Two-dimensional growth of Fe thin films with perpendicular magnetic anisotropy on GaN(0001)

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The growth and magnetism of Fe thin films on the GaN(0001) surface are studied by scanning tunneling microscopy and surface magneto-optic Kerr effect. It is found that Fe grows in a layer-by-layer mode on the pseudo-1×1 surface at room temperature, and the film develops magnetism at 1.2 ML and shows perpendicular magnetic anisotropy below 6 ML. On the bulk-terminated 1×1 surface, Fe grows in a three-dimensional mode, and ferromagnetization with in-plane anisotropy is observed only above 4.3 ML. Fe-induced $\sqrt{7}\times\sqrt{7}$ reconstruction on the pseudo-1×1 surface plays the key role in reducing the interface reaction and promoting the two-dimensional growth. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2210792]

Injection of spin polarized electrons into semiconductors (SCs) is the first step towards the realization of SC-based spintronics devices, which utilize the spin degree of freedom of electrons.[1] Ferromagnetic metals (FMs) are a good candidate for spin source because of their high Curie temperatures and well-known magnetic properties. Although the injection efficiency through a FM/SC interface is rather low in a diffusive transport regime due to the large resistance mismatch,[2] the situation can be dramatically improved in the case of Schottky contact by tunneling or hot electron emission through the interface barrier.[3,4] Previous studies on FM/SC were focused on the Fe films on the surfaces of conventional III-V semiconductors with zinc blende structure such as GaAs,[5–12] InAs,[13] and InP[14] for they have relatively small lattice mismatches with bcc Fe. Few works were devoted to the FM films, especially in the ultrathin regime, on the surfaces of other semiconductors. With the rapid development of GaN-based single crystalline materials, atomically flat GaN surfaces with well-defined orientations and reconstructions could be routinely obtained.[15,16] By delicate control of the growth kinetics, the layer-by-layer growth of Ag on GaN(0001) has recently been reported.[17] In this study, we investigate the growth and magnetism of Fe films on both the GaN(0001) pseudo-1×1 and bulk-terminated 1×1 surfaces, and find that atomically flat films could be achieved on the pseudo-1×1 surface, which show a perpendicular magnetic anisotropy (PMA).

Our experiments were carried out in an OMICRON molecular beam epitaxy scanning tunneling microscopy (MBE-STM) ultrahigh vacuum system equipped with an Oxford radio frequency plasma source and a homemade surface magneto-optic Kerr effect (SMOKE) setup. The base pressure of the system is lower than 1.0×10^{-10} mbar. A He–Ne laser with a wavelength of 632.8 nm and a silicon photodetector are used to excite and collect the SMOKE signals, while a lock-in amplifier with a precise photoelastic modulator of 50 kHz is used to increase the signal/noise ratio.

The substrates used in this experiment are commercial GaN(0001) grown by metal organic chemical vapor deposition on sapphire. After degassing at 300 °C for 2 h, the substrate was exposed to nitrogen plasma at a substrate temperature of about 700 °C for 15 min, followed by Ga deposition of ~3 ML. After removing Ga droplets on the substrate by annealing at ~700 °C for several minutes, we obtained the pseudo-1×1 surface. Bulk-terminated 1×1 surface was prepared by further annealing the pseudo-1×1 surface to remove the top Ga bilayer.[17] Fe was evaporated from Fe slugs resistively heated by a Ta boat. The flux of Fe was calibrated by the STM images of Fe films on Cu(100), approximately 0.08 ML/min in this experiment. The pressure was around 4×10^{-10} mbar during Fe deposition. Wedge-shaped samples were used in SMOKE measurements to avoid signal fluctuations in different experiments and obtain reliable thickness dependence of the magnetic properties.

