ESRF	**Experiment title:** Study of the pressure-induced valence change in Eu and Yb compounds by resonant inelastic x-ray scattering	**Experiment number**: HE1263
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Report:

The purpose of experiment HE1263 was to measure the pressure induced valence change of some Yb and Eu compounds by high-resolution resonant inelastic x-ray scattering. We were able to measure most of the proposed compounds, i.e. Yb metal, $YbAl_2$, YbS and EuS. We have already submitted the experimental report with the results on $YbAl_2$ (which have now been published [Ref. 1]). The present report is about results on YbS.

YbS is known from compressibility data and optical reflectivity measurements to undergo a progressive rearrangement of the electronic structure starting at around 100 kbar. The valence was correspondingly estimated by L_3 X-Ray Absorption to change from 2 (at ambient pressure) to 2.5 at 300 kbar [Ref. 2]. The large lifetime broadening of L_3 XAS leaves some uncertainties about these values. By performing Resonant Inelastic X-Ray Emission (RXES) measurements at the same edge, as a function of the incident and emitted energy, we could evaluate much more precisely the change in the electronic structure.

Figure 1 shows Partial Fluorescence Yield (PFY) spectra: the $L\alpha_1$ (*3d-2p*) decay channel is recorded while scanning $h\nu_{IN}$ across the L_3 edge. Two peaks arise in the spectrum due to the different Coulomb interaction of the Yb^{2+} and Yb^{3+} ions with the *3d* core-hole present in the final state of the process. The change in the relative weight of the two peaks is the indicator of the progressive valence change. In order to extract the valence value, the spectra were fitted at each pressure by two line shapes representing the two different valence contributions. The typical quality of the fit of given in the lower panel of Fig. 1. Thanks to the good contrast of the PFY spectra it is possible to reveal also the different line width of the two spectral components. This is a fingerprint of the different spread of the atomic multiplet structure, which remains visible in this rather ionic compound.

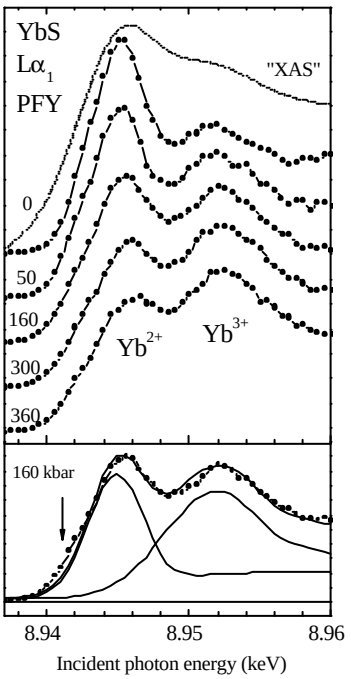

Figure 1. PFY spectra of YbS *vs.* P. A typical fit is reported (lower panel).

[1] Dallera et al., *Physical Review B* 68, 245114 (2003).
[2] K. Syassen, Physica, 139-140B, 277 (1986).

Resonant Inelastic X-Ray Emission spectra are reported in Fig. 2 for two pressure values. Spectra were measured at several incident energies across the L_3 threshold (dots in the inset). The spectra plotted with thick lines are taken at the resonance of Yb^{2+} and Yb^{3+}. The series of spectra taken at the different pressures have been decomposed in their divalent and trivalent component. In the fitting procedure the presence of fluorescent emission (at constant outgoing energy) must be taken into account as soon as the 2+ resonance is crossed. The valence value was again calculated as $v = 2 + I^{2+}/(I^{2+} + I^{3+})$, I^{2+} and I^{3+} being the intensity values of each component at its resonance.

As a third way to extract the valence we took advantage of RXES spectra measured with incident energy well below the L_3 absorption threshold. The analysis is more straightforward since no fluorescent component is present yet, and the trivalent component is better seen because the divalent resonance is still far. Of course these spectra are less intense and corrections due to the different distance from the resonances of the two components can be needed. Figure 3 shows these low intensity spectra at different pressures. With normalization to the divalent peak, the intensity of the trivalent component indicates the valence change.

Figure 4 summarizes the different valence estimates: the spread of these values gives the residual uncertainty, which is less than 0.05.

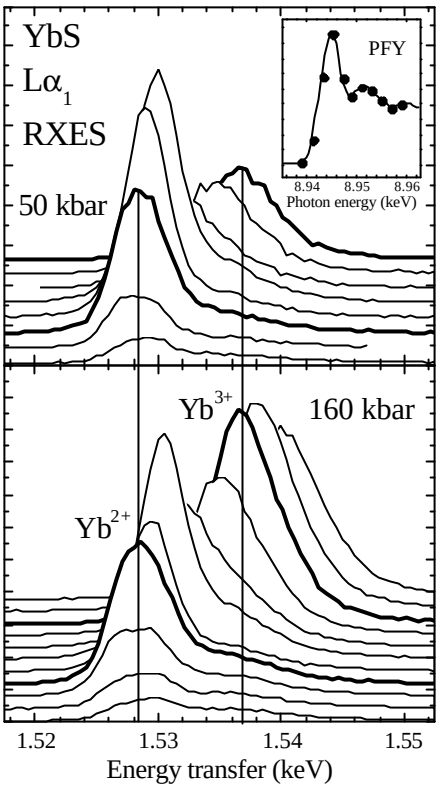

Fig. 2. RXES spectra of YbS at selected energies indicated in the inset.

The discussed results have been submitted to Physical Review B.

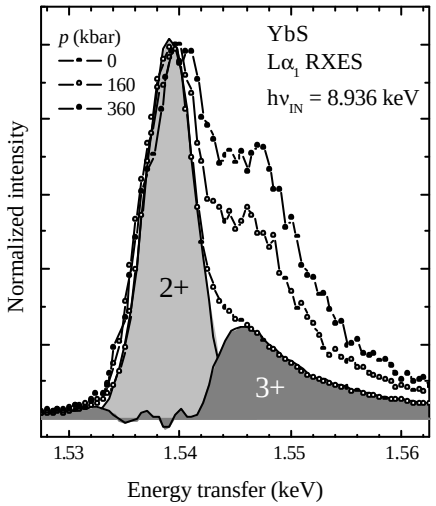

Fig. 3. RXES spectra excited well below threshold.

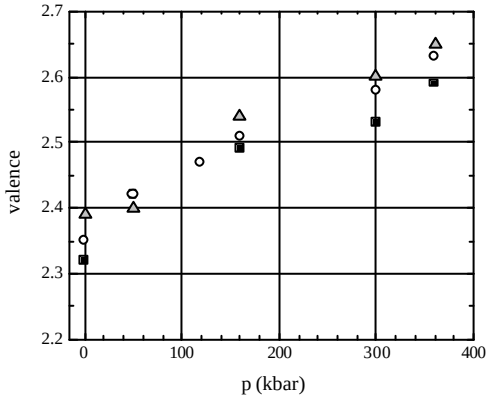

Fig. 4. Valence values estimates from the analysis of La_1 Partial Fluorescence Yield (circles) and L_3 Resonant Inelastic X-Ray Emission spectra (vs. hn_{IN} with triangles, well below threshold with squares).