



	Experiment title: Pressure-induced f -electron localization-delocalization in U-based heavy fermion	Experiment number: HE-1795
Beamline: ID-16	Date of experiment: from: 02/03/2005 to: 12/03/2005	Date of report: 20/04/2004
Shifts: 24	Local contact(s): G. Vanko	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): J.-P. Rueff* , Ph. Leininger* Laboratoire de Chimie Physique – Matière et Rayonnement, 11 rue Pierre et Marie Curie, 75231 Paris Cedex 05, France S. Raymond* , D. Braithwaite* CEA-Grenoble, DRFMC / SPSMS / MDN, 38054 Grenoble Cedex, France		

Report:

We report on the pressure-induced electronic changes under pressure in prototypical U-based heavy fermion compounds : UP_2Al_3 , and UPd_3 .

The experiment was carried out at ID-16 using the beamline Raman spectrometer laid out in the Rowland circle geometry. The spectrometer was equipped with a Ge(777) analyzer and an APD detector. The total energy resolution was estimated at 2.1 eV. The samples were loaded in a membrane pressure-cell with a 16:4:1 methanol-ethanol-water mixture as pressure transmitting medium. The medium is strictly hydrostatic up to the freezing pressure P_f (~ 14 GPa), but remains quasi-hydrostatic well above P_f ; the estimated differential stresses at 25 GPa is about 0.15 GPa [1]. Several attempts of loading the samples either with Ne or N_2 failed to produce any measurable signal. We attribute it to the mismatch between the spot size and the sample size, which is specially accentuated in the case of gas loading compared to a liquid medium. In the former case, the samples have to be smaller to sustain the higher compressibility of gases and subsequent shrinkage of the gasket hole upon loading.

Figure 1a illustrates the absorption spectra at the U- L_3 edge in UPd_2Al_3 as function of pressure. The absorption were acquired in the partial fluorescence yield mode, by monitoring the intensity of the U- $L_{\alpha 1}$ emission line, as the incident energy is swept across the edge. The spectra show almost no changes up to 20 GPa, except a slight energy-shift of the EXAFS oscillations related to contraction of the unit cell volume. This underlies further that non-hydrostaticity is not an issue here, as far as the electronic properties are concerned. On the contrary, a dramatic change occurs in the spectral lineshape above the structural transition pressure (see inset to Figure 1a) : The white line broadens and shifts towards higher energy by ~ 1.5 eV, in par with a reduction of the EXAFS oscillating pattern to a single peak. Note that the spectrum at 25 GPa is somehow intermediate between the two regimes.

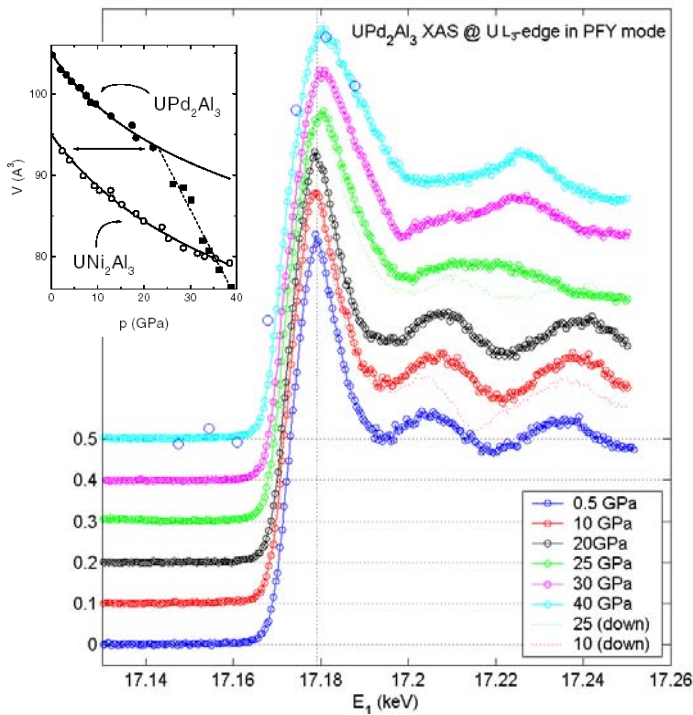


Figure 1a : U-L₃ edge absorption spectra in UPd₂Al₃ measured in the partial fluorescence yield mode. Diffraction data from Ref. [2] are shown in the inset.. Open circles denotes a measurement obtained in UPd₂Al₃ loaded with N₂ as pressure transmitting medium.

