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Effect of ion beam irradiation on magnetism of Fe(100) outermost surfaces studied by spin-polarized ion scattering spectroscopy

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ABSTRACT

We study the effect of 2 keV Ar^+ ion beam irradiation (IBI) on the outermost surface magnetism of an Fe(100) film by spin-polarized ion scattering spectroscopy (SP-ISS). We found that the coercivity of the outermost surface is enhanced with IBI. On the other hand, spin polarization is independent of IBI. These effects of IBI on surface magnetism are discussed in terms of morphology and atomic arrangement of the surface analyzed by ISS and reflection high-energy electron diffraction. The variation of coercivity with respect to the average iron film thickness d followed a power law d^{-n} with the assumption that d is linearly dependent on the IBI time.

1. Introduction

Local modification of surface magnetism by ion beam irradiation (IBI) has attracted increased attention in the past decade [1]. The target surface involves ultrathin films, multilayer films, and alloys. Magnetic patterning by IBI combined with a standard e-beam lithography technique has been well investigated [2,3]. Another approach for magnetic patterning of surfaces by IBI is topographic modification with self-assembly procedures. It is known that IBI with an appropriate incident condition can be used to form well-organized regular nanoscale patterns on the target surface [4]. The pattern formation has been partially attributed to the dependence of the sputtering yield on the surface curvature [5], although efforts are still in progress to achieve a fully quantitative description of the mechanism [6]. Since topographic nanopatterning of surfaces may help in the fabrication of high-density magnetic recording media, a number of studies have investigated the effect of morphology of thin films on their magnetism [7,8].

Previous research has demonstrated the control of parameters governing magnetic anisotropy of surfaces by IBI. For example, it has been shown that surface steps along the sidewalls of ripples created by IBI become the source of uniaxial anisotropy [9,10].The spin reorientation transition by IBI has been reported by several groups [11,12]. Moreover, ion induced-nonequivalent electronic hybridization between easy and hard axes has been claimed to induce uniaxial anisotropy on the irradiated film [13].

The above-mentioned effect of IBI has been typically discussed in terms of the magnetic hysteresis curve obtained using a vibrating sample magnetometer or the magneto-optical Kerr effect. Although the IBI effect on magnetism has been successfully observed using these techniques, the number of studies concerning the IBI effect on magnetism of outermost surfaces is very limited [14]. This is owing to the experimental difficulty that it is still not easy to analyze the magnetic property of outermost surfaces in the present time [7]. However, such analysis is needed for understanding the low-energy IBI effect on magnetism, because this effect essentially originates from the surface, as discussed later.

Recently, we reported the development of spin-polarized ion scattering spectroscopy (SP-ISS), a novel method for analyzing the magnetic structure of surfaces [15]. In SP-ISS, the electron spin-polarized $^4He^+$ ions are projected on sample surfaces, and the kinetic energy of the scattered ions is analyzed. The typical kinetic energy of the incident He^+ ions is of the order of keV, and therefore, most incident He^+ ions are neutralized at the sample surface. Since SP-ISS detects the scattered He^+ ions that survive this efficient neutralization at the surface, it has extreme surface sensitivity as conventional ISS [16]. Moreover, only the atoms located on the outermost surface layer can be selectively detected with the shadowing effect.

Neutralization of an incident He^+ ion typically occurs by the transition of a surface electron to the 1s hole of the incident He^+ ion (Auger neutralization (AN)) [16]. According to the Pauli exclusion principle, the scattered He^+ ion intensity should differ depending on whether the He^+ spin is parallel to majority or minority spins of the surface in the AN process. Moreover, the spin dependence of neutralization probability as a function of kinetic energy of scattered ions is expected to show element selectivity because the scattering

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energy correlates with the mass of target atoms. We have discussed these features of SP-ISS in the past few years [17–19].

In the present study, we investigated the IBI effect on magnetism of the Fe(100) outermost surface by SP-ISS. We observed for the first time that IBI enhances the coercivity of the outermost surface layer as reported in a number of publications for subsurfaces or bulk [3,20,21]. On the other hand, no substantial effect of IBI on the spin polarization was observed. These effects of IBI on the outermost surface magnetism are discussed in terms of surface structure. We successfully demonstrated that SP-ISS is a powerful method for element-selective magnetic hysteresis study on the outermost surfaces of soft magnetic materials.

2. Experimental method and setup

The experiments were performed in an ultrahigh vacuum chamber (base pressure of 5×10^{-11} Torr) equipped for SP-ISS and reflection high energy electron diffraction (RHEED). We grew Fe(100) films on MgO(100) single-crystalline substrates by vapor deposition of iron (purity 99.99%) using an electron beam evaporator (Omicron EFM3T) at room temperature, followed by annealing at about 700 K for 10 min. The deposition rate was about 1 nm/min. Under these growth conditions, bcc Fe films grew epitaxially with the orientation relation Fe(100)//MgO(100) and Fe[110]//MgO[100] [22]. The final thickness of the iron film was estimated to be about 100 nm.

In IBI, the Fe(100) sample was irradiated with Ar^+ ions at 2 keV with a fluence of 8×10^{12} ions cm^{-2}. This corresponds to the removal of one equivalent layer of Fe(100) by 1 min IBI according to the empirical formula for ion-induced sputtering of monatomic solids on normal incidence [23]. The incident angle measured from the surface normal was 30° and the incident direction was parallel to Fe(010). The bare MgO substrate surface finally appeared after IBI. The (SP-) ISS measurement became impossible immediately after this appearance of the MgO surface due to the charging effect. Thus, our study was limited to the iron film without pin holes induced by IBI.