Pseudo-1×1 is the most stable structure on the Ga-polar GaN(0001) surface, which contains a Ga bilayer above the truncated bulk surface. It is characterized by strong 1×1 spots with satellite peaks in electron diffraction patterns,[18,19] and appears atomically flat but featureless in STM images. Figure 1(a) shows a typical STM image of this surface prepared by our method. The surface is basically atomically flat except for small amounts of depressions. After deposition of 0.16 ML Fe on the surface at room temperature, ordered regions appear, as shown in Fig. 1(b). High resolution STM images of a reconstructed area at different bias voltages are shown in Fig. 2. Its unit cell size is about 7.6×7.6 Å^2, nearly $\sqrt{7}$ times (about 10% smaller) of the GaN(0001) base vector, thus the surface is $\sqrt{7}\times\sqrt{7}$ reconstructed. The contraction was also observed in the Mn induced $\sqrt{3}\times\sqrt{3}$ reconstruction on GaN(0001) pseudo-1×1.[20] The possible reason is that the top Ga layer of GaN(0001) pseudo-1×1 is laterally contracted according to Smith *et al.*[18] More importantly, as shown in Figs. 2(a)–2(d), the STM images exhibit a very strong bias-dependent behavior, suggesting that the surface has semiconductorlike electronic states.

At 0.48 ML [Fig. 1(c)], islands form on the $\sqrt{7}\times\sqrt{7}$ surface. All islands have a height of about 2 Å, i.e., one

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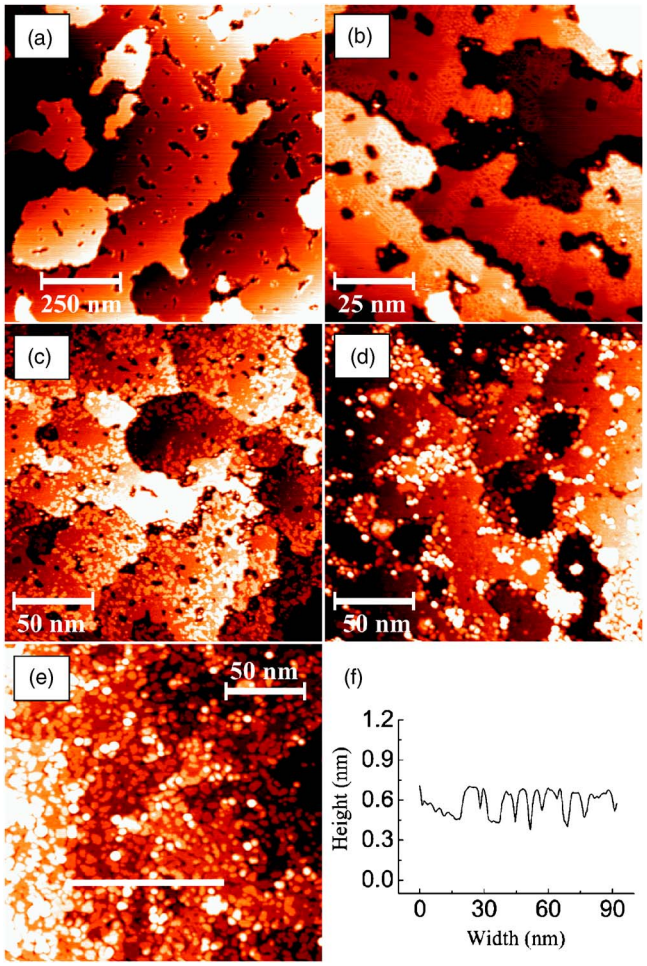

FIG. 1. (Color online) STM images of (a) clean surface of the GaN(0001) pseudo-1×1 (1000×1000 nm^2) and after deposition of (b) 0.16 ML (100×100 nm^2), (c) 0.48 ML (200×200 nm^2), (d) 2.0 ML (200×200 nm^2), and (e) 5.5 ML (200×200 nm^2) of Fe on the GaN(0001) pseudo-1×1 surface at room temperature. (f) Line profile taken from the STM image in (e).

atomic layer high. With increasing coverage, Fe grows nearly in a layer-by-layer mode. Figures 1(d) and 1(e) show the STM images of the surfaces after 2 and 5.5 ML depositions, respectively. The surfaces are essentially atomically flat and the height fluctuation does not exceed 1 ML [see Fig. 1(f)].

