Thickness dependence of magnetic state of Fe thin films grown on Al$_2$O$_3$(0001) substrates with an inclined angle

Yu Shiratsuchi$^a$, Yasushi Endo$^{a,b}$, Masahiko Yamamoto$^{a,b,*}$

$^a$Department of Materials Science and Engineering, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

$^b$Frontier Research Center, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

Received 27 August 2003; revised 19 September 2003; accepted 26 September 2003

Abstract

We have investigated the magnetic state of Fe thin films grown on Al$_2$O$_3$(0001) inclined substrates at the Fe thickness range of 5–25 ML (monolayer) (1.0–5.0 nm). For 5 ML Fe films, the magnetic state is superparamagnetic state and the effect of inclination of substrate is negligible. The magnetic state of 10 ML Fe films is in the coexistence of superparamagnetic state and ferromagnetic state, and the effect of inclination of substrate appears but is still a little. In the ferromagnetic region, above Fe thickness of 15 ML, the magnetic easy axis is influenced by the inclination of substrate. Fe thin films grown on the flat substrate have no preferred direction of magnetization in the film plane because of the three equivalent variants of the epitaxial Fe(110) on Al$_2$O$_3$(0001). On the other hand, Fe thin films grown on the inclined substrate have a uniaxial magnetic anisotropy, in which the magnetic easy axis is parallel to the step edge. The strength of uniaxial magnetic anisotropy decreases with increasing Fe thickness. This uniaxial magnetic anisotropy might be derived from the effective demagnetization field due to the magnetic charge distribution at the corrugated surface caused by the steps.

Keywords: Fe thin films; Al$_2$O$_3$; Inclined substrates; Superparamagnetism; Uniaxial magnetic anisotropy

1. Introduction

A rapid development of magnetic storage media has been witnessed during the last decade. Each element storing magnetic information must consist of ultrathin particles in order to yield further development. As the magnetic elements continue to decrease in size and approach the ultrathin region, the magnetization of the elements is influenced by the thermal fluctuation and finally the elements are in superparamagnetic state [1]. Thus, the understandings of the superparamagnetism and the critical size for the magnetic particles to keep the ferromagnetic state at room temperature are the key issues for the magnetic storage. In this study, we have investigated the thickness dependence of magnetic state of Fe thin films. At the thickness region studied here (5–25 ML (monolayer)), the magnetic state of Fe thin films developed from superparamagnetic state to ferromagnetic state via the coexistence of these two states.

The suppression of superparamagnetism is also the crucial problem, and some ideas to suppress the superparamagnetism are proposed [2,3]. The common concept to all proposals is the enhancement of the effective magnetic anisotropy, $K_{\text{eff}}$. One method to enhance $K_{\text{eff}}$ is to use the materials which possess the high crystalline magnetic anisotropy such as L$_1$$_0$-type FePt [2,4]. Another is to induce the additional magnetic anisotropy, for example, surface magnetic anisotropy [5–7]. Among the various methods to induce the magnetic anisotropy [5–9], we have investigated the use of an inclined substrate. In particular, we have used the oxide substrate. This is because the interface between a magnetic material and an oxide can possess a high interfacial magnetic anisotropy [3,7]. In this paper, we demonstrate the possibility of utilizing the inclined substrate as the method to control the magnetic anisotropy. Further, we will discuss the origin of magnetic anisotropy induced by the inclined substrate.
2. Experimental procedures

Fe thin films are prepared by molecular beam epitaxy (MBE) using our VG-80M MBE system. The pressure before and during the deposition is typically below $5 \times 10^{-9}$ and $2 \times 10^{-8}$ Pa, respectively. The growth rate is 0.005 nm/s and the thickness of Fe is varied from 5 to 25 ML. The growth temperature is fixed at 323 K. To investigate the magnetic properties, the Fe films are exposed to air. To avoid the oxidation, a 10 nm thick Au capping layer was deposited at room temperature before exposing to air. We confirmed the lack of oxidation even for 5 ML Fe indirectly from the fact that the magnetization curves at 10 K show no shift after cooling in magnetic field of 10 kOe. α-Al$_2$O$_3$(0001) was used as the substrate because it has atomically flat terraces. In this study, we used two types of substrate to control the structure of Fe films. One is the nominally flat substrate which has 0.216 nm height steps and 129.5 nm width terraces on the average (Fig. 1(a)). Another is the inclined substrate of which the inclined angle is 4°. The direction is ⟨1120⟩. The substrates inclined to this direction have the straight steps (Fig. 1(b)). On the inclined substrate, several steps with average height of 6.06 nm and terraces with average width of 65.5 nm are formed. These steps and terraces can be formed by the suitable thermal treatment. The detailed preparation procedure of the substrates is described in Ref. [10].

