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Growth and characterization of Fe nanostructures on GaN

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ABSTRACT

We have investigated the growth of Fe nanostructures on GaN(0 0 0 1) substrates at room temperature using reflection high-energy electron diffraction (RHEED), scanning tunneling microscopy (STM), and superconducting quantum interference device magnetometer. Initially, a ring RHEED pattern appears, indicating the growth of polycrystalline α-Fe. At around 0.5 nm deposition, the surface displays a transmission pattern from α-Fe films with the epitaxial relationship of Fe(1 1 0)//GaN(0 0 0 1) and Fe[1 −1 1]//GaN[1 1 −2 0] (Kurdjumov–Sachs (KS) orientational relationship). Further deposition to 1 nm results in the appearance of a new spot pattern together with the pattern from domains with the KS orientation relationship. The newly observed pattern shows that Fe layers are formed with the epitaxial relationship of Fe(1 1 0)//GaN(0 0 0 1) and Fe[0 0 1]//GaN[1 1 −2 0] (Nishiyama–Wasserman (NW) orientational relationship). From STM images for Fe layers with the KS and NW orientational relationships, it can be seen that Fe layers with the KS relationship consist of round-shaped Fe nanodots with below 7 nm in average diameter. These nanodots coalesce to form nanodots elongating along the Fe[1 0 0] direction, and they have the KS orientational relationship. Elongated Fe nanodots with the NW relationship show ferromagnetism while round-shaped Fe nanodots with the KS relationship show super-paramagnetic behavior. We will discuss their magnetic properties in connection with the change in crystalline configurations of nanodots.

1. Introduction

Spin-dependent phenomena in semiconductors have been studied from the viewpoint of the fabrication of semiconductor spintronic devices, which utilize the spin degree of freedom of electrons, as well as from the viewpoint of understanding their physical processes. Since conventional semiconductors such as Si, GaAs, and so on are non-magnetic materials, injection of spins into conventional semiconductors is required to fabricate semiconductor spintronic devices. There are mainly two ways for introducing the spin degree of freedom of electrons into them. One is to make conventional semiconductors ferromagnet, the other is to use ferromagnetic metals as spin injectors. Dilute magnetic semiconductors (DMSs) doped with transition or rare-earth metals are a promising candidate as ferromagnetic semiconductors for spintronic devices. We have reported the synthesis of Cr-doped GaN showing ferromagnetic behavior even at room temperature (RT) [1] and the observation of the tunneling magnetoresistance effect in GaCrN/AlN/GaCrN trilayer tunneling junctions [2]. On the other hand, ferromagnetic metals are also a good candidate for spin source injected into non-magnetic semiconductors. Since recent theoretical calculations [3] predict a long spin relaxation lifetime in GaN compared with that in GaAs, a heterostructure of Fe/GaN is the very interesting subject of spin-related transport phenomena in semiconductors. Several groups have reported thick Fe films (around 70 nm) grown on hexagonal GaN(0 0 0 1) [4] and cubic GaN(0 0 1) [5] substrates towards the application to spintronic devices.

In this paper, we have investigated the initial stages of Fe growth and magnetism of Fe nanostructures grown on GaN(0 0 0 1) surfaces. We will discuss their magnetic properties in connection with the change in crystalline configurations of Fe nanostructures.

2. Experimental procedure

Substrates used in this study were *n*-type GaN(0 0 0 1) templates grown by metal organic chemical vapor deposition. Just before introducing them into an ultra-high vacuum chamber equipped with reflection high-energy electron diffraction (RHEED) and scanning tunneling microscopy (STM), the substrates were chemically etched with a mixed solution of H_2SO_4, H_2O_2 and H_2O to remove oxide layers on GaN surfaces. Without annealing treatments in the UHV chamber, the substrate surfaces exhibited a

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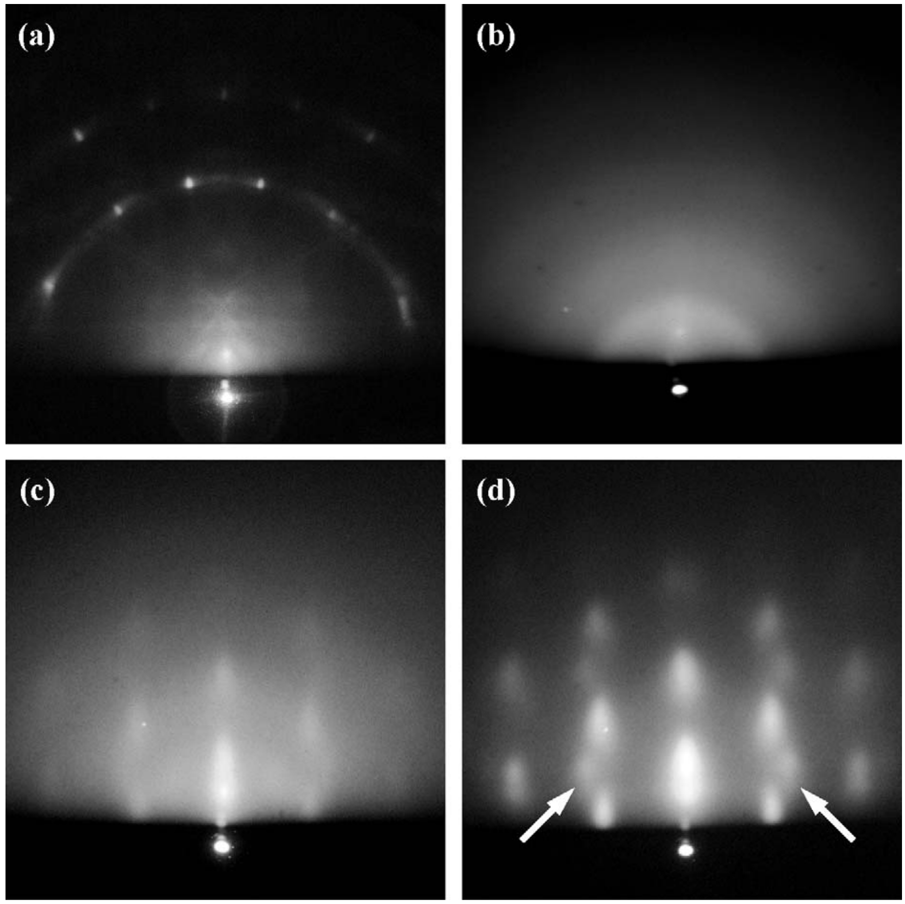

