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## Resonant inelastic X-ray scattering probe of inter- and intra-atomic interactions in uranium systems

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A central issue in the investigation of various properties of actinide systems is the nature of the 5f electrons in different systems. Many of the unusual properties found specifically in uranium (U) compounds are thought to be related to the delocalization of the partially filled U 5f electron states and their hybridization with the U 6d-7s states. However, due to the complexity of the physics involved, a complete understanding of the electronic structure of actinides has not yet been achieved.

An element-selective probe of the electronic structure is provided by inner-shell X-ray spectroscopy. Especially, hard X-ray spectroscopy is the ideal candidate for the actinide systems, since it does not require the high-vacuum environment around the sample. For the moment, the X-ray absorption near edge spectroscopy (XANES) at the L3 edge of U (~17.166 keV) has been the most commonly reported. Nevertheless, the large lifetime broadening at the 2p level (~7.4 eV for U) renders the technique little sensitive to the electronic structure. Härmäläinen and co-workers [1-2] showed that the L3 edge of lanthanides can be studied in considerably more details as compared to standard absorption spectroscopy by employing an X-ray emission spectrometer with an instrumental energy resolution similar to the core hole lifetime broadening.

We have studied the electronic structure of U systems in different oxidation states by means of resonant inelastic X-ray scattering (RIXS) via transitions between core levels and between core and valence levels at U L3 edge. The pre-edge structure near the main L3 edge is clearly resolved in U L3 XANES spectra with a help of X-ray emission spectrometer. The general model for interpreting pre-edges of lanthanides is to consider them as quadrupole transitions into the empty f states. In order to analyze the possible existence of quadrupole transitions of U compounds we carried out FDMNES [3] calculations for the different U systems. Crystal field splitting of the U 6d states were observed in valence band RIXS spectra for UO<sub>2</sub> system and reproduced theoretically using LDA+ U [4] approximation.

On the other hand, the f shell can be directly probed via dipole transitions from 3d core levels (U M<sub>4,5</sub> absorption edge). RIXS spectroscopy has also been used to record changes of the oxidation state in the U systems, by directly observing f-f interactions at U M<sub>5</sub> edge. The experimental results are supported by the atomic multiplet calculations[5]. The f-f transitions uniquely identify the ground state configuration of the uranium ion and can therefore provide an indication of the valence state of U in different complex materials.

Reference:

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