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Magnetic phase transition and anisotropy of ultrathin Fe films grown on inclined $Al_2O_3(0001)$ substrates

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We investigated the magnetic properties of ultrathin Fe films grown on inclined $Al_2O_3(0001)$ substrates at various growth temperatures. We report the evolution of the magnetism with Fe thickness t_{Fe}, growth temperature, and the effect of the inclination of the substrate orientation on the magnetic anisotropy. The films are superparamagnetic ($t_{Fe}\approx 5$ monolayer, ML), ferromagnetic ($t_{Fe}>15$ ML), or coexistent ($t_{Fe}\approx 10$ ML). The effect of inclination of the substrate is small in the superparamagnetic region and substantial in the ferromagnetic region. Fe thin films grown on the inclined substrate have a uniaxial magnetic anisotropy with the magnetic easy axis parallel to the step edge. This uniaxial magnetic anisotropy might be derived from the effective demagnetizing field due to the magnetic charge distribution at the corrugated surface. The strength of the uniaxial magnetic anisotropy decreases as the growth temperature increases. The dependence of the uniaxial magnetic anisotropy on growth temperature is caused by the change of growth mechanism, from smooth to rough with an increasing of growth temperature.
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Superparamagnetism has attracted much attention from fundamental and technological points of view. From the fundamental viewpoint, the superparamagnetic behavior of ultrathin magnetic films is not understood because factors such as the interparticle interactions and the size distribution complicate the situation.[1,2] From the technological viewpoint, strong efforts have been devoted to the study on superparamagnetism in order to utilize magnetic nanostructures as high density storage media. As the density of magnetic storage media increases, the magnetic elements enter the nanometer region and approach the superparamagnetic limit. While a number of ideas to suppress the superparamagnetic limit have been proposed, the main one is to enhance the magnetic anisotropy energy, for example, via materials possessing high magnetocrystalline anisotropy,[3] or to induce additional magnetic anisotropy.[4] In this study, we investigate the magnetic state of ultrathin Fe films grown on inclined $Al_2O_3(0001)$ substrates, the influence of growth temperature on the magnetic state, and the effect of the inclination on the magnetic state. We also demonstrate the use of the inclined substrate as a way to induce additional magnetic anisotropy.

Ultrathin Fe films were prepared by molecular beam epitaxy (MBE) using a VG-80M MBE system. The pressure before and during the deposition was typically below 4×10^{-9} and 5×10^{-8} Pa, respectively. The nominal thickness of Fe was varied from 5 to 25 monolayers (ML) and the growth temperature was varied in the range 323–773 K. The surface structure of the Fe thin films was investigated *in situ* using noncontact atomic force microscopy (NC-AFM) and reflection high energy electron diffraction (RHEED). The magnetic properties were investigated *ex situ* by means of the magneto-optic Kerr effect (MOKE), vibrating sample magnetometry (VSM), and superconducting quantum interference device (SQUID) magnetometry. To avoid oxidation, a 10-nm Au capping layer was deposited at room temperature. We confirmed the lack of oxidation even for 5 ML Fe thin films indirectly from the fact that the magnetization curves at 10 K showed no bias after cooling in a magnetic field (10 kOe). Two types of α-$Al_2O_3(0001)$ substrates were used. One is nominally flat having 0.216 nm height steps and 129.5 nm width terraces on the average as reported previously.[5] The other is the inclined substrate with an angle of 4 degrees inclined in the $\langle 11\bar{2}0\rangle$ direction. Such inclined substrates have straight steps. On the inclined substrate, several bunched steps with an average height of 6.06 nm and terraces with an average width of 65.5 nm are formed by suitable thermal treatment.[5]

The magnetism of ultrathin Fe films evolves from superparamagnetic to ferromagnetic with increasing Fe thickness. 5-ML Fe films on the flat substrate are in the superparamagnetic, with behavior dependent on growth temperature.[2] The growth dependence of the blocking temperature (T_B) and volume of Fe particles (V_{Fe}) estimated from atomic force microscopy (AFM) images are shown in Fig. 1. T_B is defined as the peak temperature of the M-T curve after zero field cooling. While V_{Fe} monotonically increases with growth temperature, T_B has a minimum at the growth temperature of 473 K. The enhancement of T_B at low growth temperature is attributed to interarticle interactions. The growth temperature

