

MOCVD growth, structure and magnetic properties of Fe films grown on GaAs (001) substrates

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Abstract

Thin iron films have been grown on (001) GaAs substrates by low pressure metal organic chemical vapor deposition (LP-MOCVD) at different temperatures with the pressure of 150 Torr. X-ray diffraction (XRD) analysis showed that all films have only one strong diffraction peak (110). The surface of Fe film became smooth with increasing the growth temperature. Magnetization measurements showed that the Fe films grown at different temperatures were ferromagnetic with easy axis parallel to the film surface and hard axis perpendicular to the substrates. The field dependence of magnetization along two axes showed a remarkable difference, implying that the samples have strong magnetic anisotropy. Furthermore, when the applied magnetic field is perpendicular to the Fe surface, a sharp jump in the hysteresis loop could be observed, followed by a broad shoulder, which is related to the interface effect, the existence of carbon and the formation of 180°/90° magnetic domains.

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1. Introduction

Ferromagnetic thin films on semiconductor substrates have been of great interest in recent years because of the possible future development of spintronics, e.g., spin injection through the ferromagnetic thin-film semiconductor interfaces [1–5]. The Fe/GaAs system has attracted much attention in this area of study because of its room temperature ferromagnetism and small lattice mismatch (1.3%) [6]. Based on the small lattice mismatch, the stable bcc phase of Fe grew epitaxially on GaAs (001), GaAs (110), GaAs (113)A, and GaAs (133)A substrates, and numerous studies optimizing its growth conditions and analyzing its interface structure have been reported [7–16]. Much attention has been concentrated on the magnetic behavior of Fe films with the thickness below 100 nm. The thin films of Fe/GaAs (001), with the thickness less than about

60 ML or 9 nm, exhibit a remarkable and unexpected in-plane uniaxial magnetic anisotropy (UMA) with an [110] easy axis [10,14,21–23]. This is dramatically different from the cubic magnetic anisotropy of bulk bcc Fe with (100) easy axes. The transformation from superparamagnetism (SP) to ferromagnetism (FM) is observed for ultrathin Fe films [18]. Up to now, most of the Fe/GaAs structures were deposited by molecular beam epitaxy (MBE) [17,20,23–26]. However, the growth of an Fe epitaxial layer on a GaAs substrate with a high-quality interface using MOCVD has been also reported [27]. The hysteresis loop along the [100] and [110] axes has been shown at low temperature (6 K) for the sample thickness below 60 nm. It can be observed that the easy axis was not clear for the thinner films (<20 nm). By comparison with that prepared by MBE, it seemed that the different growth methods could lead to different results in magnetic properties of the films.

In this paper, high quality Fe films have been prepared by using LP-MOCVD at different temperatures. The surface morphology of the Fe films was investigated with an atomic force microscope (AFM). The magnetic properties were measured

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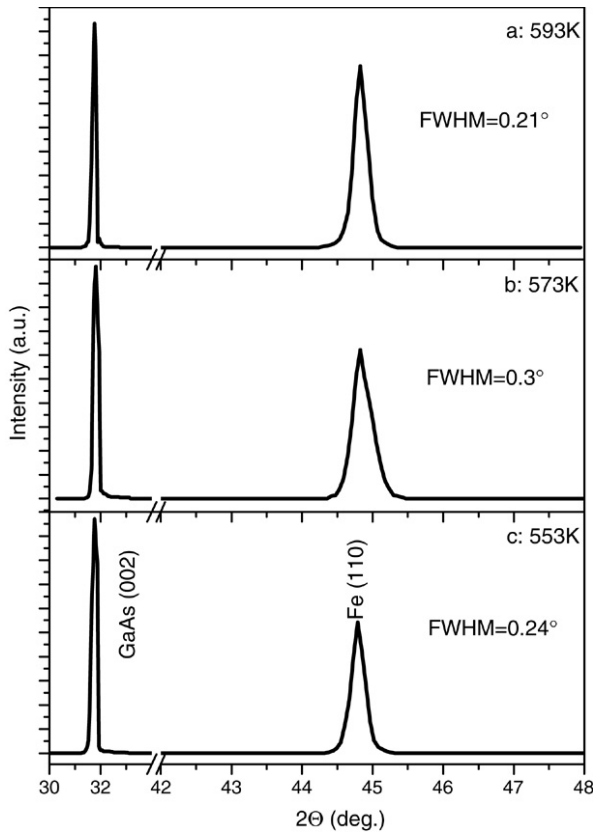


Fig. 1. XRD spectra of Fe films with different growth temperatures: (a) 593 K; (b) 573 K; (c) 553 K.

by vibrating sample magnetometer (VSM). A clear magnetic anisotropy of the Fe films was observed at room temperature.

