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Oxygen-termination effect of the spin-dependent electronic states in FeCo/Rh(001) thin film

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The emergence of perpendicular magnetic anisotropy (PMA) in magnetic thin films is essential from a practical application standpoint, as it contributes to increasing the recording density of storage devices and reducing energy consumption. Many magnetic thin films exhibiting PMA, such as Co/Pt, Co/Pd, FePt, TbFeCo, and GdFeCo, have been extensively investigated. Among them, FeCo alloy films, which are free of rare-earth and noble-metal elements, are considered promising candidates exhibiting strong PMA. Burkert *et al.* predicted that the magnetic anisotropy energy may increase in FeCo alloys when the tetragonal distortion is applied [1]. Specifically, at a c/a ratio of approximately 1.20-1.25, the magnetic anisotropy energy exceeds 700-800 μ eV/atom, which is one or two orders of magnitude greater than that of pure Fe or Co. The emergence of strong PMA was experimentally confirmed in tetragonally distorted FeCo ultra-thin films grown on Rh(001) (c/a = 1.24) with a thickness of 13-15 monolayers (ML) [2].

Furthermore, the FeCo thin films exhibiting PMA could be useful as a target for out-of-plane spin component in very low energy electron diffraction (VLEED) spin detectors [3,4]. In the present VLEED spin detector, Fe(001) $p(1\times1)$ -O films, which exhibit in-plane magnetic anisotropy and can detect only in-plane spin components (P_x , P_y), are widely used. However, to observe all components of spin polarization (P_x , P_y , P_z) with the VLEED detectors, a ferromagnetic thin film target with PMA is required in addition to the conventional Fe(001) $p(1\times1)$ -O films. Moreover, Fe(001) $p(1\times1)$ -O has an oxygen-adsorbed overlayer that serves as a protection layer from further oxidization and contamination, extending its lifetime from hours to weeks (or even several months or more with flash annealing). Thus, if the preparation method of oxygen-adsorbed protection layer on FeCo films is established, the FeCo-O films may be usable for long-term use as the VLEED targets for the out-of-plane spin component.

In this study, we fabricated the oxygen-terminated FeCo films on a Rh(001) single-crystal substrate and investigated the spin-polarized electronic structures and their lifetime utilizing spin- and angle-resolved photoemission spectroscopy (SARPES).

The samples were fabricated by using two different methods. In the first method, after cleaning the Rh(001) substrate by cycles of 2 keV Ar⁺-ion sputtering and subsequent annealing at 1000 °C, FeCo was deposited at room temperature. Then, the FeCo film was exposed to approximately 30 L of oxygen and postannealed at 300° C for 5 min. As a result, the low energy electron diffraction (LEED) pattern shows not only the 1×1 spots but also new superlattice spots [Fig. 1(b)]. Furthermore, while a clear out-of-plane spin polarization was observed at the pristine FeCo thin film [Fig. 2(a)], the spin polarization was almost diminished after oxygen exposure [Fig. 2(b)], indicating the loss of PMA.

Next, we explored a second method in which oxygen was introduced prior to the FeCo deposition, aiming to induce oxygen segregation. Specifically, after cleaning the Rh(001) substrate, oxygen exposure was exposed \sim 30 L to the substrate, followed by FeCo deposition and post-annealing at 300° C for 5 min. In this method, we obtained clear 1×1 LEED spots [Fig. 1(c)] similar to that of the pristine FeCo film [Fig. 1(a)].

More importantly, we observed a distinct out-of-plane spin polarization [Fig. 2(c)]. These results indicate that the FeCo thin film successfully retained perpendicular magnetic anisotropy even after oxygen termination.

Finally, to evaluate the protection effect of oxygen termination, we monitored the spin-polarization of both pristine FeCo and oxygen-terminated FeCo thin films over approximately one week. Figure 3 shows the time-dependent spin-polarization. Here, we estimated the time constant for an attenuation of out-of-plane spin-polarization, *i.e.* lifetime, by fitting the experimental results with an exponential function. As a result, the time constant of the oxygen-terminated FeCo thin film was approximately 5.5 times longer than that of the pristine FeCo thin film. This finding suggests that, similar to $Fe(001)p(1\times1)-O$, the oxygen termination provides an effective surface protection effect for FeCo thin films.

Based on these results, oxygen-terminated FeCo thin films appear to be highly promising as targets for outof-plane spin component in VLEED spin detector.



FIGURE 1. LEED patterns of the pristine FeCo thin film (a) and the oxygen-exposed FeCo film fabricated by the methods I (b) and II (c) taken at 90 eV.



FIGURE 2. Out-of-plane spin-resolved photoemission spectra taken at 21.2 eV (He discharge lamp). The samples correspond to those in Fig. 1. A magnetic field was applied in the out-of-plane direction prior to the measurement. Red (blue) represents the majority (minority) spin, and green represents the out-of-plane spin polarization.



FIGURE 3. Comparison of the time constant for an attenuation of out-of-plane spin polarization. Red (blue) represents FeCo thin film (oxygen-terminated FeCo thin film).

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