Relativistic Multiple Scattering Approach to Heavy Metal $L_{2,3}$-Edge XMCD

Keiko Okamoto*, Shin-ichi Nagamatsu*, Takashi Fujikawa* and Hiroshi Maruyama†

*Graduate School for Science, Chiba University, 1-33 Yayoi-cho, Inage, Chiba 265-8522 Japan
†Department of Physical Science, Graduate School of Science, Hiroshima University, 1-3-1 Kagamiyama, Higashihiroshima, Hiroshima 739-8526 Japan

Abstract. The relativistic effects on L_2- and L_3-edge XMCD spectra are theoretically studied. For the excitation from heavy elements like W and Pt, a simple rule that the L_2-edge XMCD is closely related to the L_3-edge XMCD does not work because of the relativistic effects, and presumably of other effects. In the Pt L_2- and L_3-edge XMCD, the calculated spectra are quite sensitive to the spin moments on Pt. We can succesfully explain the main interesting features in the experimental XMCD spectra for $CrPt_3$ and $MnPt_3$.

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INTRODUCTION

Typically normalized X-ray magnetic circular dichroism (XMCD) to σ_0 at L_3-edge is approximately twice as large as that at L_2-edge, but with an opposite sign. This rule can be well applied to light elements like 3d transition metals. On the other hand, L_2- and L_3-edge XMCD from W, Pt show the breakdown of the rule, which could be explained based on the strong relativistic effects. For example, Maruyama *et al*. observed prominent difference in L_2- and L_3-edge XMCD for $CrPt_3$ and $MnPt_3$ [1, 2]: the L_2- and L_3-edge XMCD give an opposite sign for $MnPt_3$, whereas the same sign for $CrPt_3$ and $Cr_{1-x}Mn_xPt_3$.

In many aspects, relativistic quantum theory provides the natural framework for the study of XMCD spectra [3, 4, 5], because spin-orbit interaction is taken into account to all orders and we have no difficulty in convergence of the core function calculations. In the present paper we study the relativistic effects on the difference in L_2- and L_3-edge XMCD spectra from heavy elements like W and Pt. So far no systematic work on these problems has been found in literature.

THEORY

For the study of XMCD spectra, useful approaches have been developed based on the expansion in terms of nonrelativistic corrections [4, 5]. In the one-electron relativistic theory, the core function $|c\rangle$ from which an electron is excited can be written as

$$|c\rangle = \begin{pmatrix} |\phi_c\rangle \\ |\chi_c\rangle \end{pmatrix} \tag{1}$$

where the 2-spinors $|\phi_c\rangle$ and $|\chi_c\rangle$ are the large and small components. To give finite XMCD at $L_{2,3}$-edge, the relativistic effects splitting 2p state into $2p_{1/2}$ and $2p_{3/2}$ play a crucial role. In this case ΔT_{11} defined by

$$\begin{aligned} \Delta T_{11} = \quad & - \quad 2\mathrm{Im}\langle\phi_c|\Delta_+^* g(\varepsilon)\Delta_+|\phi_c\rangle \\ & + \quad 2\mathrm{Im}\langle\phi_c|\Delta_-^* g(\varepsilon)\Delta_-|\phi_c\rangle \end{aligned} \tag{2}$$

is responsible for the XMCD, where $g(\varepsilon)$ is a full one-electron nonrelativistic Green's function for the photoelectron energy ε, and $\Delta_\pm$ is electron-photon interaction operator with $\pm$ circular polarization, $\Delta_\pm \propto r\mathrm{Y}_{1,\pm}(\hat{\mathbf{r}})$. We should note that the X-ray propagation is parallel to the magnetization axis. In the case of nonrelativistic theory, the core function's ϕ_c are the same for the L_2- and L_3-edge excitation, which gives a simple relation [4, 5]

$$\Delta T_{11}(L_2) = -\Delta T_{11}(L_3). \tag{3}$$

For the core $L_{2,3}$-edge excitation from 3d transition metals, the relation works so well, but the approximation is poor for the excitation from heavy elements.

