ESRF	**Experiment title:** Study of the pressure-induced valence change in Eu and Yb compounds by resonant inelastic x-ray scattering	**Experiment number:** 1263
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Report:

We exploited high-resolution x-ray absorption spectroscopy (XAS) and resonant inelastic x-ray scattering (RIXS) to investigate the pressure dependence of the Yb valence in YbAl2. We observed a large increase from v=2.25 at ambient pressure, to v=2.9 at 385 kbar, indicative of a strong rearrangement of the electronic states due to an increasing 4f-band hybridization. The reliability of our results is enhanced by the combined use of the two spectroscopies, which leads to considerably reduced error bars with respect to previous determinations of the valence in Yb intermetallics under pressure.

Our recent experiments on $YbAgCu_4$ and $YbInCu_4$ showed that XAS in the partial fluorescence yield (PFY) mode and RIXS at the Yb L3 edge are sensitive bulk probes of the Yb 4f occupation in intermediate valence Yb compounds [1]. Namely, we were able to measure the characteristic 'Kondo' temperature dependence of the Yb valence which reflects the progressive depopulation of the hybrid ground state and the thermal excitation of magnetic states. Those results also indicated that the puzzling discrepancies between thermodynamic and (low energy) photoemission data are a consequence of the surface sensitivity of the latter.

Temperature is not the only parameter determining valence in intermediate valence Yb systems. Pressure – both external and chemical – enhances the 4f-band hybridization and favours the Yb^{3+} (f^{13}) ionic configuration, characterized by a smaller volume. For obvious reasons photon in- photon out techniques are the only viable spectroscopic probes of this effect. The predicted pressure dependence was qualitatively confirmed by pioneering XAS experiments [2]. Quantitative studies of the electronic configuration under pressure are now possible thanks to the high brilliance of the ESRF and the specific set-up of beamline ID16.

We used a Si(111) monochromator and a 1m spherical Si(620) analyzer to perform PFY-XAS and RIXS measurements at the Yb L3 edge (hν=8.95 keV), with a total energy resolution ΔE=1.3 eV. In PFY-XAS scans the intensity of the emitted Yb Lα ($2p^5 3d^{10}$ -> $2p^6 3d^9$; hν =7.41 keV) fluorescence was measured while scanning the energy of the primary beam. RIXS spectra were collected by analyzing the energy of the emitted beam at fixed values of the primary energy.

The PFY-XAS spectra (Fig. 1) show a continuous transfer of spectral weight away from threshold with increasing pressure, indicative of a progressive increase of valence. At 385 kbar the lineshape is similar to that of $YbInCu_4$ (v=2.96). A quantitative estimate of the Yb valence can be obtained at all pressures by decomposing (not shown) the XAS spectra into distinct 2+ and 3+ contributions with varying relative intensities. For the two contributions we assumed identical lineshapes, and an energy separation of 7 eV which reflects the 2p-4f interaction, and the different number of 4f electrons in the two configurations. The RIXS data provide an independent estimate of the Yb valence. When the incident photon energy is swept across the L_3 absorption profile, the intensities from the $3d^9 4f^{14}$ (2+) and $3d^9 4f^{13}$ (3+) final states are subsequently enhanced (Fig. 2). The relative strength of the 3+ and 2+ resonances varies with pressure, and at 385 kbar the resonance profile is dominated by the 3+ signal, in agreement with the XAS results. A combined quantitative analysis of the XAS and RIXS data yields the pressure dependence of Fig. 3 [3].

These results indicate that PFY-XAS and RIXS are powerful spectroscopic probes of intermediate valence. They also show that hybridization and 4f occupancy in Yb systems can be tuned by applying external pressure, opening the way to more challenging investigations of the P-T phase diagram.

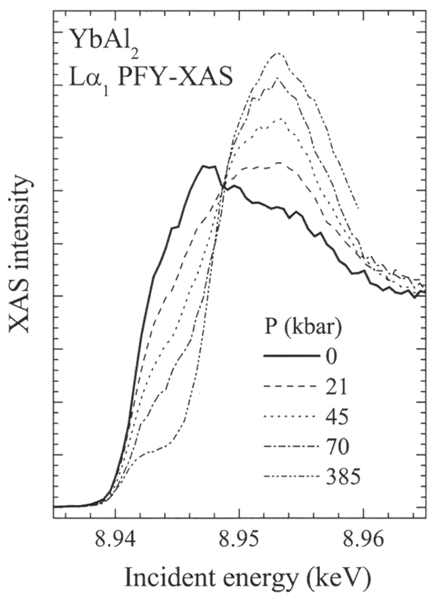

Fig. 1. PFY-XAS spectra of $YbAl_2$ at various external pressures.

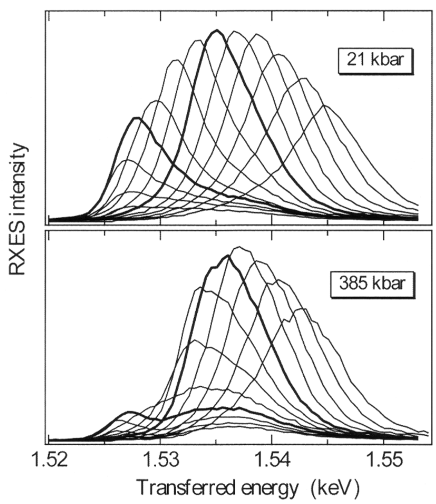

Fig. 2. RIXS spectra measured at 21 and 385 kbar across the 2+ and 3+ resonances.

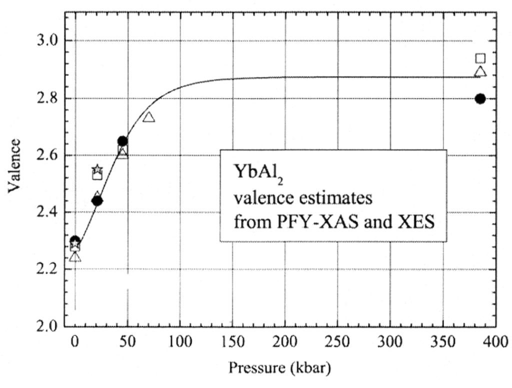

Fig. 3. Estimated pressure dependence of Yb valence from PFY-XAS and RIXS.

[1] C. Dallera et al., Phys. Rev. Lett. **88**, 196403 (2002)

[2] J. Röhler, G. Krill, J.-P. Kappler, M.-F. Ravet, D. Wohlleben, Valence instabilities, P. Wachter and H. Boppart editors, (North-Holland, 1982), p. 215.

[3] C. Dallera et al., submitted to Phys. Rev. B.