2. Experiments

Growth was carried out in a LP-MOCVD system with a horizontal rectangular quartz reactor at low pressure. The ironpentacarbonyl ($\text{Fe}(\text{CO})_5$) carried by ultrahigh purity hydrogen was used as material source. The source was kept at 283 K during deposition. The GaAs substrates were semi-insulating with (001) orientation. The substrates were cleaned in trichloroethylene, acetone and ethanol for 5 min in an ultrasonic bath, and etched in a sulfuric peroxide solution ($3\text{H}_2\text{SO}_4 + 1\text{H}_2\text{O}_2 + 1\text{H}_2\text{O}$) for 5 min before loaded into the reactor. The substrates were placed at the center of a graphite boat, and then inserted in the reactor. The substrates were heated to 873 K for 10 min in H_2 gas flow. The deposition was carried out at the pressure of 150 Torr with different temperatures (553 K, 573 K and 593 K). The flow rate of $\text{Fe}(\text{CO})_5$ was kept at 8 sccm/min.

The structure and composition of the films were studied by XRD. As for the magnetic property of the samples, the field dependences of magnetization (M – H curves) were measured by the VSM system at room temperature.

3. Results and discussion

In order to study the effect of substrate temperature (T_s) on crystallinity, the iron films were grown on (001) GaAs

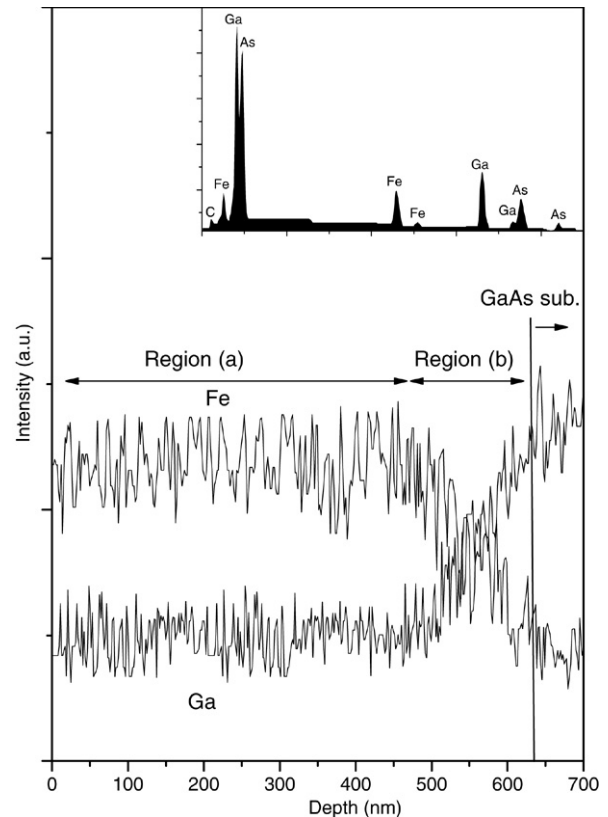


Fig. 2. Depth profile of Fe films measured by Auger electron spectroscopy. The inset is energy dispersive spectroscopy of the samples.