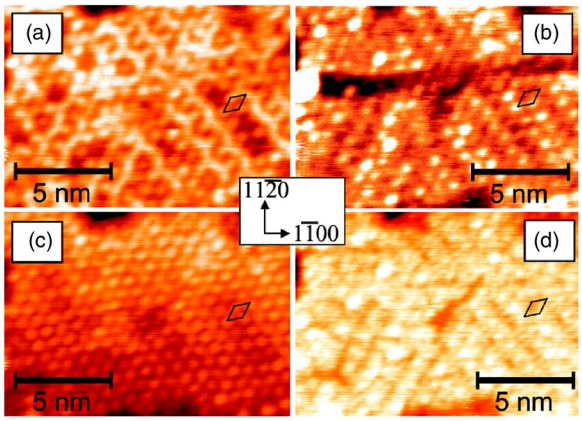

FIG. 2. (Color online) Bias-dependent STM images of the Fe-induced $\sqrt{7}\times\sqrt{7}$ reconstruction on GaN (0001) pseudo-1×1 surface: (a) +2 V, (b) −3 V, (d) +1 V, and (d) −1 V. The black rhombus indicates a unit cell of the reconstruction.

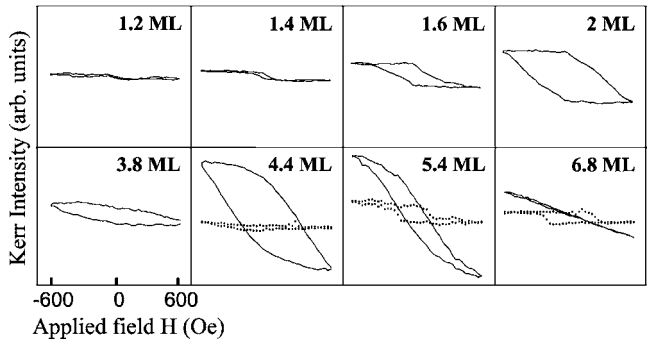

FIG. 3. (Color online) Representative hysteresis loops measured by SMOKE at 100 K. The polar loops are plotted in solid lines and the longitudinal loops in dotted lines. All polar loops have the same scale, which, however, is different from that of the longitudinal ones.

This kind of morphology holds for all coverages (up to 7 ML) studied.

The above results are different from those of the Fe films grown on Ga-rich GaAs surfaces in which Fe favors three-dimensional (3D) growth.[6–9] The difference may come from the Fe-induced $\sqrt{7}\times\sqrt{7}$ reconstruction. In terms of the semiconducting electronic structure, the formation of this reconstruction not only wets the GaN surface but also provides a stable template to promote the two-dimensional (2D) growth. The decrease in interface roughness can significantly increase the spin injection efficiency.[21] Thus, a high efficiency spin injection can be expected in Fe/GaN(0001) pseudo-1×1.

The magnetism of the Fe films was measured with SMOKE at both room temperature and 100 K. The representative hysteresis loops measured at 100 K were shown in Fig. 3. At 100 K, the polar signal appears as early as 1.2 ML, while the longitudinal signal is absent, suggesting that the magnetic anisotropy is perpendicular to the surface. The coercivity increases dramatically with increasing Fe thickness. Above 2 ML, the films cannot be saturated by the maximum magnetic field (600 Oe) of our system, leading to a decrease in the magnetic signal until 3.8 ML. Above 5 ML, the polar loops become slanted, and both the coercivity and the remanence of the polar loops decrease, while the longitudinal signal appears gradually. Above 6 ML, the remanence of the polar loops decreases to zero and the easy axis rotates to in plane. It suggests that a spin reorientation transition takes place at ~6 ML.

PMA is generally observed in ultrathin Fe films grown on various metal surfaces.[22] However, on the conventional III-V semiconductor surfaces, such as GaAs(001), the Fe films usually show an in-plane uniaxial magnetic anisotropy, and it is commonly believed that the interface plays an important role in the magnetic properties.[21] The Fe/GaAs interface is dominated by Fe–As bonding. In the case of Fe/GaN(0001) pseudo-1×1, the Ga bilayer separates Fe from N, reducing their bonding significantly. Therefore it is possibly a weaker interface interaction that mainly leads to the appearance of PMA.

