L- and *M*-edge Resonant Inelastic X-ray Scattering in Transition-Metal Compounds

Michel van Veenendaal

Dept. of Physics, Northern Illinois University, De Kalb, IL 60115, U.S.A
Advanced Photon Source, Argonne National Laboratory, 9700 S Cass Avenue, Argonne, IL 60439, U.S.A

Introduction

Recently, resonant inelastic x-ray scattering (RIXS) in transition-metal compounds at the transition-metal L ($2p \rightarrow 3d$) and M ($3p \rightarrow 3d$) edges has attracted considerable attention [1-3]. In this technique, an incoming x-ray with energy $\hbar\omega$ excites an electron from a core level into the valence shell and one measures the energy $\hbar\omega'$ of the outgoing x-ray resulting from the recombination of the core hole with a valence electron. Since there is no core hole in the final state, the energy lost by the scattered photon can be related to excitations in the valence shell.

Results

Although the spectral line shape of this RIXS can often be calculated with numerical methods, the analytical understanding of this spectroscopy is still limited. We demonstrate how an effective scattering approach [4] can provide insight into the polarization dependence of RIXS. Within this framework, RIXS can be expressed as one-particle hole operators $W^{\pm}_{Qq}$ that are weighted by an angular dependence $T_{Qq}(\mathbf{e}, \mathbf{e}')$, where $\mathbf{e}$ and $\mathbf{e}'$ are the polarization vectors of the incoming and outgoing x-rays, respectively. The angular dependencies can be dramatic as shown in Fig. 1 for a divalent copper atom in D_{4h} symmetry. The ground state has a hole in the x^2-y^2 orbital. The calculation is done for a 90° scattering geometry, i.e., the x-rays are detected perpendicular to the incoming x-ray. The polarization of the incoming x-ray is linear, and the angle of the incoming polarization vector with the scattering plane is given by φ. The angle of the incoming x-ray with the surface normal is θ. The intensity of the elastic line is predominantly determined by the scattering operator $W^{\pm}_{00}$ that contains the hole operator. The angular dependence is $T_{00}(\mathbf{e},\mathbf{e}')= \mathbf{e}^*\cdot\mathbf{e}'$. Note that if the incoming polarization vector $\mathbf{e}$ is in the scattering plane ($\varphi=0°$), it is perpendicular to both outgoing polarization vectors and the elastic scattering is close to zero. Small elastic scattering still occurs due to higher-order scattering operators.

Using the analytical expressions for the scattering operators [5], it is also possible to show that no spin flips are allowed for holes in x^2-y^2 and xy orbitals but are allowed for holes in the other $3d$ orbitals. Let us now consider, the situation of normal incidence ($\theta=0°$). Ghiringhelli *et al.* [2] demonstrated that rotating the incoming polarization vector out of the scattering plane ($\varphi=0\rightarrow 90°$) increases the elastic peak and decreases the $x^2-y^2 \rightarrow xy$ scattering. Let us first consider the outgoing polarization vector $\mathbf{e}'_1=\mathbf{y}$. For the polarization vector in the scattering plane ($\mathbf{e}=\mathbf{x}$), $\mathbf{e}'_1\cdot\mathbf{e} =\mathbf{y}\cdot\mathbf{x}=0$ and $\mathbf{e}'_1\times\mathbf{e} =\mathbf{y}\times\mathbf{x}=-\mathbf{z}$, and for the polarization vector perpendicular to the scattering place ($\mathbf{e}=\mathbf{y}$), $\mathbf{e}'_1\cdot\mathbf{e} =\mathbf{y}\cdot\mathbf{y}=1$ and $\mathbf{e}'_1\times\mathbf{e} =\mathbf{y}\times\mathbf{y}=\mathbf{0}$. Therefore for $\varphi=0\rightarrow 90°$, $T_{00} =\mathbf{e}'_1\cdot\mathbf{e}=0\rightarrow 1$, and $T_{00} =\mathbf{e}'_1\times\mathbf{e}=3i/2\rightarrow 0$. Since the scattering is dominated by the terms without spin flip, the RIXS with $\mathbf{e}$ in the scattering plane has a strong $x^2-y^2\rightarrow xy$ scattering from $W^{\pm}_{10}$. Whereas with $\mathbf{e}$ perpendicular to the scattering plane, the RIXS has a strong elastic $x^2-y^2\rightarrow x^2-y^2$ component due to $W^{\pm}_{00}$. The other outgoing polarization vector $\mathbf{e}'_2=\mathbf{z}$ remains at a 90° angle with the incoming polarization vector for $\varphi=0\rightarrow 90°$. This gives $\mathbf{e}'_2\cdot\mathbf{e}=0$ and $\mathbf{e}'_2\cdot\mathbf{e}=\mathbf{y}\rightarrow -\mathbf{x}$ for $\varphi=0\rightarrow 90°$. The scattering for $\mathbf{e}'_2$ is therefore determined by $W^{\pm}_{1,\pm 1}$. The $x^2-y^2\rightarrow yz/zx$ scattering intensity does not change when rotating the incoming polarization vector out of the scattering plane.

Acknowledgments

This work was supported by the U.S. Department of Energy (DE-FG02-03ER46097) and NIU's Institute for Nanoscience, Engineering, and Technology under a grant from the U.S. Department of Education. Work at Argonne National Laboratory was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38.

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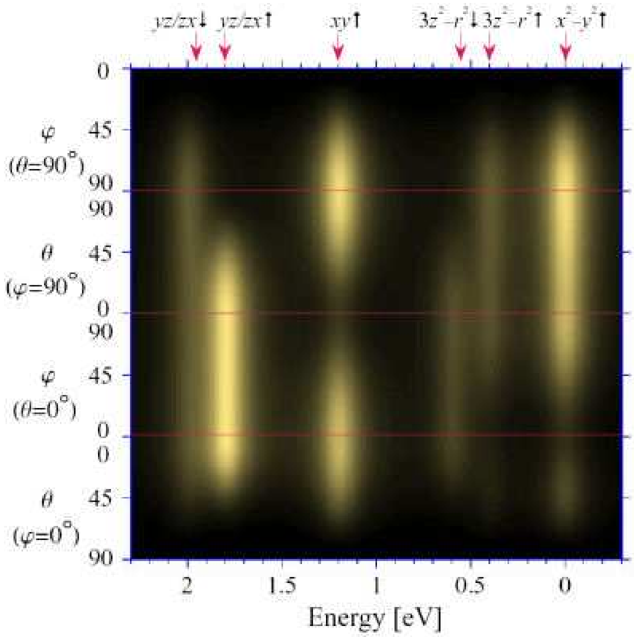

Figure 1: Density plot of the RIXS intensities at the L_3 edge for a 90° scattering geometry as a function of the angle θ of the incoming radiation with the surface normal and the angle φ between e and the scattering plane. The ground state has a hole in the x^2-y^2 orbital.