substrates with the T_s ranging from 553 to 593 K. It is well known that the lattice constant of Fe is slightly bigger than half of the lattice constant of GaAs and the lattice mismatch between Fe and GaAs is 1.3%. XRD measurement with $\text{Cu K}\alpha$ radiation ($\lambda = 0.154178$ nm) was used to make θ – 2θ scans to determine (110) d -spacing lattice constant for the Fe films, just as shown in Fig. 1. In this figure, the diffraction peak at $2\theta \approx 44.7^\circ$ can be associated with the (110) plane of the bcc Fe phase, and no other diffraction peak was detected. Three samples have similar results except that the peak width and intensity are different. Furthermore, the intensity of Fe (110) diffraction peak is increasing with the increase of the growth temperature. The full width at half-maximum (FWHM) of the (110) diffraction peaks are 0.21° for sample *a*, 0.3° for sample *b* and 0.24° for sample *c*. From Fig. 1, it can be calculated that the (110) d -spacing lattice constant is 0.20242 ± 0.00004 nm, which is close to the bulk α -Fe of 0.20265 nm. Fig. 2 shows the depth profiles of Fe film measured by Auger electron spectroscopy (AES). The sample was prepared at 593 K with the thickness of about 550 nm. As shown in Fig. 2, the profile pattern consists of two regions: region (a) and region (b). They correspond to the regions from the surface to the depth of 510 nm and from the depth of 510 nm to the substrate, respectively. In region (a) a signal from Fe is detected and the intensity of the signal is constant. In region (b) the signal of Fe decreases toward the interface of the Fe/GaAs substrate. The decrease of the signal intensity of Fe is caused by the diffusion of Ga or As atoms. The composition

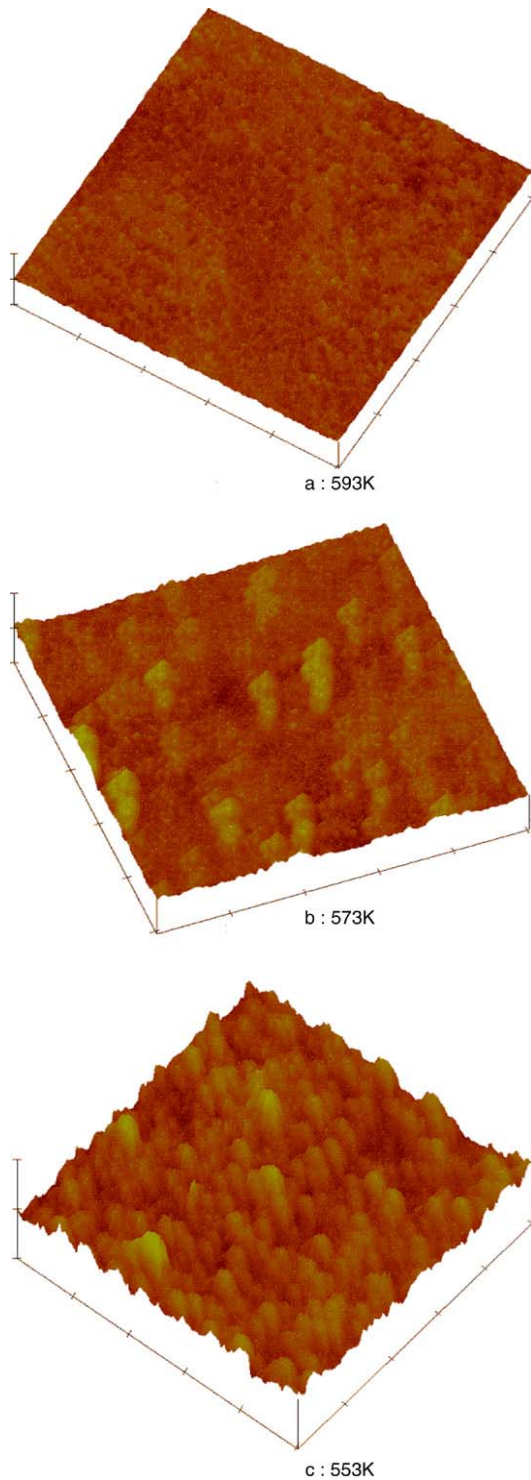


Fig. 3. AFM images of Fe films grown at the temperature of (a) 593 K, (b) 573 K and (c) 553 K. Image size is $0.5 \times 0.5 \mu\text{m}$ with 150 nm high.

of Fe film was studied by EDS (see the inset of Fig. 2). No obvious O signal has been detected and little of carbon (weight ratio $\approx 1.2\%$) may come from the decomposition of $\text{Fe}(\text{CO})_5$. Fig. 3 shows the surface morphology of the Fe films prepared at 553 K, 573 K and 593 K, respectively. The thickness of the Fe films is about 550 nm. The scanned area is $5 \times 5 \mu\text{m}^2$. The AFM pictures clearly show the