Electron spin-polarized $^4He^+$ ions were generated from Penning ionization of spin-polarized metastable He 2^3S_1 atoms (He*) [24]:

$$He^* + He^* \rightarrow He^+ + He^0 + e^-.$$

We employed an optical pumping (OP) technique, using the D_0 line of 1083 nm radiation (He* $2^3S_1 \rightarrow 2^3P_0$), for spin polarization of He*. The spin angular momentum is conserved in Penning ionization; thus, if He* is spin polarized by OP, spin-polarized He^+ ions are generated [25]. The OP radiation was provided by an ytterbium-doped fiber laser (Keopsys KPS-BT2-YFL-1083-FA) via an ytterbium-doped fiber amplifier (Keopsys KPS-BT2-YFA-1083-200-COL). The wavelength of the 1083 nm seed light from the fiber laser was precisely tuned to the D_0 line. The spin directions of He^+ ions (up or down) were controlled by the polarization of OP radiation. The spin polarization of the He^+ ion beam P_{He+} was about 0.2.

The entire apparatus was surrounded by a three-axis coil to compensate for the Earthâ€™s magnetic field. An additional coil produced a weak guiding field (0.1 Oe), which was parallel to the vertical axis. Thus, the spin direction of the incident He^+ ion beam was defined by the guiding field.

The scattered He^+ ions were measured using a rotatable hemispherical sector analyzer (Omicron SHA50). The measurements were conducted in a constant pass energy mode with a pass energy of 318 eV. The scattering plane was parallel to the surface normal.

3. Results and discussion

Before discussing the magnetism on the outermost surfaces, the effect of IBI on the Fe(100) surface structure is discussed. The structure analysis is essential for discussing the origin of magnetism. We investigated the surface atomic arrangement by ISS and the surface periodicity and morphology by RHEED.

Fig. 1 shows the incident angle α scan of ISS on the Fe(100) surface (a) without IBI and (b) with IBI for 40 min. The scattering angle θ and the incident energy E_0 were 150° and 1780 eV, respectively. The scattering plane was parallel to Fe(001) as shown in the inset (c). The inset (d) shows the ISS spectrum on the Fe(100) surface without IBI. This spectrum was obtained with $\alpha = 0°$ and the exit angle $\beta = 30°$, where α (β)is defined as the angle between the incident (exit) direction and the surface normal. The peak at 1320 eV originates from the scattering from iron atoms and that at 20 eV is attributed to secondary ions.

The intensity variation without IBI, shown in Fig. 1 (a), exhibits a sharp increase at 72° (peak (i)). This is due to the focusing effect to the iron atoms located at the outermost surface atomic layer. The geometrical relationship between the atomic arrangement and the shadow cone is illustrated in Fig. 2, where the shadow cone was calculated using the Thomas–Fermi–Moliere potential [26]. By taking the critical angle of the focusing effect at an angle with 80% of maximum intensity [27], the angle of peak (i) is well explained by the bulk termination structure of the Fe(100) outermost surface. This is consistent with recent calculation on the atomic position of the Fe (100) surface [28]. Some humps at smaller *lpha* are due to the focusing effect to the iron atom at the subsurface atomic layer. The hump is enhanced in the α scan with lower energy (920–1100 eV) compared with iron peak energy (1240–1400 eV) as shown in Fig. 1 (b). In this low energy range, the scattered He^+ ions experience neutralization followed by re-ionization [29]. Since multiple collisions enhance the probability of re-ionization, the α scan with energy lower than the peak energy reflects the atomic arrangement in deeper atomic layers. In fact, the angles of peaks (ii)–(iv) are well explained by the bulk structure of iron as shown in Fig. 2. It is noted that there are small differences in the peak angle between Fig. 1 (a) and (b). This is due to the ion neutralization effect as follows.

If we assume that the scattered ion intensity I is proportional to the survival probability from ion neutralization, I is given by

$$I \propto \left(1-P_N^{in}\right)\left(1-P_N^{out}\right), \tag{1}$$

where P_N^{in} and P_N^{out} represent neutralization probabilities in the incoming and outgoing trajectories, respectively. This assumption is

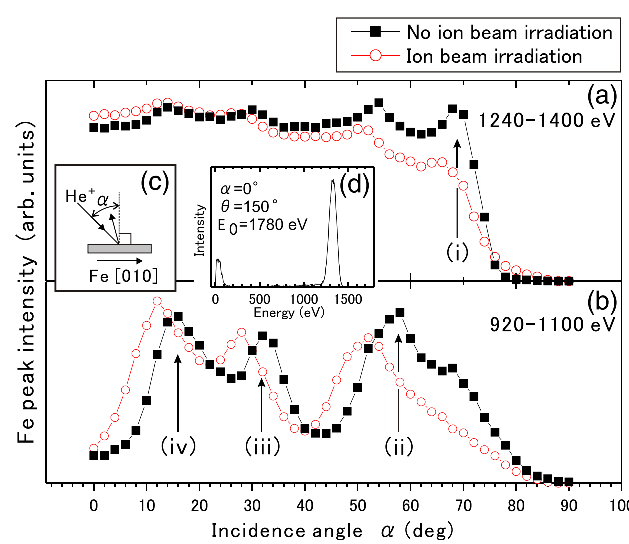

Fig. 1. α dependence of scattered He^+ ion intensity from an Fe(100) surface without IBI (solid black squares) and with IBI for 40 min. E_0 and θ were 1780 eV and 150°, respectively. Energy window is (a) 1240–1400 eV that corresponds to the iron peak; (b) 920–1100 eV that corresponds to the low-energy tail of the iron peak. (c) The scattering plane is parallel to Fe(001). (d) The ISS spectrum with $\alpha = 0°$.