In a surface emitting spin LED, polarized light can only be obtained with perpendicular magnetized FM layer. So Fe/GaAs based spin LED, which always shows in-plane magnetic anisotropy, can only work in an external perpendicular magnetic field.[1] The PMA of Fe/GaN(0001) pseudo-

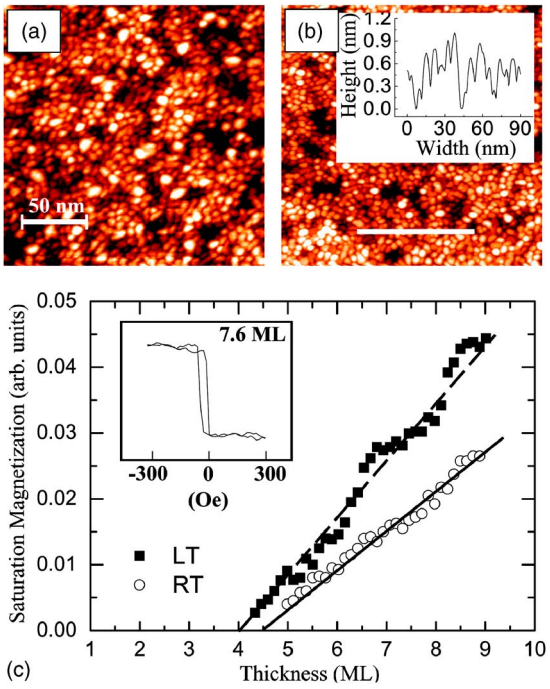

FIG. 4. (Color online) STM images of (a) 3 ML and (b) 7.6 ML Fe grown on the GaN(0001) bulk-terminated 1×1 surface (200×200 nm^2). The inset of (b) shows the line profile. (c) Thickness dependence of the saturation magnetization of the longitudinal hysteresis loops of the Fe films on the bulk-terminated 1×1 surface measured at 100 K and room temperature. The inset shows the hysteresis loop of 7.6 ML film measured at 100 K.

1×1 makes it prospectively useful in the fabrication of spin LED free of external field.

On the bulk-terminated 1×1 surface, Fe grows in a totally different way. Fe forms 3D clusters immediately under various growth conditions. Figures 4(a) and 4(b) show STM images of the surface after 3 and 7.6 ML Fe depositions. The lateral size of the clusters is about 5 nm and the surface roughness is about 1 nm [see the line profile in Fig. 4(b)]. The morphology does not change much at different thicknesses and is basically similar to those of the Fe films grown on surfaces of GaAs, InP, and InAs.[12–14]

SMOKE measurements of the Fe films on the bulk-terminated 1×1 surface show that only longitudinal signals can be detected for all thicknesses studied, which indicate that the easy axis is always in plane. The thickness-dependent saturation magnetization of the longitudinal loops [see Fig. 4(c)] demonstrates that the magnetic signal can only be observed above 4.3 ML even at 100 K. Figure 4(c) also indicates that the extrapolation line for the magnetization-thickness curve at large thickness does not cross the origin, instead, shows an intercept at about 4 ML. Moreover, the intercept does not change much when measured at room temperature, which means that the amount of nonmagnetic Fe is influenced little by temperature. So we can conclude that the superparamagnetism of Fe clusters is not the main cause for the nonmagnetic layer, and nonmagnetic compounds must form.

In summary, we found that Fe can grow in a 2D mode on the pseudo-1×1 surface, and the film has PMA below 6 ML. The properties make the Fe/GaN system promising to yield a high efficiency spin injection in spintronics devices.

