Resonane Inelastic X-ray Scattering on Dy at High Pressures

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Unusual phase phase transitions driven by electron correlation effects occur in many f-band metals (lanthanides and actinides alike) from localized phases to itinerant phases at high pressures[1]. Some of these transitions are accompanied by large volume collapses (for example, 15 % in Ce at 1 GPa [1], 10 % in Pr at 20 GPa [2] and 6 % in Dy at 73 GPa [3]) and dramatic changes in crystal structures (for example, high symmetry closed packed structures to low symmetry monoclinic structures). The dramatic changes in specific volumes and crystal structures reflect equally significant changes in the underlying electronic structure of these correlated f-electron metals. Therefore, the direct examination of electronic structure by various spectroscopic methods can provide critical insight into the nature of the transitions.

The goal of this study is, therefore, to determine the pressure-induced f-electronic structure changes in 4f rare-earth metals across the electronic correlation-driven phase transitions. The most relevant information to electron correlation effects is contained explicitly in the M band (corresponding to the 3d → 4f transition in rare-earths) at approximately 1 keV. However, high-pressure investigation of such a low energy transition is not feasible by using a conventional x-ray absorption technique, because of strong x-ray absorption by diamond and/or metal gasket. In fact, such low energy x-ray does not even penetrate through the bulk of f-electron metal samples and, thus, the studies have been remained largely on surface at ambient conditions.

Utilizing an intense high energy-resolution (~1eV) hard x-ray from the SP12XU beamline at the Spring-8, we have performed high pressure Resonant Inelastic X-ray Scattering (RIXS) at the LIII edge of Dy to 105 GPa in a diamond-anvil cell (DAC). By scanning the incident energy in steps of 2 eV through the LIII edge, 2p->5d, at 7790 eV and monitoring the decay of this excited state though Lα1 emission, 3d->2p, at 6495.2 eV we can gain information on M-edge x-ray absorption (XAS) not normally obtainable through a DAC. The RIXS technique also provides similar information as standard LIII XAS but without the 2p core-hole broadening effects seen in standard XAS.

Our preliminary result is summarized in Fig. 1 showing the pressure-induced change in Dy RIXS spectra at high pressures. A typical RIXS spectrum of Dy is comprised of two peaks resonanced with the 2p->4f transition at 7786 eV and the 2p->5d at 7794 eV. These resonances are identified by the maximums of the peaks at 1289 eV and 1299 eV energy transfer, as plotted in Fig. 1. These spectra clearly indicate the significant change in electronic structure with increasing pressures. We also performed high resolution partial flourescence yield (PFY) scans which provide similar information as LIII XAS without the 2p core-hole broadening effects that mask features related to preedge 2p->4f quadrupolar excitations. As pressure increases, this preedge feature stays relatively fixed, yet the position of the main 2p->5d edge shifts considerably from 7792.3 eV at ambient pressure to 7790.8 eV at 70 GPa. We are currently analyzing these pressure-induced changes in both RIXS and PFY to 105 GPa in terms of Anderson's volume collapse model.

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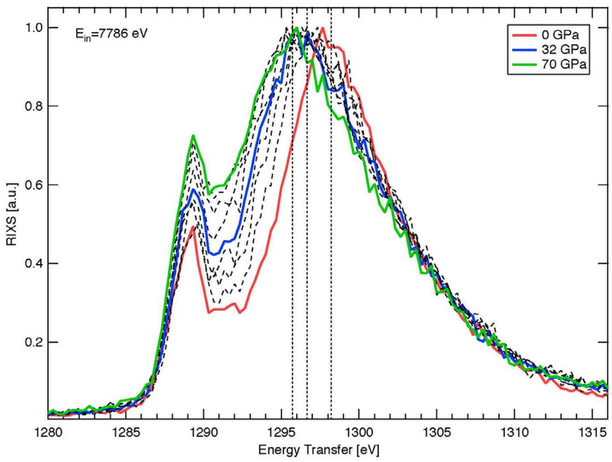

Figure 1. Pressure-induced changes in the RIXS spectra of Dy measured at the incident energy at 7786 eV.

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