High-resolution Resonant X-ray scattering Study on 3d eg^1 systems in Ultrathin Films and Heterostructures of $RNiO_3$ (R=rare earth)

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The success of the interfacial engineering on the band structure leads to the astounding advances in the science and technology of semiconductor heterostructures in the past four decades. In contrast, interfaces of Mott materials with inherent multiple components (charge, spin, orbital and lattice) offer spectacular opportunities of realizing novel quantum phases of radical diversity [1], due to the many-particle physics. On the other hand, despite the recently observed emergent phenomena in transition metal oxide (TMO) heterostructures [2-5], exploring and understanding correlated electrons at nano and even atomic scale have been experiencing formidable challenges in experiment and theory. To extract the accountable rules from the complexity and facilitate functional tailoring, investigation on the correlation-driven metal-insulator transition (MIT), the kernel of the Mott physics, with advanced techniques at heterointerfaces is vital.

The perovskite nickelates family, $RNiO_3$ is known as a prototype of bandwidth-control MIT. In addition, due to the small ligand-to-TM charge-transfer energy, $RNiO_3$ is subject to self-doping which may be utilized to drive the properties without disorder from chemical doping. To elucidate the electronic structure and low-lying charge excitation evolving across the MIT, we performed high-resolution resonant inelastic X-ray scattering (RIXS) experiments at Ni *L*-edge and O *K*-edge. The element-sensitive and site-specific characteristics and resonant enhancement of this technique are ideal to probe ultrathin layers that might be buried in multi-component heterostructures. Resonant inelastic soft x-ray scattering (RIXS) is sensitive to a wide range of elementary excitations like crystal-field, inter-band and charge-transfer excitations of electronic nature [6], but also to magnetic excitations and phonons in solids [7,8]

Beamline BL07LSU of the SPring-8 provides a unique combination of desired soft X-ray energies and high brilliance, which is critical to measure these ultrathin materials. With the soft X-ray scattering endstation, 100 meV resolution can be achieved to provide a detailed picture of low-lying excitation for comprehensive understanding on the electronic structure.

Figure 1 shows the Ni *L*-edge X-ray absorption spectra of $NdNiO_3$ and $LaNiO_3$ at room temperature. The two-peak in $NdNiO_3$ is clearly observed. Unfortunately, La *M*-edge X-ray absorption overlaps with the Ni *L*-edge and dominates X-ray absorption in the energy region of interest, thus we couldn't make the comparison with the MIT for the case of replacing the rare-earth elements in $RNiO_3$-based system.

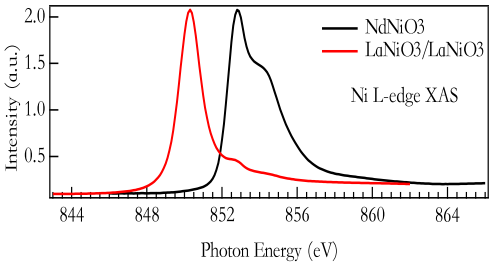

Figure 1. Ni L_3-edge X-ray absorption spectra of $NdNiO_3$ and $LaNiO_3$ at 300K. The two-peak in $NdNiO_3$ is observed.

Ni *L*-edge RIXS spectra may still be a possible means to study the MIT effect in $RNiO_3$-based systems. However, the intensity of Ni

L-edge RIXS has been so weak that we cannot obtain one decent RIXS spectrum.

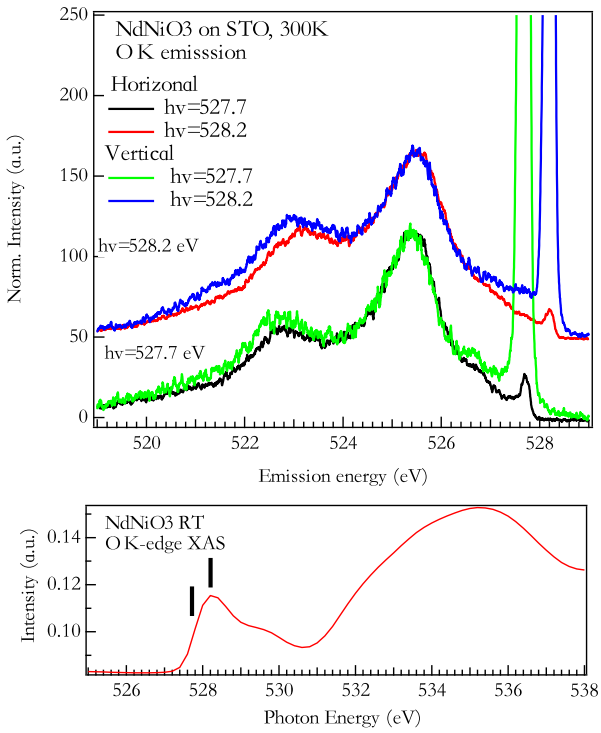

Figure 2. Upper panel: O *K-edge* RIXS at the first absorption resonance of $NdNiO_3$ and 0.5 eV below; bottom panel: O *K*-edge X-ray absorption spectra of $NdNiO_3$.

Thus, we decide to measure the O *K*-edge RIXS spectra. Figure 2 shows the O *K*-edge X-ray absorption spectra of $NdNiO_3$. The vertical bars indicate that the excitation energies for O *K*-edge RIXS spectra to be recorded. The lower panel of Figure 2 shows the O *K*-absorption spectrum where the vertical bars indicate the corresponding excitation energies for the RIXS spectra to be recorded. The upper panel of Figure 2 shows the O *K*-RIXS emission spectra excited on selected energies: the maximum of the 1st O *K* resonance and at 0.5 eV below the maximum on an energy loss scale. When tuning the x-ray energy to the first $NdNiO_3$ O *K*-absorption peak, the O 1*s* electron is excited into empty O 2*p* states strongly hybridized with the Ni 3*d* states. Two different detection geometries are compared: depolarized (polarized) geometry means that the scattered X rays are detected along (perpendicular to) the direction of the electric field vector of the incident x rays.

The RIXS spectra in Figure 2 are dominated by two intense broad peaks (maximum about 2.4 eV and 5 eV energy loss) with a low intensity tail structure (centered around 7 eV energy loss). Here at the SPring-8 with high resolution (100 meV), we record the elastic peak energy which allows the determination of the corresponding loss energies. Moreover, we observe excitations at lower energies (about 1 eV). The main contribution is attributed to *dd* excitations at about 1 eV which are mediated by the O 1*s*-core hole state. This means that d^8 final states of the excited Ni ion are reached. The *dd* excitations have been observed in Ni *L*-edge RIXS with the strongest component at 1 eV energy loss, which is corroborated by Ni *M*-edge RIXS at much higher resolution. Note also the extra intensity at about 1.25 eV energy loss that is observed in the polarized geometry but absent in the depolarized geometry.

References

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