

X-ray magnetic circular dichroism spectroscopy for voltage-controlled magnetic anisotropy in FePt/MgO multilayer system

An electric field at a surface is known to exhibit useful phenomena, such as the pinch-off phenomenon in field-effect transistors and the induction of Mott transitions, superconducting phases, ferromagnetic phases, and magnetic anisotropy. All of these phenomena are attributable to one or both of two factors: electron accumulation or electric dipole induction. In particular, electric-field control of magnetic properties at room temperature has attracted much attention because of its great potential for enabling the construction of ultralow-power-consumption electric devices. Voltage-controlled magnetic anisotropy (VCMA) in Fe/MgO-based tunnel junctions [1] has shown that the magnetization of nanomagnets can be controlled in fast periods (down to 0.1 ns) by electric fields, as indicated by bistable precessional magnetization switching [2] and ferromagnetic resonance excitation [3]. Because the microscopic origin of VCMA should be purely electronic, the VCMA effect can be an ultimate technology for the operation of spintronics devices, such as nonvolatile random access memory, where high-speed operation with high writing endurance is indispensable.

In this research [4], we showed a mechanism to enhance electronic VCMA proved by X-ray magnetic circular dichroism (XMCD) spectroscopy at SPing-8 BL39XU and BL25SU beamlines. We prepared monatomic layers of Pt at an Fe-MgO interface by

molecular-beam epitaxy in an ultrahigh vacuum as shown in Fig. 1. Since the Pt atoms have a considerable amount of spin polarization because of their hybridization with Fe and large spin-orbit interactions, the influence of the modified electron orbitals of Pt on the level of magnetocrystalline anisotropy energy (MAE) should be significant.

An L_{10} -FePt/MgO system was prepared to conduct XMCD spectroscopy as shown in Fig. 1. X-ray absorption spectroscopy (XAS) and its XMCD spectra were recorded *in situ* at the Pt L_3 and L_2 edges by detecting the X-ray fluorescence yields. In this configuration, positive external voltages induce electron accumulation at the Pt-MgO interface. As shown in Fig. 2, a clear VCMA effect, that is, a change in the magnetization hysteresis curve, was observed. Then, conventional sum-rule analysis can characterize the magnetic moments from XAS/XMCD spectra at the Pt absorption edge. The results confirm clear external voltage induction of the effective spin magnetic moment $m_S - 7m_T$ and an increase in the number of hole in Pt. While voltage application does not contribute a significant difference to the orbital magnetic moment m_L , the effective spin magnetic moment increased by 13%, and the hole number of Pt in the $5d$ -orbital increased by 0.019 when the external voltage was switched from positive to negative. From the magnetization angle dependence, we conclude that the voltage-induced changes in the effective spin magnetic moment originate from voltage-induced changes in the magnetic dipole T_z term m_T in the Pt atom, which has been revealed by high-precision XAS/XMCD measurements in SPing-8.

Equation 1 shows the perpendicular MAE (ΔE) when the spin-orbit interaction is treated in the second order [5] to discuss the MAE.

$$\Delta E \cong \frac{\lambda'}{4\mu_B} (\Delta m_{L,\downarrow} - \Delta m_{L,\uparrow}) - \frac{21}{2\mu_B} \frac{\lambda'^2}{E_{\text{ex}}} \Delta m_T \quad (1)$$

$\Delta m_{L,s}$ ($= m_{L,s}^{\perp} - m_{L,s}^{\parallel}$) and Δm_T ($= m_T^{\perp} - m_T^{\parallel}$) express the changes in the orbital magnetic moment and the magnetic dipole moment between the perpendicularly (\perp) and in-plane (\parallel) magnetized electronic states, respectively. Here $\Delta m_{L,\downarrow(\uparrow)}$ expresses the contribution from the minority (majority) spin band. The measured orbital magnetic moment equals $m_L^{\perp(\parallel)} = \Delta m_{L,\downarrow}^{\perp(\parallel)} + \Delta m_{L,\uparrow}^{\perp(\parallel)}$. In this case, λ' is the effective spin-orbit interaction coefficient in the d -band.

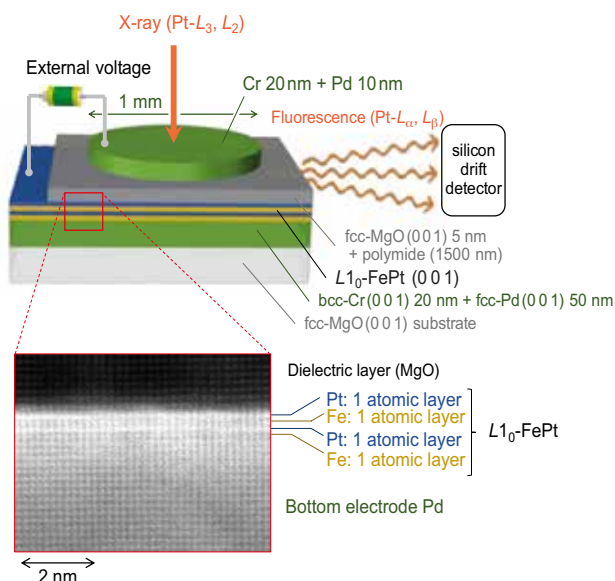


Fig. 1. Schematic of sample structure and experimental design.