Fig. 1. Evolution of RHEED patterns as a function of the thickness of Fe layers. (a) GaN(0 0 0 1) (1×1), (b) 0.3 nm, (c) 0.5 nm, (d) 1.0 nm.

1×1 RHEED pattern, as shown in Fig. 1(a). Fe was evaporated on the 1×1 surfaces at room temperature from a pure Fe rod by using an electron-beam evaporator in the chamber. After taking out the samples outside the UHV chamber, magnetization of the samples was measured using superconducting quantum interference device (SQUID) magnetometer. Assuming that the magnetic moment per Fe atom is equal to 2.22 μ_B (Bohr magneton), the deposition rate is calculated as 0.4 nm/min on the value of the saturation magnetization of ferromagnetic samples.

3. Results and discussion

Fig. 1 shows the evolution of RHEED patterns along the GaN[1 1 −2 0] azimuth as a function of Fe growth on the GaN(1×1) surfaces shown in Fig. 1(a). From the RHEED observation, it can be seen that the initial growth of Fe on the GaN(1×1) proceeds in three stages. Initially, a ring RHEED pattern appears as shown in Fig. 1(b), indicating the growth of polycrystalline Fe. After a deposition of 0.5 nm, a 3D transmission pattern shown in Fig. 1(c) is observed. The RHEED pattern indicates the growth of α-Fe (bcc Fe) having an epitaxial relationship with Fe(1 1 0)//GaN(0 0 0 1) and Fe[1 −1 1]//GaN[1 1 −2 0]. Further deposition of Fe (1 nm in total thickness) produces an appearance of additional 3D transmission spots as indicated by arrows in Fig. 1(d). The appearance of these RHEED spots shows that α-Fe also grows with the orientational relationship being Fe(1 1 0)// GaN(0 0 0 1) and Fe[0 0 1]//GaN[1 1 −2 0]. We can rationalize these preferred epitaxial relationships in terms of the purely geometrical row-matching model for fcc(1 1 1)/bcc(1 1 0) epitaxial systems, that is, so-called Nishiyama–Wasserman (NW) and Kurdjumov–Sachs (KS) orientational relationships [6]. The NW relationship is simply row-matching along the fcc[1 −1 0] and bcc[0 0 1] directions (fcc[1 −1 0]//bcc[0 0 1]). The KS relationship is the close-packed row-matching (fcc[1 −1 0]//bcc[1 −1 1]) by rotating the fcc overlayer by around 5° relative to the NW. Since the GaN(0 0 0 1) plane has the same symmetry as the fcc(1 1 1) plane, we can apply the NW and KS relationship to the Fe(1 1 0) on GaN(0 0 0 1) system. The Fe[1 −1 1]//GaN[1 1 −2 0] relationship observed in Fig. 1(c) corresponds to the KS relationship (bcc[1 −1 1]//fcc[1 −1 0]). There are two orientational relationships KS1 and KS2 as shown in Fig. 2(a). The lattice mismatches along the Fe[1 −1 1] and Fe[−1 1 2] directions are 22% and 15%, respectively. For the Fe[0 0 1]//GaN[1 1 −2 0] relationship observed in Fig. 1(d), the corresponding one is the NW relationship (bcc[0 0 1]//fcc[1 −1 0]). There is one orientational relationship as shown in Fig. 2(b). The lattice mismatches along the Fe[0 0 1] and Fe[1 −1 0] directions are 10% and 27%, respectively. It turns out that the Fe overlayer showing the RHEED pattern of Fig. 1(d) consists of domains with the KS and NW orientational relationships. It should be noted that because of 3-fold symmetry of the GaN(0 0 0 1) plane, there are three equivalent domains in each orientational relationship (KS1, KS2, or NW).

Fig. 3(a)–(c) shows STM images for Fe overlayers on GaN(0 0 0 1) exhibiting ring, the NW relationship, and the KS + NW relationship RHEED patterns, respectively. The respective thicknesses are 0.3, 0.8, and 1 nm. From the images, it can be seen that RT deposition of Fe on GaN(0 0 0 1) produces nanodots, and that as the thickness of Fe increases, the size of Fe nanodots increases. The average sizes of nanodots in Fig. 3(a)–(c) are 3, 4, and 8 nm, respectively. We also notice that nanodots both without a preferred orientational relationship (Fig. 3(a)) and with the NW relationship (Fig. 3(b)) have round shapes. On the other hand, at