differences in roughness of three samples. From Fig. 3, it can be observed that the root mean square (rms) surface roughness (σ) of samples is 5.102 nm (a: 593 K), 8.100 nm (b: 573 K) and 11.259 nm (c: 553 K), respectively. The surface of Fe film is smooth (rough) when the growth temperature is high (low). The magnetic properties of Fe films grown at different temperatures were studied with a vibrating sample magnetometer system. The sample's size is approximately $0.3 \times 0.3 \text{ cm}^2$. Fig. 4 shows the results of the magnetic moment measurement when the applied magnetic field is parallel to or perpendicular to the surface of Fe films at room temperature. When the applied field is parallel to the surface, the magnetization curves along these directions exhibit a quasi-rectangular shape for all the samples, just as shown in Fig. 4. However, when the applied magnetic field is perpendicular to the Fe films, a sharp jump in the hysteresis loop could be observed, followed by a broad shoulder. This process can be divided into two distinct dynamic regimes, as shown in the figure: the low dynamic regime, extending to magnetic field $< 100\text{Oe}$, and the high dynamic regime when magnetic field $> 100\text{Oe}$. According to Raquet et al., [19] magnetization reversal in the low dynamic regime is predominantly by domain-wall motion, and in the high dynamic regime is predominantly by domain nucleation. A number of reasons could be responsible for this trend: these include surface partial oxidation because of the exposure to the air, interface effect, surface roughness, the existence of carbon or the formation of $180^\circ/90^\circ$ magnetic domains [20]. We discounted the surface partial oxidation because no obvious O signal was detected by EDS and this effect would be very weak if there is any. As for the surface roughness, we did not find large difference in $M-H$ curves (applied field perpendicular to the film plane) of the samples grown at different temperatures with root mean square (rms) surface roughness (σ). So this reason can also be excluded. Therefore, the resulting broad shoulder can be attributed to the interface effect, the existence of carbon and the formation of $180^\circ/90^\circ$ magnetic domains, but it is not clear which is the dominating reason. Furthermore, from Fig. 4, the square loop behavior in-plane and the slow approach to saturation out-of-plane for the samples indicated that the Fe films grown at different temperatures are ferromagnetic with easy axis along the film surface. The magnetic moment per unit volume (emu/cm^3) of sample a is $1620 \text{ emu}/\text{cm}^3$, which is smaller than that of bulk $\alpha\text{-Fe1707}$ emu/cm^3 at room temperature. The growth-temperature dependence of the coercivity (H_c) and the saturation magnetization (M_s) is shown in Fig. 5, when the applied field is parallel to the film surface. By comparison, it can be seen that the 593 K (573 K) sample has the lowest (highest) H_c but the highest (lowest) M_s among the three samples. This is in agreement with the XRD observation that sample a (b) has the narrowest (widest) FWHM value. According to Scherrer's formula, the grain size of sample a (b) will be much larger (smaller) than the other two samples. Since the ferromagnetic coupling is strongly dependent on the exchange interaction between neighboring magnetic ions, decrease of the grain size (or domain) can even change a system from the ferromagnetic

