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Thesis/dissertation Title

Growth and magnetism of epitaxial Fe nanofilms on ammonia-terminated and three-dimensionally structuralized Si{111}

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Chapter 1

In this study, the growth mechanism and magnetic properties of Fe on the molecules modified planar and three-dimensional (3D) architecture Si substrates were observe by in-situ reflection high-energy electron diffraction (RHEED) in ultra-high vacuum and by ex-situ vibration sample magnetometer (VSM) at room temperature (RT). The main results and discussion in this dissertation are constructed in to four parts, the brief content is shown in the following: Chapter 1 describes the general background and motivation of the dissertation. Chapter 2: Fe growth on clean and ammonia saturated Si(111)7×7 surface studied by in situ electron diffraction. Chapter 3: Magnetic property of Fe film on clean and ammonia saturated Si(111)7×7 surface. Chapter 4: Surface investigation of Fe thin film on the 3D Si pyramid using in situ RHEED. Chapter 5: Magnetic property of Fe thin film on the 3D Si{111}. Last chapter, Chapter 6, covered the dissertation summary and the future suggestion for further development. To the academic development of topological magnetic vortex in nano scale, these findings will lead to the development of practically possible technological innovation. It will have extremely high industrial effect including the realization of carbon-neutral society.

Chapter 2

I have investigated the growth of Fe(111) islands on clean and NH_3-saturated Si(111)7×7 surfaces by Fe deposition at thickness Θ_{Fe} = 1-10 ML at RT using RHEED. By analyzing the transmission diffraction spots of the Fe islands, we estimated the average horizontal and vertical crystalline domain sizes, the crystalline domain volume, and the relative number of crystalline domains from the spot intensity and FWHM. Fe diffraction spots appeared from 3 ML deposition on the clean surfaces and 5 ML deposition on the NH3-saturated surfaces; the rate of increase in spot intensity on the former was higher than that on the latter, indicating the easier formation of Fe crystalline islands on the clean surfaces. The horizontal domain size on the clean surfaces decreased from ~12 Å at ΘFe = 3 ML to ~9-10 Å at 10 ML. In contrast, that on the NH3-saturated surfaces increased from ~11 Å at 5 ML to ~12-13 Å at 10 ML. The vertical domain size on both the clean and NH3-saturated surfaces was almost constant at 3-4 Å. The effective domain volume increased and the effective number of domains remained constant between 3 and 4 ML on the clean surfaces and between 5 and ~7-8 ML on the NH3-saturated surfaces. These findings were attributed to the horizontal growth of the domain base stacking layer with ~3-4 Fe (111) atomic layers. On the clean surface, more than two domain stacking layers were grown for 4-10 ML, with decreasing horizontal size and volume with increasing ΘFe, while less than two domain stacking layers were grown on the NH3-saturated surfaces until 10 ML. These size and volume analyses indicate strong and weak 3D features of Fe crystalline islands on the clean and NH3-saturated surfaces, respectively.

I discussed the origin of the difference in the Fe island growth on clean and NH3-saturated Si(111)7×7 surfaces. On the clean surfaces, the topmost Si adatoms initiate Fe nucleation growth centers, and Fe islands epitaxially grow horizontally and vertically from the centers during the Fe deposition. The density of nucleation centers originating from the reactive Si adatoms restricts the horizontal Fe domain size owing to the conflict between neighboring growing Fe domains. On the NH3-saturated surfaces, where half of the Si adatoms are inactivated by dissociated NH2 molecules, the Fe horizontal domain size can become larger owing to the lower density of nucleation centers originating from the remaining bare adatoms. I suggest that Fe nucleation centers are initiated by either the two-adatom, three-adatom (bend), or three-adatom (center triangle) model among the six possible models. The models can explain the obtained horizontal Fe domain size and the ratio of the numbers of Fe domains on the clean and NH3-saturated Si(111)7×7 surfaces estimated from the diffraction spot intensities and domain volumes at the initial stage.

The two-adatom, three-adatom (bend), and three-adatom (center triangle) models, respectively, result in ~2.2, ~0.6, and ~0.10 nucleation centers per 7×7 half unit cell on clean surfaces, corresponding to densities of ~7.0×10ˆ13, ~1.9×10ˆ13,and ~3.2×10ˆ12 cmˆ-2, and in ~0.86, ~0.21, and ~0.018 nucleation centers per half unit cell on NH3-saturated surfaces, corresponding to densities of ~2.7×10ˆ13, 6.7×10ˆ12, and 5.6×10ˆ11 cmˆ-2. The wider horizontal crystalline domains on NH3-saturated surfaces imply the higher crystallinity of the Fe epitaxial islands despite the co-existence of the amorphous region compared with that on clean surfaces. The results of this study suggest that the partial termination of reactive Si(111)7×7 surfaces by characteristic molecules improves the crystalline quality of epitaxially grown highly reactive metal islands on the surfaces by reducing the density of nucleation centers to one per 7×7 half unit cell, corresponding to a density of ~3.8×10ˆ13 cmˆ-2. This idea can be applied to other highly reactive metals such as 3d transition metals and to lanthanide metals with interesting properties to design future nanodot devices.