The magnetic properties were investigated by means of a magneto-optic Kerr effect (MOKE), a vibrating sample magnetometer (VSM), and a superconducting quantum interference device (SQUID) magnetometer. The magnetization curves were measured using MOKE or VSM at room temperature. The measurements are performed in two configurations in which the magnetic field is parallel and perpendicular to the film plane. The temperature dependence of the magnetization $M(T)$ was measured using SQUID magnetometer at the range of 10–300 K in a constant magnetic field. Changes are measured while heating process after field cooling (FC) and zero-field cooling (ZFC). If the system is in a superparamagnetic state, it should show the blocking phenomena in the magnetization. The blocking temperature was determined as the peak temperature of the $M(T)$ curve after ZFC.

The surface structure of the Fe films was investigated using non-contact atomic force microscopy (NC-AFM). The investigation of surface structure was performed in situ before Au coating to eliminate the influence of the Au capping layer.

3. Results and discussion

3.1. Transition from superparamagnetism to ferromagnetism

Fig. 2 shows the magnetization curves for Fe thin films in the applied magnetic field parallel to the film plane. The inclined angle of substrate is (a)–(c) 0 (flat) and (d)–(f) 4°. Fe thickness is (a), (d) 5, (b), (e) 10, and (c), (f) 15 ML, respectively. The magnetic field is applied parallel to ⟨1100⟩ direction of α-Al$_2$O$_3$ (0001) substrate. The magnetization curves for 5 ML Fe do not saturate at the maximum magnetic field of 4.5 kOe. These magnetization curves can be reproduced by Langevin function. Besides, the magnetization changes with the temperature for 5 ML Fe show the blocking phenomena. These two features show that 5 ML Fe films are in superparamagnetic state. Further, in our previous paper [11], we reported the superparamagnetic behavior of ultrathin (5 ML) Fe films and clarified that the dominant factor of superparamagnetic behavior is continuously changed with the growth temperature. The details are beyond the focus of this paper and referred to elsewhere [11]. As the Fe thickness increases, the magnetic state of the film develops to ferromagnetic state. 10 ML Fe films are in transition region from superparamagnetic state to ferromagnetic state. As shown in Fig. 2(b) and (e), although the magnetization curves are hard to saturate, they show the remanence and coercivity. This means that magnetic state of 10 ML Fe is the coexistence of superparamagnetic state and ferromagnetic state. 15 ML Fe films are
completely in the ferromagnetic state as shown in Fig. 2(c) and (f). The magnetic anisotropy for ferromagnetic Fe thin films is described below. Here, we describe the coexistence state of superparamagnetic and ferromagnetic from another experimental result. Fig. 3 shows the magnetization changes with the temperature for 10 ML Fe films. The inclined angle of substrate is (a) 0 (flat), and (b) 4°. For both cases, although $M(T)$ after FC and ZFC differ from each other, the magnetization at the higher temperature than the blocking temperature does not decrease with increasing temperature. This implies that 10 ML Fe films are not only in pure superparamagnetic state but ferromagnetic components exists in the films. The ratio of superparamagnetic and ferromagnetic Fe should be dependent on the inclined angle of substrate. The Fe film grown on the inclined substrate has more ferromagnetic term than the Fe films on the flat substrate. This is confirmed from the point that the normalized magnetization value at 10 K after ZFC for Fe grown on the inclined substrate is larger than that for Fe grown on flat substrate; the value is 0.9 for Fe on the inclined substrate, while it is 0.2 for Fe on the flat substrate. The ratio is also dependent on the growth temperature. The growth temperature dependence will be reported in the near future.

3.2. Magnetic anisotropy induced by the inclined substrate

The magnetic anisotropy for ferromagnetic Fe films is described in this section. Above Fe thickness of 15 ML, Fe thin films are in ferromagnetic state. The magnetization curves in the magnetic field perpendicular to the film plane saturate harder than those parallel to the film plane. This means that the magnetization lies almost in the film plane for all studied Fe films. In the following, we describe the in-plane magnetic anisotropy. Magnetization
curves of 15 ML Fe films, in which the applied magnetic field is (1100) and (1120) of Al$_2$O$_3$ substrate, are shown in Fig. 4. Each direction of applied magnetic fields is corresponding to the direction parallel or perpendicular to the step edge in case of the inclined substrates. As shown in Fig. 4(a), two magnetization curves are identical for Fe thin films grown on the flat substrates. This means that Fe thin films grown on the flat substrate have no preferred directions of magnetization in the film plane. This is due to the three equivalent variants of the epitaxial Fe(110) on Al$_2$O$_3$(0001). On the other hand, Fe thin films grown on the inclined substrate have a uniaxial magnetic anisotropy as shown in Fig. 4(b). The magnetic easy axis is parallel to the step edge. This uniaxial magnetic anisotropy also appeared in 10 ML Fe film, which is in the transition region from superparamagnetic state to ferromagnetic state, as well as 20, 25 ML Fe films.