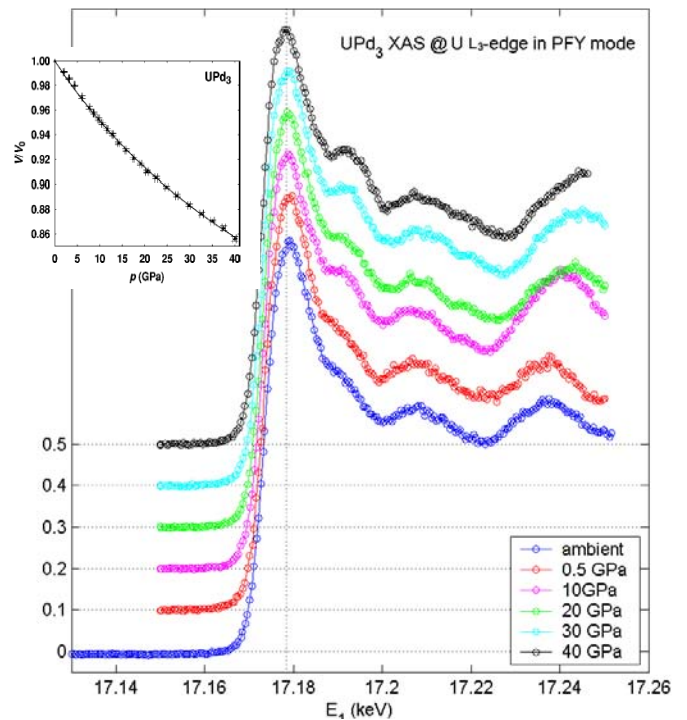


Figure 1b : U-L₃ edge absorption spectra in UPd₃ measured in the partial fluorescence yield mode. Diffraction data from Ref. [3] are shown in the inset.

In contrast to UPd₂Al₃, the absorption spectra measured in UPd₃ barely vary upon pressure increase (see Figure 1b). The main effect relates to the continuous growth in intensity of the resonance appearing 15 eV above the white line energy with increasing pressure (and a shift of the second EXAFS oscillation). This resonance is a common feature to most of the U-compounds, and is attributed to multiple scattering events [4]. Our results corroborate the diffraction data, which show a smooth contraction of the volume cell (see inset to Figure 1b) with increasing pressure.

These results are in stark contradiction with the orthodox picture of the valence change with pressure, as observed for instance in Yb compounds (see e.g. Ref. [5]). There, the valence change can be simply sketched as a transfer of n_f 4*f* electrons, according to the valence equilibrium: $\text{Yb}^{2+} \Leftrightarrow \text{Yb}^{3+} + 5d$. This picture manifests itself in the absorption spectra by two distinct white lines reminiscent of the two valent states. None of this in the 5*f*-electron Uranium compounds, where a localization – delocalization description seems more appropriate. LDA+U band structure calculations are planned to further investigate the influence of volume change on the 5*f* electronic properties.

References :

- [1] G. Fiquet et al., *American Mineralogist*, **84**, 856 (1999)
- [2] A. Krimmel et al., *J. Phys. Cond. Matter* **12**, 8801 (2000)
- [3] S. Heathman et al., *Phys. Rev. B* **67**, 180101 (2003)
- [4] E. A. Hudson, *Phys. Rev. B*, **52**, 13815 (1995)
- [5] E. Annese et al., *Phys. Rev. B*, **70**, 075117 (2004)