Chapter 3

10 ML and 20 ML Fe(111) film were formed on Si(111)7×7 and NH3-saturated Si(111)7×7 surfaces grown by SPE method. Ex situ VSM at RT revealed that 10 ML Fe deposition show canted M-H curve implying the ferromagnetic has out-of-plane easy axis while, the deposition of 20 ML Fe on both surfaces show rectangular M-H curve indicating the magnetization has in-plane easy axis. The reasonable explanation for this are at 10 ML the M-H curve show canted magnetizations due to the inhomogeneity in the deposition, whereas thicker deposition, at 20 ML, Fe has uniform thickness. Furthermore, at higher coverage, ΘFe = 20 ML, NH3 layer caused the pinning effect of the domain-wall resulting in larger coercive force compare to Fe on the clean Si(111)7×7 surface.

Chapter 4

Surface investigation of atomically ordered Si{111}7×7 facet surface on a 3D pyramid Si(001) substrate has been successfully conducted using in situ RHEED. The RHEED pattern show the superimposed of Si{111}7×7 reconstruction and 2×1 pattern arising from the facet surface and the Si(001) substrate, respectively. For advanced applicability, 20 ML Fe was deposited on the 3D Si{111} pyramid arrays and investigated by RHEED. There are two type of pattern were observed. The first observed pattern is chevron pattern (thin streaks) at a certain azimuth angle indicating Fe{111} was grown epitaxially on the facet Si{111} surfaces and has a wide crystalline domain size. Indeed, the chevron pattern has the same angle as the fabricated pyramid, that is, 54° to the facet normal direction. The second pattern are broaden modulated spots along the c axis. These pattern did not change at the given azimuth angles rotation, the contribute to the given incident direction, indicate that partially the pure Fe(111) was formed fiber oriented Fe(111) on the top of the apex. The results of this study can contribute to the realization of high-performance 3D nanoscale integrated devices.

Chapter 5

In this chapter, I demonstrated different magnetism property of 3D Si {111} structures. Firstly, the magnetic property of ferromagnetic Fe thin film (ΘFe = 30 nm) on the 3D Si{111} pyramid with atomically flat and reconstructed {111} facet surfaces. The grown Fe film shown peculiar ferromagnetic properties which will depend on the geometric structures, conversely, this phenomena are not observed in planar Fe nanofilms. I reasonable argue that, the different magnetization in the 3D pyramid facet planes induced the creation and annihilation of vortices due to the shape anisotropy.

Secondly, I observe the ferromagnetic behavior of Fe nanofilm on 3D pyramid Si {111} base on thickness dependence. I found that the M-H curve have step-wise hysteresis loop as the function of the thickness and 3D architected structure. Different variant of slop exist on the M-H curve indicates different speed motion of the asymmetric vortex (off apex position). Using the potential energy as a function of vortex motion, I conclude that the stable position of the vortex is on the local minima on each face of the 3D pyramid, instead of on the apex. Increasing the thickness make the effect of shape anisotropy become less and less resulting in different stable magnetic configuration.

Lastly, I varied the shape of the original pyramid by fine-tuning the width and length ratio, so-called 3D Si facet-line, and study the magnetic property. It revealed that, when the facet-line has given an external magnetic field perpendicular to the facet-line direction, the M-H curve shown that the longer the facet-line is, the canter the shape is, this is indicating magnetic easy axis with the majority of the magnetic moment have out-of-plane component. In contrast, when the external magnetic field given at the parallel direction, the M-H curve shown almost rectangular shape despite the different width and length ratio of the facet-line, which implies that the systems has massive in-plane component and negligible out-of-plane component . By fine-tuning the 3D properties, such as size (width and length ratio) and thickness of ferromagnetic thin film, one can control the magnetic property of 3D shape nanostructure. These results demonstrated a novel technique which will lead to the development of nanomaterial science and technology.

Summary

I have successfully controlled the shape and crystallinity of Fe(111) metal film islands on atomically-flat Si(111) surfaces with 7×7 superstructures, utilizing precise surface treatments and electron diffraction measurements for in-situ growth with spot-analysis methods. In addition, with an original idea of substrate surface modification by a NH3 molecular seed-layer insertion between the films and surfaces, which optimizes the nucleation centers for metal growth, I definitely increased in-plane crystallinity of the grown Fe islands and observed the magnetic properties which show the pinning effect. In the future, molecules are promising to use as the domain wall pinning to fine tuning the magnetization reversal process. Furthermore, I successfully created epitaxial Fe films on atomically-flat facet surfaces of 3D fabricated pyramid microstructures confirmed with in situ electron diffraction, and revealed the peculiar ferromagnetic properties of pyramidal Fe nano films originated from the vortex motion reflecting the 3D shape effect. These results will lead to future designation for magnetic vortex, i.e., the control of spin orientation in space utilizing characteristic 3D shaped

ferromagnetic films.