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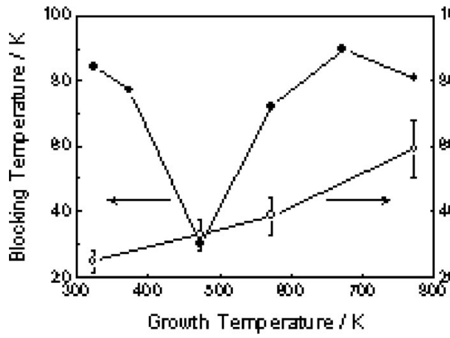

FIG. 1. Changes of the blocking temperature and estimated Fe volume with growth temperature for 5-ML Fe films. The blocking temperatures are determined from the peak temperature of the M-T curve after ZFC. The Fe volume is estimated from AFM images.

dependence of T_B with increasing growth temperature is attributed to the emergence of the nonlinear dependence of the magnetization with the magnetic field as the dominant factor.

The 10-ML Fe film exhibits coexistence of superparamagnetism (SP) and ferromagnetism (FM). In the following, we describe this state using two types of experimental results, i.e., the magnetization curves at room temperature (RT) [Fig. 2(a)] and the change of magnetization with temperature at a magnetic field of 100 Oe [Fig. 2(b)]. As shown in Fig. 2(a), while the magnetization curve is hard to saturate and indicates the existence of SP, the remanent magnetization and the coercivity are not zero, indicating the presence of FM. Furthermore, the change of magnetization with temperature after field cooling (FC) and zero field cooling (ZFC) differ in the low temperature region with the ZFC curve having a peak as shown in Fig. 2(b). These features show that 10-ML Fe films are not in the pure SP or FM state but a coexistent state of the two.

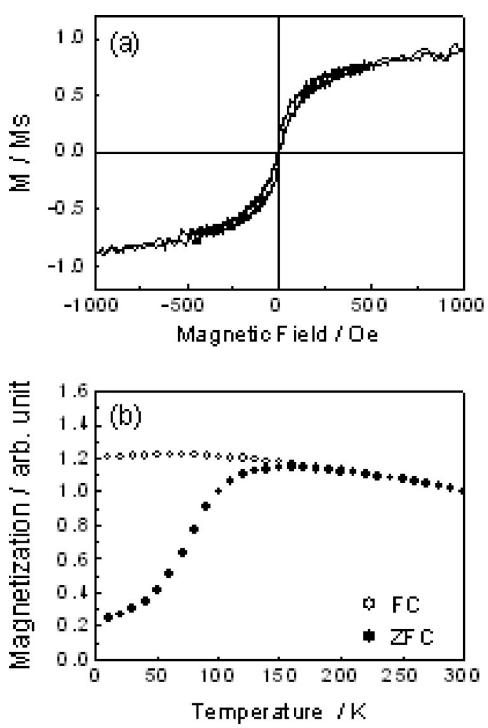

FIG. 2. (a) Magnetization curves and (b) changes of magnetization with temperature for 10-ML Fe films on flat $Al_2O_3(0001)$ substrate.

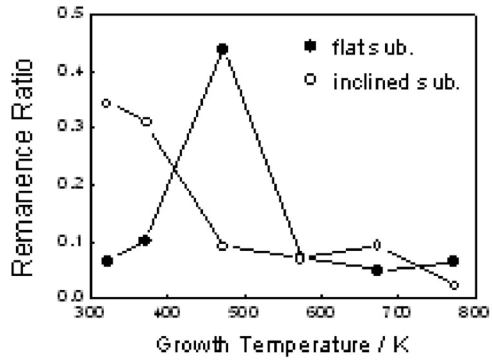

FIG. 3. Growth temperature dependence of remanence ratio for 10-ML Fe films. The remanence ratio is determined from the magnetization curve measured at RT. The applied magnetic field is parallel to $\langle 1\bar{1}00\rangle$ of the $Al_2O_3(0001)$ which is parallel to the step direction in case of the inclined substrate. The open (○) and closed (●) symbols represent the values for Fe thin films grown on the inclined and flat substrate, respectively.

Here we consider the ratio of the two magnetic states. Although quantitative determination of the ratio is difficult, we can consider the remanence ratio of the magnetization curve measured at RT as yielding the ratio of the two states, since the SP components do not show remanence at RT. Figure 3 shows the growth temperature dependence of the remanence ratio at RT. While the FM component is maximum at a growth temperature of 473 K for the Fe films grown on the flat substrate, it decreases monotonically with increasing growth temperature for Fe on the inclined substrate. This difference might be due to the change of growth mechanism of Fe on the flat and inclined substrates.

