Development of Ab-initio Multiplet Approach for Transition Metal 2*p* Resonant Inelastic X-ray Scattering

Hidekazu Ikeno

[1] Nanoscience and Nanotechnology Research Center, Research Organization for the 21st Century, Osaka Prefecture University, 1-2 Gakuen-cho, Naka-ku, Sakai Osaka 599-8570, Japan

In 3*d* transition-metal (TM) compounds, 3*d* electrons play a central role in determining their macroscopic properties.Both soft x-ray absorption spectroscopy (XAS) and electron energy-loss spectroscopy (EELS) at TM $L_{2,3}$-edges, which mainly monitor $2p \rightarrow 3d$ electric dipole transitions, have been widely used for the investigation of 3*d* electronic structures in TM compounds.

Recently, TM 2*p* and 1*s*2*p* resonant inelastic x-ray scattering (RIXS) attract increasing attentions as new probes of 3*d* orbitals. RIXS measures scattering of the high-enery photons near the absorption edges of selected elements. Figure 1 shows the schematic picture for the electronic transtions in the 2*p* RIXS process of 3*d* TM element. The first transition from initial to intermediate states corresponds to the TM-$L_{2,3}$ x-ray absorption process. The final states after photon emissions correspond to the final states for optical absorption. Other elementary excitations, including phonons and magnons, can also be observed by tuning the incident and emitted photon energies. From RIXS we can obtain 2 dimensional spectra related to the electronic excitations in materials and hence can obtain much more information than XAS/EELS. In order to extract the information about the electronic structures from experimental spectra, however, reliable theoretical tools which have predictive performance and that are free from adjustable parameters are indispensable.

The aim of this work is to develop an *ab-initio* approach for the transition metal 2*p* RIX. Similar to the case of TM-$L_{2,3}$ XAS/EELS, the shape of TM-2*p* RIXS is dominated by the multiplet effects arising from the strong electronic-correlations between core-2*p* and 3*d* electrons. The present author has already developed a full relativistic configuration interaction (CI) calculation for XAS and EELS spectra. In this method, the exchange correlation interactions among a 2*p* hole and 3*d* electrons are rigorously calculated in order to take the multiplet effects into account. Experimental XAS at TM $L_{2,3}$-edge and with different *d*-electron numbers, coordination numbers, and symmetries have been successfully reproduced without any adjustable parameters[1,2].

In this work, this method has been extended to calculate RIXS spectra. First, the relativistic CI calculations are performed for initial, intermediate and final states respectively. Then, the differential scattering cross-section can be calculated by the Kramers-Heisenberg formula as

$$F(!_{!"}, !_{!"\#}) = \sum_f \left| \sum_! \frac{\langle f|\hat{T}_2|!\rangle\langle !|\hat{T}_1|!\rangle}{!_i!!\omega_{in}-!_!!!\Gamma_!} \right|^! \delta(!_! + !!_{!"} \; ! \; !_! \; ! \; !!_{!"\#}),$$

where $!_!, !_f, !_!$ are eigenvalues of initial, final and intermediate states, $\hat{T}_1, \hat{T}_!$ are transition operators corresponding first and second transitions, and $\Gamma_!$ is natural width corresponding to the 2*p* core-hole lifetime. Figure 2 shows the Mn-2*p* RIXS speactra of MnO calculated with various incident photon energies. The spectral shapes obtained by the present work are consistent with the experimental spectra reported in ref [3]. The

result demonstrates the power of the *ab-initio* multiplet method for quantitatively reproducing 2*p* RIXS spectra as well as $L_{2,3}$ XAS and EELS.

This work was partly supported by the Grant-in-Aid for Young Scientists (B) 24760537 from the Japan Society for the Promotion of Science (JSPS).

References

[1] H. Ikeno et al., J. Phys.: Condens. Matter **21,** 104208 (2009)
[2] H. Ikeno, T. Mizoguchi, and I. Tanaka, Phys. Rev. B **83,** 155107 (2011)
[3] G. Ghiringhelli et al., Phys. Rev. B **73,** 035111 (2006).

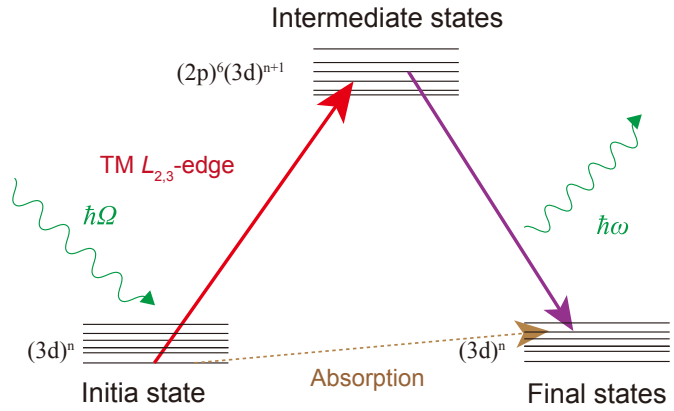

FIG. 1. Schematic picture for the electronic transitions in the 2p RIXS of 3d transition metal.

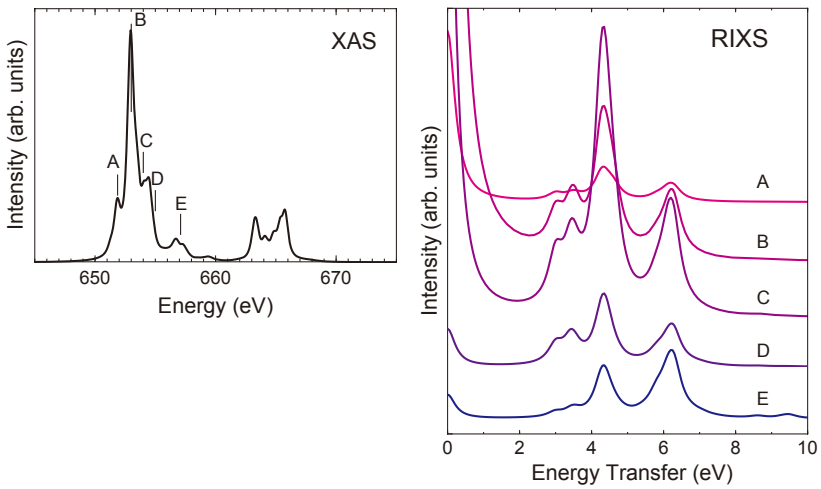

FIG. 2. Theoretical Mn-$L_{2,3}$ XAS and Mn 2p RIXS of MnO. The incident energies of RIXS spectra are indicated by vertical bars on the XAS spectrum.