Probing spin-polarized electronic structures of half-metallic Heusler alloys using resonant inelastic soft X-ray scattering in a magnetic field

Toward the next-generation spintronics application using the spin degrees of freedom, it is essential to optimize the spin-polarized electronic structures of magnetic materials under *operando* conditions in magnetic fields. In particular, the spin polarization in the vicinity of the Fermi level (E_F) for the conducting electrons and the size of the spin-dependent band gap are key parameters to designing functional devices. However, it was not straightforward to probe the spin-polarized electronic structures of the buried magnetic systems using the *standard* spin-polarized photoemission technique, which is rather surface-sensitive, since the magnetic layers are usually covered by capping layers in the device.

Resonant inelastic X-ray scattering (RIXS) is a photon-in/photon-out spectroscopy technique using a high-brilliance synchrotron light source to probe the electronic structures of any kind of specimens such as metal, insulator, liquid, and gas phases in an element-specific manner by tuning the incoming photon energy to the core-level absorption edges. Moreover, it is possible to apply any external perturbation, such as a magnetic field, during the measurements. This gives the great advantage of enabling the probing of the buried electronic states under the *operando* conditions for the active magnetic layers in the device structures.

Recently, we established a versatile experimental technique to study the spin-polarized electronic structures of the magnetic materials by RIXS at the transition metal (TM) $2p_{3/2}$ absorption edges in an external magnetic field [1-3]. The effective magnetic field of the TM 3*d* states yields the spin-polarized 2*p* core states due to the effective Zeeman splitting, as shown in Fig. 1, and thus the magnetic circular dichroism (MCD) in the RIXS spectra can probe the spin-polarized electronic structures owing to the strong selection rule of the dipole-allowed transition between the core-level 2*p* states and the 3*d* states for the circularly polarized light.

We utilized this technique to probe the spin-polarized electronic structures of half-metallic Heusler alloys [2,3] in which one of the spin subbands is metallic and the other is semiconducting with a band gap, as illustrated in Fig. 1 [4]. To investigate the intrinsic electronic structures, we measured high-quality single-crystalline samples of ferromagnetic Co_2MnSi and ferrimagnetic Mn_2VAl. The experiments were performed at SPring-8 **BL07LSU** HORNET end-station, where linear and circular polarizations are available. Moreover, the beam position and energy resolution do not change upon polarization switching in BL07LSU, and thus the switching technique is suitable for MCD experiments. In addition, a compact magnetic circuit with an external magnetic field of 0.25 T is installed for RIXS-MCD measurements, as shown in Fig. 2 [1].

Figures 3(a) and 3(b) respectively show the photon energy dependence of the RIXS spectra at the Co and Mn $2p_{3/2}$ edges for Co_2MnSi. The fluorescence components, which shift linearly with increasing incoming photon energy, are dominant for both edges. The fluorescence signals in the RIXS spectra on Co_2MnSi occur at around 1.0 eV for Co and 1.5 eV for Mn away from the elastic line, as shown in Figs. 3(a) and 3(b), respectively. This suggests that the partial density of states (PDOS) differs between the Co and Mn 3*d* states in Co_2MnSi. On the other hand, the fluorescence signals in the RIXS on Mn_2VAl branch off from the elastic line, as shown in Fig. 3(c), indicating the difference in Mn 3*d* PDOS between the two materials [2-4].

Figure 3(d) shows the Co $2p_{3/2}$ RIXS-MCD spectra for Co_2MnSi with a clear MCD contrast, which are attributed to the spin-polarized electronic structures of the Co 3*d* states. The *dd* excitation around 1.5 eV is observed at around 778 eV, indicating that the Co 3*d* states on the unoccupied side are rather localized.

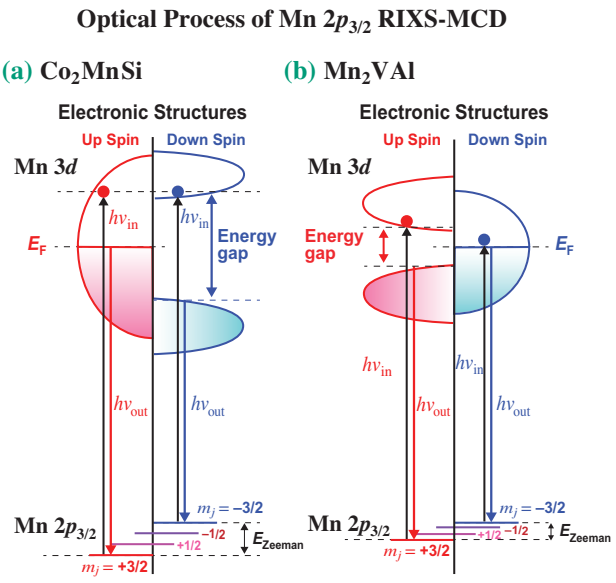

Fig. 1. Optical process of Mn $2p_{3/2}$ RIXS-MCD and the electronic structures of **(a)** Co_2MnSi and **(b)** Mn_2VAl.

