. It is noteworthy that $t = 3$ nm has the broadest range, which implies that the Fe clusters are actively connecting in the 3 nm-thick film and is consistent with the STM result that show clusters with various shapes. The values of average cluster sizes obtained from those distributions are 3.8, 8.4, 11.5 and 10.8 nm for $t = 1, 3, 5$ and 10 nm, respectively.

Fig. 4(a)–(d) shows the distributions of the Fe cluster height obtained from STM images corresponding to film thickness, $t$, of 1, 3, 5 and 10 nm, respectively. The ranges of the distributions are 1.6 (0.5–2.1), 2.2 (0.7–2.9), 1.2 (1.7–2.9) and 2.0 (2.7–4.7) nm for $t = 1, 3, 5$ and 10 nm, respectively. The broadest range for the height also appears in the 3 nm-thick film, which indicates that the clusters have various heights at this thickness and may be due to the active connections between the clusters in the 3 nm-thick film. The average cluster heights are 1.1, 1.3, 2.3 and 3.6 nm for $t = 1, 3, 5$ and 10 nm, respectively. The average cluster height increases as the film thickness increases.

In order to investigate the overall features of each Fe film, Fig. 5 summarizes the average values of the cluster size and the cluster height obtained from each distribution. The average cluster size monotonically increases up to $t = 5$ nm and then slightly decreases for $t = 10$ nm. It is believed that the connecting and coarsening between the Fe clusters mainly occurs within a certain terrace of step until the thickness reaches $t = 5$ nm. Then as the thickness further increases the coarsening of the Fe clusters across to the step edge allows the clusters to freely connect with clusters on the lethal step. For the cluster height, the average value increases with film thickness. Comparing the average values of the heights and the deposited thicknesses, differences are observed except for $t = 1$ nm. The differences are 1.7, 2.7, and 6.4 nm for $t = 3, 5$, and 10 nm, respectively. These differences also increase with film thickness, which implies that increasing the film thickness enhances the connectivity of the Fe clusters. It is believed that Fe films above 3 nm thick consist of two

![Fig. 4. Distributions of the cluster height of the Fe film with film thickness of (a) 1 nm, (b) 3 nm, (c) 5 nm and (d) 10 nm.](image)

![Fig. 5. Thickness dependence on the average cluster size and height for the Fe films. The average values are obtained from distributions of the cluster size and height.](image)
parts: a continuous film in the lower part and cluster coagulation in the upper part. Thus, above a thickness of 3 nm, the size and height of the clusters in these films is from the upper part. In addition, when the height, size, and shape of the clusters are considered, the average volumes can be simply calculated and are 8.3, 45.3, 80.1 and 220.0 nm³ for $t = 1, 3, 5$ and 10 nm, respectively.

In order to investigate the details of the connected states of the Fe clusters, the surface roughness was measured from STM images. Fig. 6 presents the roughness of Fe films with the different film thicknesses. The value of the roughness steeply decreases up to $t = 3$ nm and then gradually decreases upon further increasing the film thickness. It is emphasized that the clusters are isolated from each other in the 1 nm-thick Fe film since the value of roughness is the same as the thickness, 1 nm. Unlike the average cluster height, which increases with thickness, the surface roughness decreases as the film thickness increases since the cluster height is determined by the length between the fixed lowest point and the highest point of each cluster.

Fig. 7(a)–(d) shows schematic diagrams of the growth procedures as the film thickness increases. The inset depicts the connected state between the clusters and shows that the 3, 5, and 10 nm thick Fe films consist of two parts. For $t = 1$ nm, isolated Fe clusters are formed, which have a circular shape when the Fe clusters are observed in the [00-1]SrTiO$_3$ direction. For $t = 3$ nm, the clusters connect and have various shapes: circular, square, and rectangular. For $t = 5$ nm, the connections occur between the clusters on the lethal steps and the Fe clusters have the square and rectangular shape. For $t = 10$ nm, the Fe clusters are completely connected and the clusters have an oblate circular shapes. The average volume of the clusters, which is derived from the size and height of the clusters, increases with the inclined angle.

The morphology of the Fe films is strongly dependent on their film thickness. The change of morphology is expected to influence the magnetic properties of the Fe film. Thus, when morphology of Fe film is adequately controlled by factors such as the film thickness, the growth temperature, the growth rate, etc. it is possible to achieve the desired magnetic properties.

![Fig. 6. Change in the surface roughness of Fe films as a function of film thickness.](image)

![Fig. 7. Schematic representation of the growth in the Fe films with film thickness of (a) 1 nm, (b) 3 nm, (c) 5 nm and (d) 10 nm. The heights of the lower parts are also presented in (b)–(d). Inset of each figure shows a cross sectional view of the Fe film.](image)
4. Conclusion

Film thickness dependence on morphology of Fe film grown on the inclined SrTiO$_3$ (100) substrate has been investigated by STM observation.

(1) Three-dimensional clusters are formed for the Fe films with a film thickness between 1 and 10 nm and the clusters are aligned in parallel to the step edge on the inclined SrTiO$_3$ (001) substrate.

(2) The shape of the Fe clusters changed with the film thickness: a circular shape for $t = 1\, \text{nm}$, circular, square, and rectangular shapes for $t = 3\, \text{nm}$, square and rectangular shapes for $t = 5\, \text{nm}$, and oblate circles for $t = 10\, \text{nm}$.

(3) The average Fe cluster size increases, and the average Fe cluster height decreases as the film thickness increases. The average volume calculated from the size and height of the clusters increases with thickness. The widest distribution ranges for the size and height appeared in the 3 nm-thick film. The surface roughness decreases as the film thickness increases and shows the steepest decrease for the 3 nm-thick film. The coarsening and the connecting between the Fe clusters actively occur in the 3 nm-thick film. Fe films thicker than 3 nm consist of two parts: a continuous film in the lower part and cluster coagulation in the upper part.

(4) The morphology of the Fe film less than 3 nm is governed by the strain induced at the interface between the Fe film and the SrTiO$_3$ substrate and films greater than 5 nm thick, the connections between Fe clusters and the coarsening process govern the surface features.

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