Fig. 5 shows the thickness dependence of uniaxial magnetic anisotropy energy for Fe thin films grown on the inclined substrates. The anisotropy energy, $K_u$, is estimated from the Eq. (1)

$$K_u = (\int_{M_t} M H dM)_{\text{hard}} - (\int_{M_t} M H dM)_{\text{easy}}, \quad (1)$$

where $M_t$ and $M_s$ are remanence and saturation magnetization, respectively. The uniaxial magnetic anisotropy energy decreases exponentially with increasing Fe thickness.

In order to learn the origin of in-plane magnetic anisotropy, the surface structures were investigated. Fig. 6 shows NC-AFM images of Fe thin films grown on the flat substrates. The thickness of Fe is (a) 10, (b) 15 ML, respectively. Fe has spherical shape in-plane below the thickness of 10 ML, and develops to the ellipsoidal shape at 15 ML. Further, the directions of long axis of ellipsoids can be observed in three directions as shown by the arrows. This means that three equivalent variants of Fe(110) exist in the film plane. This epitaxial relationship is shown schematically in Fig. 6(c) and is confirmed by RHEED pattern. As mentioned above, Fe thin films grown on the flat substrate have no preferred direction of magnetization in the film plane. According to Gradmann et al. [12], no uniaxial magnetic anisotropy can exist if a film has in-plane $n$-fold symmetry with $n > 2$. This result is in coincidence with the in-plane identical magnetization curves as shown in Fig. 4(a).

On the other hand, Fe thin films grown on the inclined substrates have a uniaxial magnetic anisotropy. As mentioned above, Fe films grown on Al$_2$O$_3$(0001) have no preferred direction of the magnetization. Thus, the uniaxial magnetic anisotropy is the resultant feature of the substrate inclination. Some origins of the uniaxial magnetic anisotropy for the magnetic thin films grown on the inclined substrate can be considered, i.e. the shape anisotropy due to the wire structure, the step induced magnetic anisotropy [13–15], the magnetostatic interactions [16] and so on. In this paper, we discuss the origin of uniaxial magnetic
anisotropy in view of the surface structure and magnetostatic interactions. Fig. 7 shows the surface structure of 10 ML Fe films grown on the inclined substrate. Although Fe forms the particles on the terrace, the particles attach together and forms continuous film. Further, considering the surface structure of inclined substrate (Fig. 1(b)) and Fe film (Fig. 7(a) and (b-1)), the Fe film keeps the morphology of inclined substrate. However, a part of Fe grows selectively at the under part of the step as shown by the arrows (Fig. 7(b-2)). Although the wire structure can be formed by such selective growth, Fe should not form the wire at the step edge since the size along the step edge of such Fe is not so long (Fig. 7(b-3)). Thus, we consider that Fe forms the continuous film on the whole inclined substrate and the origin of uniaxial magnetic anisotropy is not the shape anisotropy due to wire structure but the magnetostatic interactions due to the magnetic charge distribution at the surface caused by the regularly aligned steps. Another reason for eliminating the shape anisotropy due to the wire structure is the decrease of $K_u$ with Fe thickness. If the uniaxial magnetic anisotropy is derived from the shape anisotropy due to the wire structure, it is expected that $K_u$ should be constant or not strongly correlate with the Fe thickness. The schematic representation of the magnetic charge distribution in the continuous film on the inclined substrate, when the magnetization is saturated perpendicular to the step, is shown in Fig. 8. The magnetic charge distribution is determined by the topological corrugation. In such a complex shape, the demagnetizing field cannot be calculated analytically. However, we can assume that the demagnetizing field is localized around the surface and the effect of demagnetizing field decreases as the thickness.
increases. Further, in such a system as the corrugated surface, the exponential decrease in experimentally obtained uniaxial magnetic anisotropy is reported in Ref. [16] in spite of the lack of theoretical background. In our case, the experimentally obtained $K_u$ decreases with increasing Fe thickness, and can be fitted with the Eq. (2)

$$K_u = K_{u0} \exp\left(-t/t_0\right),$$

where $K_{u0}$ and $t_0$ represent the amplitude and decay for the exponential function, respectively. Fitting result is also shown in Fig. 5 as a solid line. A good agreement is found between the experimental and calculated data. The obtained value of $K_{u0}$ and $t_0$ are $2.0 \pm 0.3 \times 10^5$ J/m$^3$ and $1.0 \pm 0.1$ nm, respectively. As mentioned above, although the exponential decrease in $K_u$ has no theoretical background, the obtained $K_{u0}$ value is large compared to the magnetocrystalline anisotropy, $K_1$ for bulk $\alpha$-Fe $4.8 \times 10^5$ J/m$^3$ [17].

Thus, the use of inclined substrate should have the possibility to enhance the effective magnetic anisotropy in ultrathin magnetic films.