RESULTS AND DISCUSSION

At first we calculate the W $L_{2,3}$-edge XMCD spectra for Co monolayer on W(110) surface. This system shows an

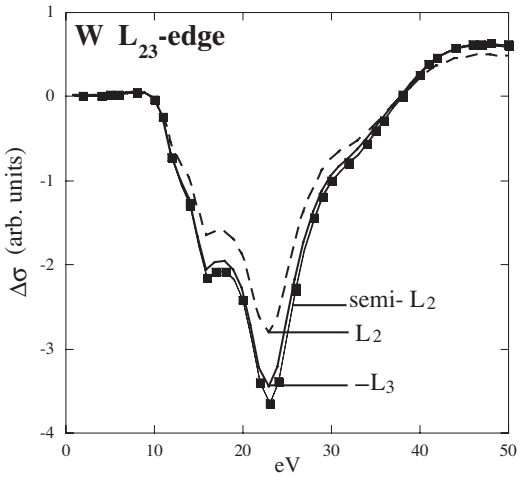

FIGURE 1. Calculated W XMCD spectra $\Delta T_{11}(L_2)$ at L_2- and $-\Delta T_{11}(L_3)$ at L_3-edge for Co monolayers on W (110) substrate. For the comparison $\Delta T_{11}(L_2)$ is shown for the semi-relativistic calculation.

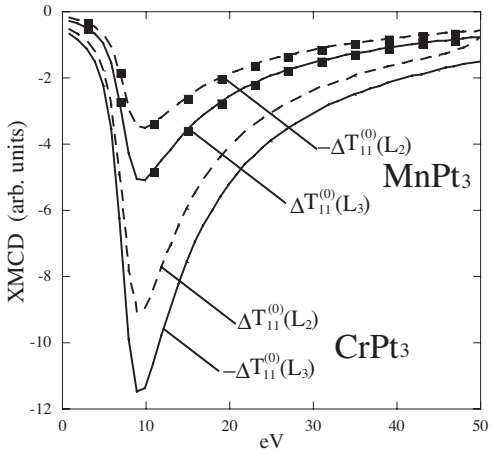

FIGURE 2. Atomic XMCD spectra for $CrPt_3$ at L_2-($\Delta T_{11}^{(0)}(L_2)$; dashed line) and L_3-edge ($-\Delta T_{11}^{(0)}(L_3)$; solid line), and for $MnPt_3$ at L_2- ($-\Delta T_{11}^{(0)}(L_2)$; dashed line with squares) and L_3-edge ($\Delta T_{11}^{(0)}(L_3)$; solid line with squares).

in-plane magnetization and larger orbital magnetic moments for the in-plane magnetization axis [6]. Figure 1 shows the calculated results for the relativistic theory compared with the semi-relativistic theory where ϕ_c is the same both for the L_2- and L_3-edges and eq. (3) works accurately. The semi-relativistic result shows the quite similar spectrum to the relativistic L_3-edge XMCD because the radial nonrelativistic 2p core function is quite similar to the relativistic $2p_{3/2}$ core function [7]. The relativistic effect makes the XMCD at L_2-edge different from the one at L_3-edge as shown in the figure, although the difference is small.

Next we calculate the Pt $L_{2,3}$-edge XMCD spectra for $CrPt_3$ and $MnPt_3$ alloys. Maruyama *et al*. have measured the XMCD spectra for these alloys [1, 2]. As the Pt atom is much heavier than W, we can expect more prominent relativistic effects in those XMCD spectra. Tohyama *et al*. have studied the magnetic order in $CrPt_3$ (ferrimagnetic) and $MnPt_3$ (ferromagnetic): Small amount of magnetic moments on Pt ($-0.26\mu_B$ in $CrPt_3$, $0.14\mu_B$ in $MnPt_3$) are estimated by neutron scattering analyses [8]. Figure 2 shows the atomic XMCD $\Delta T_{11}^{(0)}(L_2)$ and $-\Delta T_{11}^{(0)}(L_3)$ for $CrPt_3$, $-\Delta T_{11}^{(0)}(L_2)$ and $\Delta T_{11}^{(0)}(L_3)$ for $MnPt_3$ at Pt L_2- and L_3-edges. Atomic XMCD only reflects the magnetic moments on the X-ray absorbing atom Pt, which are in different direction. We thus expect that $\Delta T_{11}^{(0)}(L_2)$ for $MnPt_3$ has different sign from that for $CrPt_3$. We find the larger difference $|\Delta T_{11}^{(0)}(L_2) + \Delta T_{11}^{(0)}(L_3)|$ in Pt L_2- and L_3-edges than that found in W L_2- and L_3-edge XMCD spectra as expected. These atomic XMCD vanish in the case Pt atoms are nonmagnetic. The observed Pt L_2- and L_3-edge spectra for these systems show the same sign for $CrPt_3$, whereas they have a different sign for $MnPt_3$ [1, 2]. Atomic XMCD is rather sensitive to the magnetic moment on Pt, however it cannot show the same sign. How do the multiple scattering relativistic calculations influence the XMCD spectra? Figures 3 and 4 show the calculated results compared with the experimental XMCD spectra, where we observe the remarkable change from the atomic XMCD: The observed feature where Pt L_2- and L_3-edge XMCD show the same sign in $CrPt_3$ is well explained by these calculations. Except for Pt L_2-edge XMCD, the agreement between the theory and the experiment is rather good. For these calculations we have used the spin moments estimated from the neutron scattering experiment [8], $3.37\mu_B$ on Cr and $3.60\mu_B$ on Mn in $TMPt_3$ (TM=Cr, Mn). For the spin moments on Pt we assume that they are nonmagnetic. The neutron scattering experiments provide not so reliable values on Pt atoms for these systems.

