High-Magnetic-Field XMCD Using Pulsed Magnet

X-ray magnetic circular dichroism (XMCD) is a powerful and unique means to study magnetism because of element and shell selectivity. Although there have been many excellent works on the XMCD of magnetic materials, most of them focus on ferromagnetic materials. This is partly because a sufficiently high magnetic field for magnetizing antiferromagnetic or paramagnetic materials was not available at synchrotron X-ray facilities. Fields of 1-2 T are generally used for conventional XMCD experiments, and even the highest field using a large superconducting magnet is about 10 T.

Over several years, we have been developing experimental techniques for high magnetic field X-ray experiments in the 40 T range. We utilize a very small pulsed magnet with an inner diameter of 3-5 mm and a portable capacitor bank to perform several experiments, such as those involving X-ray diffraction [1] and X-ray absorption spectroscopy (XAS) [2]. In this work, we applied this technique to XMCD spectroscopy and investigate the valence fluctuating state of Eu atoms in $EuNi_2(Si_{0.82}Ge_{0.18})_2$. The ground state of this material at low temperatures is nonmagnetic owing to the strong c-f (conduction electron – f electron) hybridization. Therefore, so far, no one has considered it as a candidate material for the XMCD study. The high magnetic fields of up to 40 T can induce the magnetic moment and enable us to study the XMCD. We can expect the characteristic features in the XMCD spectra due to the Eu valence fluctuation, i.e., the quantum mechanical mixing of Eu^{2+} and Eu^{3+} states.

The XMCD experiment with the Eu $L_{2,3}$-edge ($2p_{1/2,3/2} \rightarrow 5d$) was carried out at beamline **BL39XU**. Figure 1 shows the photograph of the miniature magnet used. The crystal is powdered and diluted in order to achieve an effective sample thickness of about 10 μm for the transmission measurement. A diamond X-ray phase plate was introduced to generate circularly polarized X-rays. The XMCD signal ($\Delta\mu t = \mu^+t - \mu^-t$) is determined as the difference in absorption intensity between the right (μ^+t) and left (μ^-t) circular polarizations. μ^+t and μ^-t were measured by successively applying two shots of a pulsed field at a fixed X-ray energy while reversing the photon helicity every shot.

The measured XMCD spectra of $EuNi_2(Si_{0.82}Ge_{0.18})_2$ are shown in Fig. 2 together with the XAS spectra obtained at several magnetic fields [3]. Here, we only show the L_2 edge spectra. The valence fluctuation manifests itself as a double-peak structure in the XAS spectra. The higher energy XAS peak is attributed to the Eu^{3+} state, while the lower energy peak is attributed to the Eu^{2+} state. The XAS spectrum strongly depends on the magnetic field and exhibits a valence change induced by the magnetic field. We found that the double-peak structure corresponding to the two valence states is observed in the XMCD spectra as well as in the XAS spectra. The double peaks indicate that the electrons in the Eu $5d$ orbital, to where electron is finally excited from $2p$ levels, are magnetically polarized not only in the Eu^{2+} state (f^7; J=7/2 in the ground state) but also in the Eu^{3+} state (f^6; J=0 in the ground state). We also observed a similar double peak structure in the L_3-edge XMCD spectra [3]. It is worth noting that it is nontrivial to obtain a finite XMCD signal for the nonmagnetic Eu^{3+} state.

To clarify the origin of the valence-selective XMCD, we plot the field dependence of the integrated intensity of the XMCD peak (I_{mcd}), as well as the magnetic polarization of the $5d$ electrons (P_{5d}), in Fig. 3. The degree of magnetic polarization of the Eu $5d$ electrons in each valence state is defined here as $P_{5d} = \int\Delta\mu t \, dE / \int\mu t \, dE$, where $\Delta\mu t$ and $\mu t = (\mu^+t + \mu^-t)/2$ are the XMCD and absorption intensities, respectively. $\int\Delta\mu t \, dE \, (=I_{mcd})$ is deduced by the integration of the XMCD peak shown in Fig. 2 for each valence state. $\int\Delta\mu t \, dE$ is obtained from the integrated intensity of the absorption peak. Curve fitting analysis is performed in order to evaluate the absorption intensities $\int\Delta\mu t \, dE = I_2$ and I_3 for the Eu^{2+}

Fig. 1. Photograph of solenoid miniature magnet for XMCD measurement. The inner diameter is 3 mm. Maximum fields of up to 50 T can be generated. For a repetition of more than 300 shots, the maximum field is limited to 40 T for the XMCD experiment.

and Eu^{3+} states, respectively, at different magnetic fields. The magnetization (M) of the sample at 4.2 K [4] is shown in Fig. 3 in order to compare it with the dependence of I_{mcd} and P_{5d} on the field. An increase in magnetization caused by the valence change is observed at around 35 T. In addition, the I_{mcd} of Eu^{2+} and that of Eu^{3+} scale together with the magnetization, as expected for XMCD experiments. However, P_{5d} qualitatively differs between the Eu^{2+} and Eu^{3+} states, suggesting the different origins of the XMCD in such valence states.