4. Conclusion

We have investigated the thickness dependence of magnetic state of ultrathin Fe films grown on Al$_2$O$_3$(0001) substrates. 5 ML Fe films are in superparamagnetic state and the superparamagnetic behaviors are dominated by the interparticle interactions. 10 ML Fe films are in the coexistence state of superparamagnetic and ferromagnetic. Although the effect of inclination of substrate is negligible in the superparamagnetic term, it appears remarkably in the ferromagnetic term. Above Fe thickness of 15 ML, the magnetic state of Fe is ferromagnetic state. Fe thin films grown on Al$_2$O$_3$ (0001) flat substrates have no preferred direction of the magnetization in the film plane. On the other hand, Fe thin films grown on Al$_2$O$_3$ (0001) inclined substrates have a uniaxial magnetic anisotropy. This uniaxial magnetic anisotropy should be derived from the effective demagnetizing field due to the magnetic charge distribution at the corrugated surface caused by the steps on the inclined substrate.

Acknowledgements

This work is partially supported by a Grant-in-Aid for General Scientific Research (S) and Exploratory Research from the Japanese Ministry of Education, Culture, Sports, Science and Technology. This work is also partially supported by ‘Priority Assistance of the Formation of Worldwide Renowned Centers of Research-The 21st Century COE Program (Project: Center of Excellence for Advanced Structural and Functional Materials Design)’ from the Japanese Ministry of Education, Sports, Culture, Science and Technology.

References

[1] R.C. O’Handley, Modern Magnetic Materials, Wiley-Interscience, New York, 2000, p. 306.
[2] K.R. Coffey, M.A. Parker, J.K. Howard, High anisotropy L1$_0$ thin films for longitudinal recording, IEEE Transactions on Magnetics 31 (1995) 2737.
[3] C.L. Chien, Granular magnetic solids, Journal of Applied Physics 69 (1991) 5267.
[4] Y. Endo, N. Kukuchi, O. Kitakami, Y. Shimada, Lowering of ordering temperature of fct Fe–Pt in Fe/Pt multilayers, Journal of Applied Physics 89 (2001) 7065.
[5] L. Neel, Anisotropie magnetique superficielle et superstructures d’orientation, Le Journal de Physique et le Radium 15 (1954) 225.
[6] P.F. Garcia, A.D. Meinhaldt, A. Sana, Perpendicular magnetic anisotropy in Pd/Co thin film layered structures, Applied Physics Letters 47 (1985) 178.
[7] J.W. Cai, S. Okamoto, O. Kitakami, Y. Shimada, Large coercivity and surface magnetic anisotropy in MgO/Co multilayer films, Physical Review B 63 (2001) 104418.
[8] W.H. Meskelejohm, C.P. Bean, New magnetic anisotropy, Physical Review 105 (1957) 904.
[9] M. Grimsditch, A. Hoffmann, P. Pavassori, H. Shi, D. Lederman, Exchange-induced anisotropies at ferromagnetic–antiferromagnetic interfaces above and below the Neel temperature, Physical Review Letters 90 (2003) 257201.
[10] Y. Shiratsuchi, M. Yamamoto, Y. Kamada, Surface structure of self-organized sapphire (0001) substrates with various inclined angles, Japanese Journal of Applied Physics 41 (2002) 5719.
[11] Y. Shiratsuchi, M. Yamamoto, Y. Endo, D. Li, S.D. Bader, Superparamagnetic behavior of ultrathin Fe films grown on Al$_2$O$_3$ (0001) substrates, submitted for publication.
[12] U. Gradmann, J. Koreck, G. Waller, In-plane anionic surface anisotropies in Fe(110), Applied Physics A 39 (1986) 101.
[13] A. Berger, U. Lincke, H.P. Oepen, Symmetry-induced anisotropy in ultrathin epitaxial cobalt films grown on Cu(1 1 1 3), Physical Review Letters 68 (1992) 839.
[14] H.J. Choi, Z.Q. Qiu, J. Pearson, J.S. Jiang, D. Li, S.D. Bader, Magnetic anisotropy of epitaxial Fe films grown on curved W(001) with a graded step density, Physical Review B 57 (1998) R12713.
[15] E.J. Escorcia-Aparicio, J.H. Wohfe, H.J. Choi, W.L. Ling, R.K. Kawakami, Z.Q. Qiu, Magnetic phases of thin Fe films grown on stepped Cr(001), Physical Review B 59 (1999) 11892.
[16] A. Encinas-Oropesa, F. Nguyen Van Dau, Origin of magnetic anisotropy in thin films deposited on step-bunched substrates, Journal of Magnetism and Magnetic Materials 256 (2003) 301.
[17] R.C. O’Handley, Modern Magnetic Materials, Wiley-Interscience, New York, 2000, p. 189.