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[1] I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 2 (2004).
[2] B. G. Schmidt, D. Ferrand, L. W. Molenkamp, A. T. Filip, and B. J. van Wees, Phys. Rev. B **62**, R4790 (2000).
[3] E. I. Rashba, Phys. Rev. B **62**, R16267 (2000); D. J. Monsma, R. Vlutters, and J. C. Lodder, Science **281**, 407 (1998).
[4] H. J. Zhu, M. Ramsteiner, H. Kostial, M. Wassermeier, H. P. Schönherr, and K. H. Ploog, Phys. Rev. Lett. **87**, 016601 (2001); A. T. Hanbicki, B. T. Jonker, G. Itskos, G. Kioseoglou, and A. Petrou, Appl. Phys. Lett. **80**, 1240 (2002); A. T. Hanbicki, O. M. J. van't Erve, R. Magno, G. Kioseoglou, C. H. Li, B. T. Jonker, G. Itskos, R. Mallory, M. Yasar, and A. Petrou, *ibid.* **82**, 4092 (2003).
[5] G. A. Prinz, G. T. Rado, and J. J. Krebs, J. Appl. Phys. **53**, 2087 (1982); J. J. Krebs, B. T. Jonker, and G. A. Prinz, *ibid.* **61**, 2596 (1987).
[6] P. N. First, J. A. Stroscio, R. A. Dragoset, D. T. Pierce, and R. J. Celotta, Phys. Rev. Lett. **63**, 1416 (1989).
[7] M. W. Ruckman, J. J. Joyce, and J. H. Weaver, Phys. Rev. B **33**, 7029 (1986).
[8] S. A. Chambers, F. Xu, H. W. Chen, I. M. Vitomirov, S. B. Anderson, and J. H. Weaver, Phys. Rev. B **34**, 6605 (1986).
[9] B. T. Jonker, G. A. Prinz, and Y. U. Idzerda, J. Vac. Sci. Technol. B **9**, 2437 (1991).
[10] M. Gester, C. Daboo, R. J. Hicken, S. J. Gray, A. Ercole, and J. A. C. Bland, J. Appl. Phys. **80**, 347 (1996).
[11] E. M. Kneedler, B. T. Jonker, P. M. Thibado, R. J. Wagner, B. V. Shanabrook, and L. J. Whitman, Phys. Rev. B **56**, 8163 (1997); P. M. Thibado, E. Kneedler, B. T. Jonker, B. R. Bennett, B. V. Shanabrook, and L. J. Whitman, *ibid.* **53**, R10481 (1996).
[12] Y. B. Xu, E. T. M. Kernohan, D. J. Freeland, A. Ercole, M. Tselepi, and J. A. C. Bland, Phys. Rev. B **58**, 890 (1998).
[13] Y. B. Xu, E. T. M. Kernohan, M. Tselepi, J. A. C. Bland, and S. Holmes, Appl. Phys. Lett. **73**, 399 (1998); Y. B. Xu, D. J. Freeland, M. Tselepi, and J. A. C. Bland, Phys. Rev. B **62**, 1167 (2000).
[14] F. Zavaliche, W. Wulfhekel, and J. Kirschner, Phys. Rev. B **65**, 245317 (2002).
[15] A. R. Smith, R. M. Feenstra, D. W. Greve, J. Neugebauer, and J. E. Northrup, Phys. Rev. Lett. **79**, 3934 (1997).
[16] Q. K. Xue, Q. Z. Xue, R. Z. Bakhtizin, Y. Hasegawa, I. S. T. Tsong, and T. Sakurai, Phys. Rev. Lett. **82**, 3074 (1999); Z. T. Wang, Y. Yamada-Takamura, Y. Fujikawa, T. Sakurai, and Q. K. Xue, Appl. Phys. Lett. **87**, 032110 (2005).
[17] K. Wu, Q. Z. Xue, R. Z. Bakhtizin, Y. Fujikawa, X. Li, T. Nagao, Q. K. Xue, and T. Sakurai, Appl. Phys. Lett. **82**, 1389 (2003).
[18] A. R. Smith, R. M. Feenstra, D. W. Greve, M.-S. Shin, M. Skowronski, J. Neugebauer, and J. Northrup, J. Vac. Sci. Technol. B **16**, 2242 (1998).
[19] J. E. Northrup, J. Neugebauer, R. M. Feenstra, and A. R. Smith, Phys. Rev. B **61**, 9932 (2000).
[20] Y. Cui and L. Li, Surf. Sci. **522**, L21 (2003).
[21] G. Wastlbauer and J. A. C. Bland, Adv. Phys. **54**, 137 (2005).
[22] C. Liu and S. D. Bader, J. Vac. Sci. Technol. A **8**, 2727 (1990).