We have now two possible origins of the valence-selective Eu^{3+}–XMCD: (i) the effect of the excited state of Eu^{3+} (J=1) and (ii) the magnetic polarization of the conduction electrons through c-f hybridization. Since the first excited state of Eu^{3+} is magnetic (J=1), the first possible origin (i) should have some effects. The energy separation between the first excited and ground states is about 480 K. If we take the Van Vleck term of Eu^{3+} into account, the contribution of the magnetization is found to be too small to explain the experimental result. However, in compounds exhibiting valence fluctuations, the mixing between the Eu^{3+}: J=0 and Eu^{3+}: J=1 states can be enhanced by the strong c-f hybridization. Therefore, the effect of the Eu^{3+}: J=1 state is one of the plausible origins of the Eu^{3+}-XMCD observed in the present work. The second possible origin is that the conduction electrons are polarized by the strong hybridization with the $4f$ electrons in the magnetic Eu^{2+} states, and these polarized conduction electrons induce the magnetic polarization of the $5d$ electrons in the nonmagnetic Eu^{3+} state. Owing to the strong c-f hybridization, this mechanism is also regarded as a plausible one.
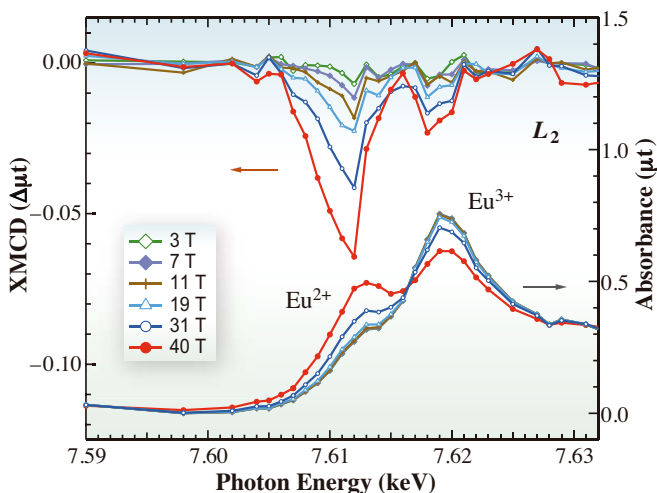
Although further studies are required to quantitatively understand the valence-selective XMCD, it is likely that the c-f hybridization strength plays an important role. It is very unique to use the high-field XMCD to clarify the strong correlation between the conduction and localized electrons in other valence fluctuating or heavy fermion compounds.
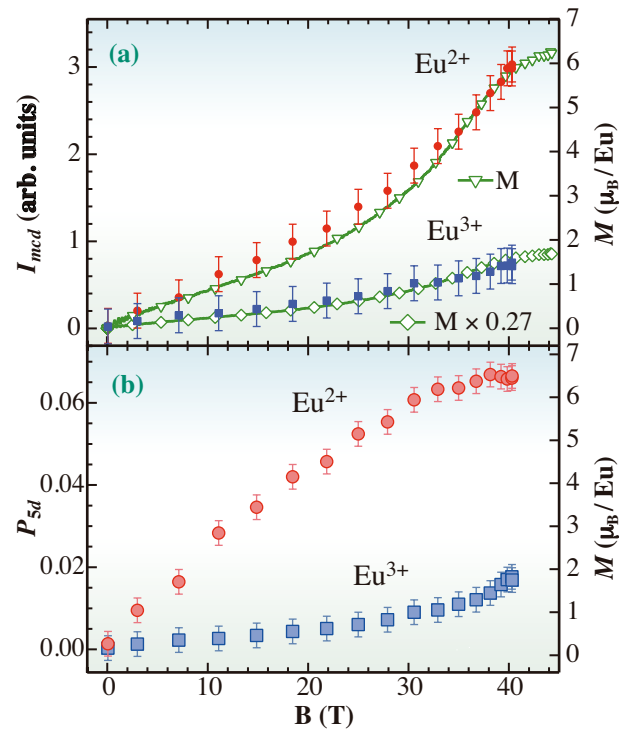

Fig. 3. Magnetic field dependences of **(a)** XMCD intensity and **(b)** normalized XMCD intensity P_{5d}.

Fig. 2. XMCD and XAS spectra of L_2 edge in $EuNi_2(Si_{0.82}Ge_{0.18})_2$ at several magnetic fields at 5 K.

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