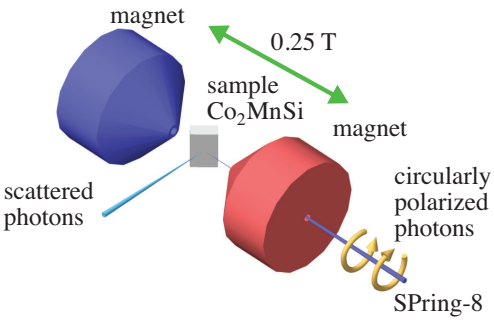

Fig. 2. Geometry of the RIXS-MCD measurements in an external magnetic field.

Moreover, the negative MCD signals (blue), which are mainly due to the Co $2p_{m_j=-3/2}$ states, reflect the down-spin components of the Co 3*d* states. Therefore, the gap structures between the elastic line and the *dd* excitation consist of the band gap of the Co 3*d* down-spin states. On the other hand, the positive MCD signals (red), originating mainly from the Co $2p_{m_j=+3/2}$ states, show the up-spin components. Interestingly, the positive MCD components are observed in the down-spin gap (green downward arrow), indicating the half-metallic electronic structures of the Co 3*d* states.

Figures 3(e) and 3(f) display the Mn $2p_{3/2}$ RIXS-MCD spectra for Co_2MnSi and Mn_2VAl. The distinct MCD contrasts indicate the high spin polarization of the Mn 3*d* states in both cases. The RIXS-MCD spectra for Co_2MnSi show the negative fluorescence MCD with the energy gap from the elastic line, reflecting the down-spin gap of the half-metallic electronic structures of the Mn 3*d* states shown in Fig. 1(a). On the other hand, the negative RIXS-MCD signals in Mn_2VAl are observed in the vicinity of the elastic line, suggesting that the down-spin states contribute to E_F. This is consistent with the predicted electronic structures of Mn_2VAl, in which the half-metallic gap opens the up-spin side, as shown in Fig. 1(b) [2,4,5]. Therefore, RIXS-MCD allows us to reveal which spin subband opens the half-metallic gap, providing important information for designing functional spintronic devices.

By utilizing RIXS-MCD, it will be possible to analyze the density of spin-polarized carriers induced at the buried interface of heterojunctions or magnetic layers under *operando* conditions. RIXS-MCD is a promising tool for probing useful information for designing spintronics devices.

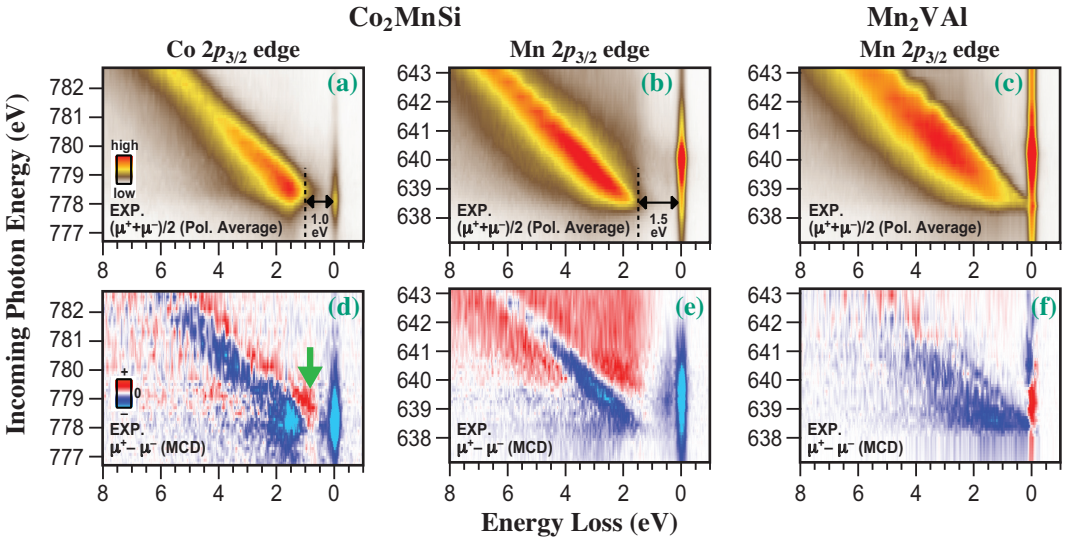

Fig. 3. Photon energy dependence of RIXSMCD spectra at the **(a)** Co and **(b)** Mn $2p_{3/2}$ edges for Co_2MnSi and the **(c)** Mn $2p_{3/2}$ edge for Mn_2VAl. Those RIXS-MCD spectra are plotted in **(d)**–**(f)**, respectively.

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