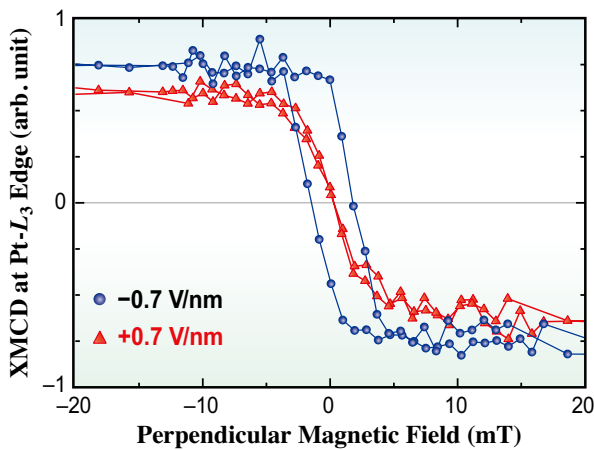
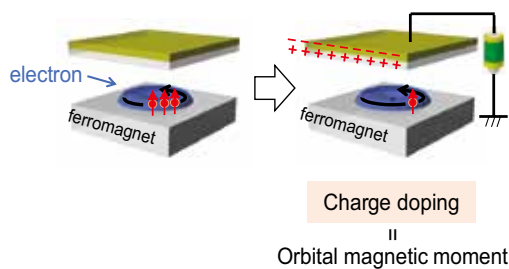


Fig. 2. Voltage-controlled magnetic anisotropy in FePt/MgO multilayer.

The first term is the MAE related to the orbital magnetic moment. When the majority spin band is full, $m_{L,\uparrow}^{(l)}$ can be neglected. Then, the perpendicular MAE is proportional to the measured changes in the orbital magnetic moment; this relation is known as Bruno’s model. This mechanism originates from the charge doping inducing anisotropy in the orbital magnetic moment as shown in Fig. 3(a). Note that $m_{L,\uparrow}^{(l)}$ cannot be neglected in the case of Pt because of the small size of the exchange splitting.

(a) Orbital magnetic moment mechanism (Bruno model)



(b) Electric quadrupole mechanism

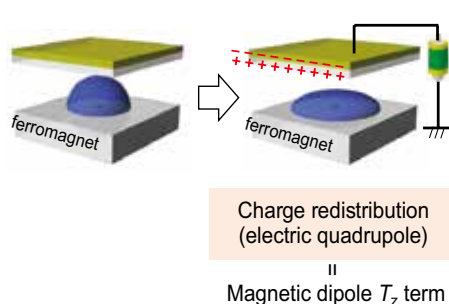


Fig. 3. Microscopic origin of VCMA in ultrathin ferromagnetic metals.

The second term represents the perpendicular MAE related to the changes in the magnetic dipole T_z term m_T . It accounts for the spin-flip excitations between the exchange-split (E_{ex}) majority and minority spin bands. Because an electric field applied to a metal/dielectric is inhomogeneous because of the strong electrostatic screening effect in metals, such an electric field, including higher-order quadratic components, can couple with the electric quadrupole correlated to the magnetic dipole T_z term. The induced energy splitting of each orbital changes the MAE through spin-flip excitation, as shown by the second term in Eq. 1 and in Fig. 3(b).

From our first-principles study, the VCMA effect in the FePt/MgO system mainly originates from the second term in Eq. 1 for the Pt atom, which corresponds to the experimentally observed changes in the magnetic dipole T_z term m_T . However, in our FePt/MgO system, the VCMA from the orbital magnetic moment and the electric quadrupole partially cancel one another. It should be noted that the VCMA from the electric quadrupole mechanism is larger than that of the orbital magnetic moment. However, the total VCMA is only 9% of the VCMA from the electric quadrupole mechanism. If a material with both VCMA having the same polarity is to be designed, an electronic VCMA larger by more than a factor of 10 would be feasible [4,5].

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References

[1] T. Maruyama *et al.*: Nat. Nanotechnol. **4** (2009) 158.
 [2] Y. Shiota *et al.*: Nat. Mater. **11** (2012) 39.
 [3] T. Nozaki *et al.*: Nat. Phys. **8** (2012) 492.
 [4] S. Miwa, M. Suzuki, M. Tsujikawa, K. Matsuda, T. Nozaki, K. Tanaka, T. Tsukahara, K. Nawaoka, M. Goto, Y. Kotani, T. Ohkubo, F. Bonell, E. Tamura, K. Hono, T. Nakamura, M. Shirai, S. Yuasa and Y. Suzuki: Nat. Commun. **8** (2017) 15848.
 [5] G. van der Laan: J. Phys. Condens. Matter **10** (1998) 3239.