Another effect of substrate inclination appears in the magnetic anisotropy of the ferromagnetic components of 10-ML Fe films. The magnetic easy axis is influenced by the large steps on the inclined substrate. Fe thin films grown on the flat substrate have no preferred direction of magnetization in the film plane, because the three equivalent epitaxial Fe(110) orientations form on the $Al_2O_3(0001)$ substrate and no uniaxial magnetic anisotropy can exist if a film has in-plane n-fold symmetry with $n>2$.[6] The epitaxial relationship and three variants of Fe(110) were previously confirmed by RHEED pattern, and three kinds of ellipsoidal Fe(110) islands due to such epitaxial growth were observed by AFM.[7]

Fe thin films grown on the inclined substrate however do have uniaxial magnetic anisotropy parallel to the steps. Although various origins of uniaxial magnetic anisotropy have

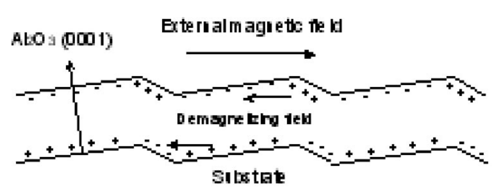

FIG. 4. Schematic representation of a continuous film on the inclined substrate and the magnetic charge distribution at the corrugated surface when the magnetization is saturated perpendicular to the step.

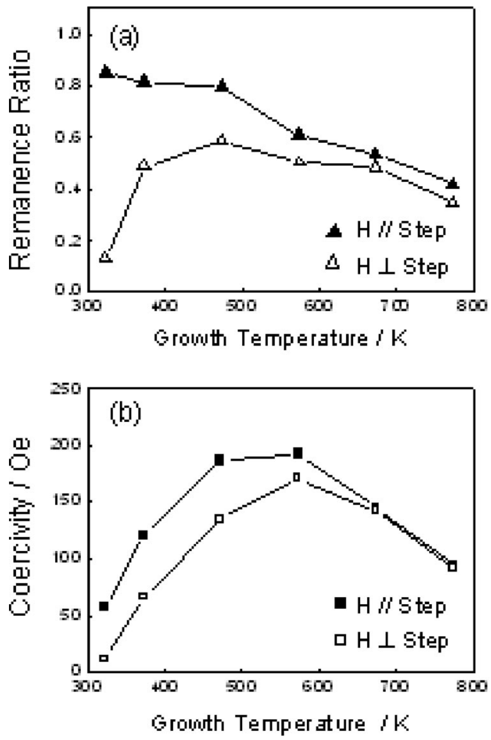

FIG. 5. Growth temperature dependence of the (a) remanence ratio and (b) coercivity for 20-ML Fe films on the inclined substrate. The closed (▲, ■) and open (△, □) symbols represent values for the magnetic field applied parallel and perpendicular to the step edge, respectively.

been proposed,[8–11] in our case it might be due to the effective demagnetizing field caused by the magnetic charge distribution at the corrugated surface, as shown in Fig. 4, since Fe films on the inclined substrate retain the morphology of the inclined substrate.[7] In such a case, the uniaxial anisotropy energy should decrease as the surface of magnetic film becomes rough. We utilized a change of growth temperature as a means to alter the surface roughness. Generally, a low (high) growth temperature should cause a smooth (rough) surface, respectively.[12] Figure 5 shows the growth temperature dependence of the remanence ratio and the coercivity of 20-ML Fe films grown on the inclined substrate. The remanence ratio decreases for the magnetic field applied parallel or perpendicular to the steps, go to similar values with increasing growth temperature. This means that the uniaxial magnetic anisotropy should decrease with increasing growth temperature. Also, the coercivity of Fe thin films have a maximum around the growth temperature of 573 K, implying that the Fe should form larger particles at higher growth temperature. These features show that the Fe films should become rough with increasing growth temperature.

In summary, we have investigated the evolution of the magnetic state with Fe thickness for ultrathin Fe films, and the induced magnetic anisotropy due to the inclination of the substrate. Magnetism evolves from superparamagnetic to ferromagnetic via a coexistence of the two states. In the transition region, the ratio of ferromagnetic and superparamagnetic components is dependent on the growth temperature and is also influenced by the inclination of the substrate. This is attributed to a change of growth mechanism caused by the large steps on the inclined substrate. The inclination of substrate influences the in-plane magnetic anisotropy and induces the uniaxial magnetic anisotropy.

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