Figure 5 shows the calculated relativistic XMCD spectra for $CrPt_3$ for different spin moments ($-0.02 \sim -0.04\mu_B$) on Pt: The spectra are sensitive to the moments. As observed in Fig.3, the XMCD $\Delta T_{11}(L_2)$ and $\Delta T_{11}(L_3)$ show the same sign and nearly the same order of magnitude for nonmagnetic Pt. For the spin moments μ_B(Pt)=-0.02 and $-0.03\mu_B$, we observe that $\Delta T_{11}(L_2)$ is much smaller than $\Delta T_{11}(L_3)$. For μ_B(Pt)= -0.04, $\Delta T_{11}(L_3)$ suddenly changes its sign. These results show that the relativistic effects and multiple scattering effects make the XMCD sensitive to the magnetic structure on Pt.

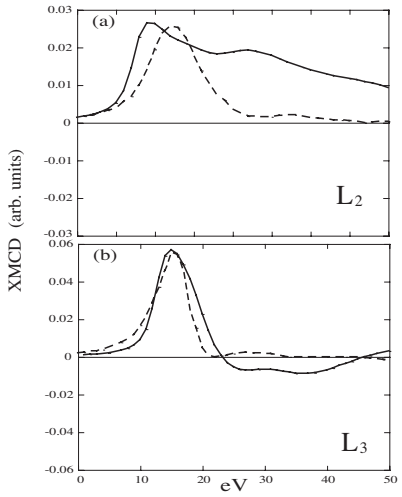

FIGURE 3. The calculated (solid line) and experimental (dashed line) Pt XMCD spectra for $CrPt_3$ at (a) L_2- and (b) L_3-edge.

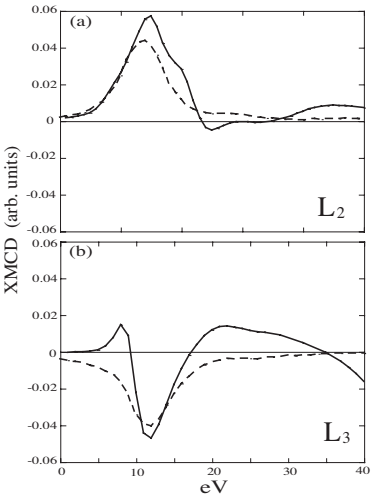

FIGURE 4. The calculated (solid line) and experimental (dashed line) Pt XMCD spectra for $MnPt_3$ at (a) L_2- and (b) L_3-edge.

CONCLUSION

We have investigated how the relativistic effects influence the XMCD spectra from some alloys including heavy metals. Prominent relativistic effects are observed in the calculated XMCD spectra. In particular, the calculated Pt L_2- and L_3-edge XMCD show the same sign in $CrPt_3$ as far as we assume small amount of spin moments on Pt. The calculated XMCD spectra for $MnPt_3$ are not so sensitive to the moments on Pt in contrast to those for $CrPt_3$.

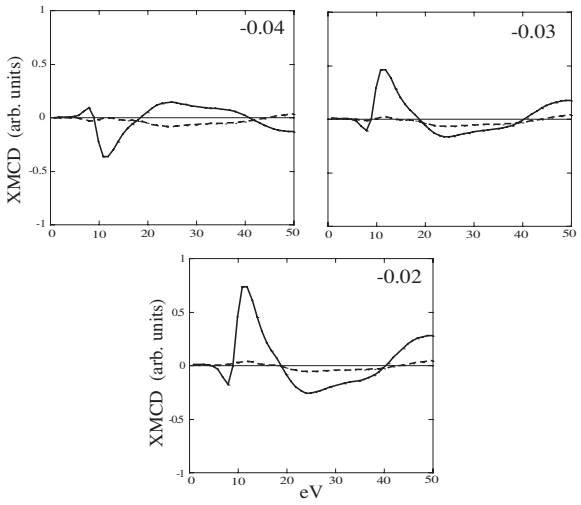

FIGURE 5. The calculated XMCD spectra at L_2- (dashed lines) and L_3-edge (solid lines) for $CrPt_3$ for different spin moments on Pt: $\mu_B(\mathrm{Pt}) = -0.02, -0.03, -0.04$.

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