Anatomy of Interfacial Spin-Orbit Coupling in Co/Pd Multilayers

Element-specific orbital magnetic moments and their anisotropies in perpendicularly magnetized Co/Pd multilayers are investigated using Co *L*-edge and Pd *M*-edge angle-dependent X-ray magnetic circular dichroism. We show that the orbital magnetic moments in Co are anisotropic, whereas those in Pd are isotropic. First-principles density-functional-theory calculations also suggest similar results through the Co/Pd interfacial proximity effects. We conclude that the orbital-resolved anatomy of Co/Pd interfaces reveals that the orbital moment anisotropy in Co and spin-flipped transition related to the quadrupoles in Pd are essential for the appearance of perpendicular magnetic anisotropy, yielding important guidelines for the design of *spin-orbitronic* materials.

Ultrathin Co/Pd multilayers are artificial nanomaterials that exhibit perpendicular magnetic anisotropy (PMA), due to the spin-orbit interactions, achieved through interfacial chemical bonding. Regarding applications, after the development of artificially synthesized PMA, researchers have pointed out the possibility of ultra-high density recording media [1]. Extensive studies have examined the interfaces of ultra-thin magnetic multilayers and nanostructures. Studies on Co atoms performed using X-ray magnetic circular dichroism (XMCD) have suggested the enhancement of orbital magnetic moments at the interfacial Co that is adjacent to Pd [2]. It is well known that the PMA emerges due to the cooperative effects between spin moments in 3*d* transition metals (TMs) and large spin-orbit interactions in nonmagnetic 4*d* or 5*d* TMs. Co/Pd interfaces and multilayers have also been employed to demonstrate the photo-induced precession of magnetization and the creation of skyrmions using the interfacial Dzyaloshinskii-Moriya interaction, and magnetization reversal using the spin-orbit torque phenomena. Despite these promising studies on Co/Pd interfaces, the interfacial PMA, including the anisotropic orbital magnetic moments, has not been fully understood for both Co and Pd sites. Bruno and van der Laan theoretically proposed an orbital moment anisotropy in 3*d* TMs within the second-order perturbation (the weak coupling) of the spin-orbit interaction for more than half-occupied electrons [3, 4]. However, in the case of strong spin-orbit coupling in 4*d* or 5*d* TMs, the validity of this perturbative formula has been debated. In order to study the mechanisms of PMA in Co/Pd multilayers, the orbital magnetic-moment contributions of each element have to be explicitly considered. However, it is challenging to study the anisotropy of the orbital magnetic moments of both Co and Pd elements using one specific experiment, due to the challenges in detecting the induced magnetic moments, and of Pd in particular.

For the interfacial Pd, the magnetic dipole contribution through the quadrupole interactions between dipoles is assessed quantitatively. We focus on the anisotropy of orbital moments at the Co/Pd interfaces using XMCD and first-principles density functional theory (DFT) calculations, which provide element- and layer-resolved contributions that reveal the mechanism of PMA. Here, we discuss the anisotropies of both spin and orbital moments of Co and Pd using angle-dependent XMCD data and DFT calculations.

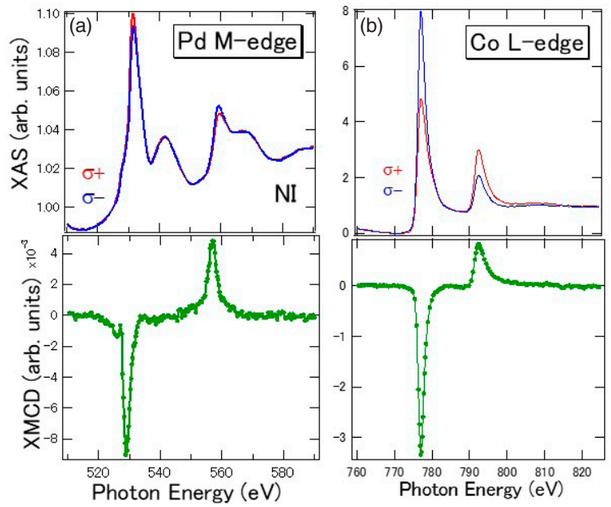

Figure 1: XAS and XMCD of **(a)** Pd *M*-edge and **(b)** Co *L*-edge in perpendicularly magnetized Co (0.69 nm)/Pd (1.62 nm) multilayer measured in the normal incidence (NI) configuration.