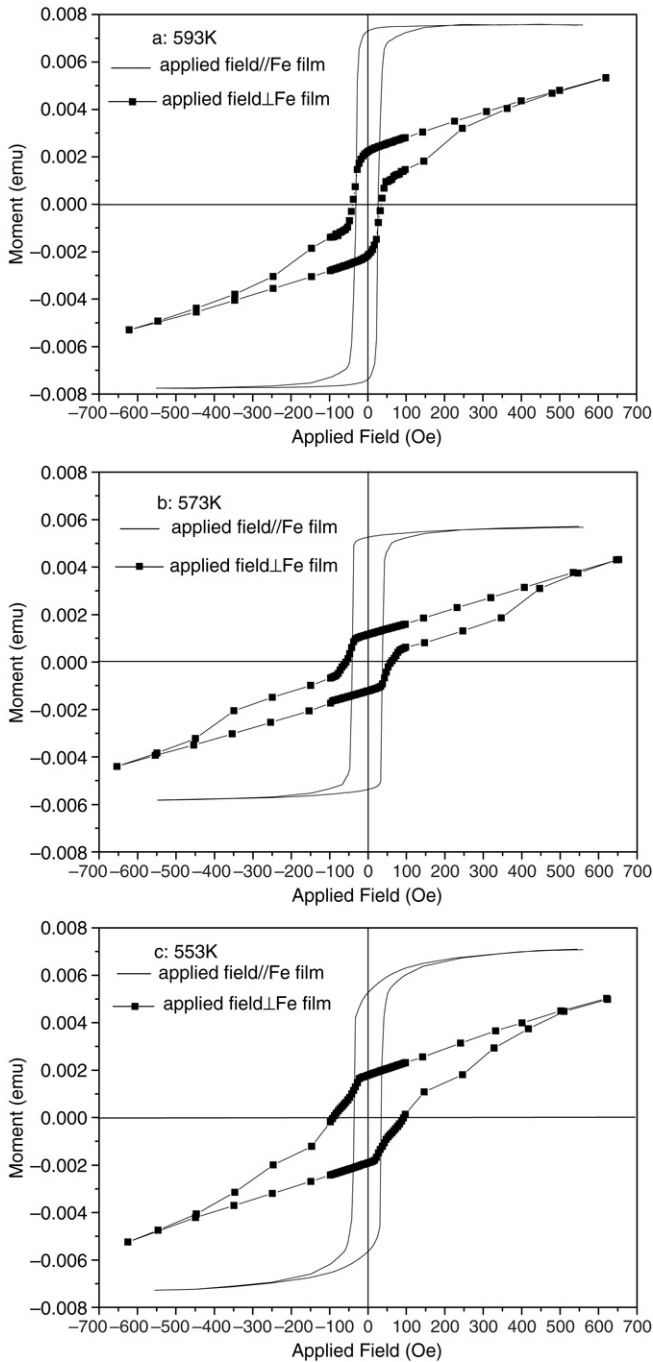


Fig. 4. Magnetization vs. applied field along and perpendicular to the surface of the films grown at different temperatures: (a) 593 K; (b) 573 K; (c) 553 K.

state to the paramagnetic state. Thus, samples with larger grains will present a larger saturation magnetization (M_s) at low magnetic field. Similarly, a smaller coercivity (H_c) value will also be expected in samples with larger grains due to the easier inversion for larger magnetic domains. It is also found that the ratio between the remnant magnetization and saturation magnetization (M_r/M_s) increased from 0.76 to 0.97 with the increase of the growth temperature (see the inset of Fig. 5), when the applied field is parallel to the film plane.

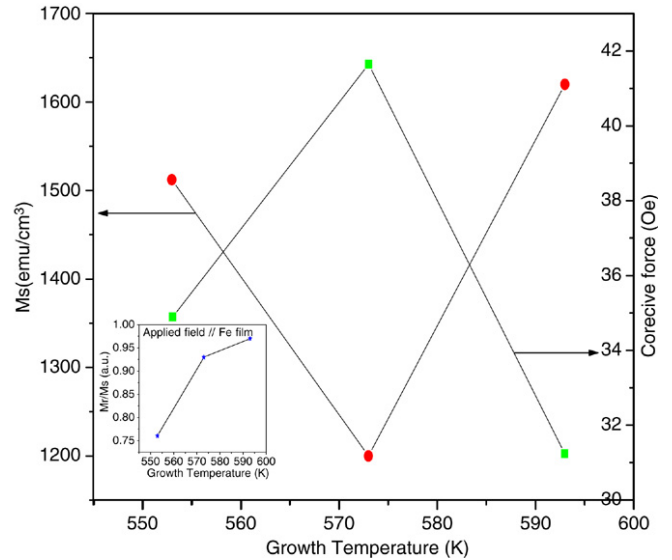


Fig. 5. The growth temperature dependence of M_s and coercivity measured at room temperature, when the applied field was parallel to the sample surface. The inset shows the growth-temperature dependence of M_r/M_s .

4. Conclusion

Fe films with strong preferred orientation (001) were prepared on GaAs (001) substrates by LP-MOCVD with the pressure of 150 Torr at different temperatures. The surface roughness of Fe films decreases with increasing growth temperature. Magnetization measurements showed that there were remarkable differences between in-plane and out-of-plane magnetic properties of Fe films. The sample prepared at 593 K has the biggest M_s and M_r/M_s ratio but the lowest H_c due to its high crystal quality. Furthermore, when the applied magnetic field is perpendicular to the Fe surface, one can clearly observe a sharp jump in the hysteresis loop, followed by a broad shoulder. It is considered that the broad shoulder may be caused by the interface effect, the existence of carbon and the formation of $180^\circ/90^\circ$ magnetic domains. The detailed reasons need to be further studied.

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