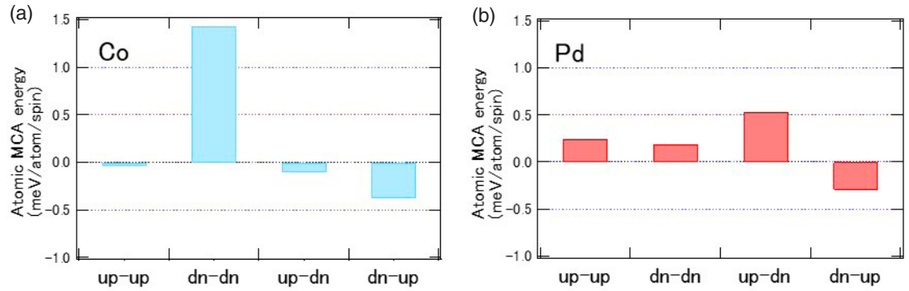

Figure 2: Bar graph of the second-order perturbative contribution of the spin-orbit interaction to the magneto-crystalline anisotropy (MCA) energy at the interfacial atomic sites of **(a)** Co and **(b)** Pd for the Pd (8 monolayer (ML))/Co (4 ML) deduced from a DFT calculation, using in-plane lattice constant $a_{ll} = 0.391$ nm.

We prepared two kinds of samples of Co/Pd multilayered structures: Co (0.69 nm)/Pd (1.62 nm) for PMA and Co (1.03 nm)/Pd (1.62 nm) for in-plane anisotropy with stacking of five periods on the Si substrates [5]. Sample surfaces were sputtered by Ar ions before the XMCD measurements in order to remove the oxygen contamination. We performed XMCD experiments at BL-7A. Total electron yield mode was adopted. A magnetic field of ±1.2 T was applied along the direction of the incident polarized soft X-ray.

We successfully observed clear XMCD signals in Pd *M*-edges after the removal of surface contamination, as shown in Fig. 1. Although the XAS line shapes overlap with those of O *K*-edge absorption, clear XMCD signals induced by the proximity with Co layers are observed due to the proper surface cleaning. The Pd *M*-edge XMCD line shapes in both PMA and in-plane anisotropy samples are almost identical, which suggests isotropic orbital moments in Pd, within the detection limits. This indicates that the isotropic finite orbital moments in Pd do not directly contribute to the PMA. On the other hand, for the PMA sample, clear Co *L*-edge XAS and XMCD with angular dependence reveal the enhancement of orbital moments in the surface normal direction because of PMA.

Next, we discuss the quadrupole-like contribution of the interfacial Pd layer from the results of angular dependent XMCD. Our Pd XMCD results indicate that the Pd orbital moments induced at the interface are isotropic. Note that the magnetic dipole term m_T is an order of magnitude smaller than the orbital moments, i.e., 0.005 μ_B, comparable with the detectable limits. Therefore, the relatively large spin-orbit coupling constant and small Pd exchange splitting contribute to the appearance of PMA by means of quadrupole-like interactions through the interfacial proximity effects.

Figure 2 represents the contributions of the crystalline magnetic anisotropy at each atomic site by DFT calculations. Four types of spin transition processes occur between the occupied and unoccupied states within the second-order perturbation of the spin-orbit interaction. For Co sites, the transition between down-down spin states is dominant, suggesting the conservation of spin states in the transition, which can be explained using the Bruno model assuming a large spin splitting. In contrast, for Pd, the spin-flipped transitions between up-down and down-up states become dominant due to the small band splitting, hence both spin-preserved and spin-flipped processes occur near the Fermi level. These results are consistent with the angular-dependent XMCD in Co and Pd sites.

In summary, we investigated the orbital-resolved interfacial electronic structures in the Co/Pd system and estimated the spin and orbital moments and magnetic dipole contribution [6].

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