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Valence Instabilities of Superconducting $CeCu_2Si_2$ and $CeCu_2Ge_2$ Heavy Fermions at High Pressure and Low Temperature investigated by RIXS

Experiment number: HE-2636

Beamline: ID-26

Date of experiment
from: 04/**12/2007** to: 11/**12/2007**

Date of report: 13/02/2008

Shifts: 18

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Received at ESRF:

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Report:

$CeCu_2Si_2$ is the first discovered heavy fermion superconductor with a critical temperature T_c of 0.7 K [1]. Along with the isostructural compound $CeCu_2Ge_2$, superconductivity in $CeCu_2Si_2$ shows up in the vicinity of a magnetic quantum critical (QCP) at a pressure P_c. While the location of the QCP is well defined in $CeCu_2Ge_2$ where it is shifted towards higher pressures, P_c in CeCu2Si2 is supposedly negative (cf. Fig. 1). In both compounds, the superconducting transition temperature $T_c(P)$ exhibits a maximum at a pressure P_v above the magnetic QCP. At this pressure, transport properties point out a change of degeneracy of the ground state and a concomitant change of valence. It is proposed that the enhancement of superconductivity around P_v is produced by charge fluctuations that are important in the vicinity of this pressure [2]. Thus, the valence transition can only be observed through a line $P_v(T)$ in the T-P phase diagram, similarly to pure cerium. But contrary to Ce, the $P_v(T)$ phase line in $CeCu_2Si_{2,}$ is expected to terminate at a lower temperature below 20 K. Determining the valence change in the $P_v(T)$ region is therefore of prime importance to understand the electronic properties (including the superconductivity) in these materials.

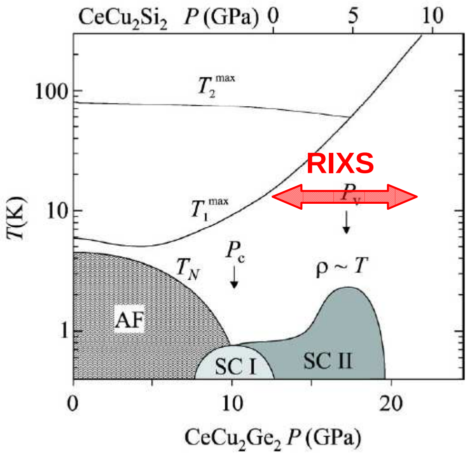

Figure 1: Phase diagram of $CeCu_2Si_2$. The red arrow indicates the measured region.

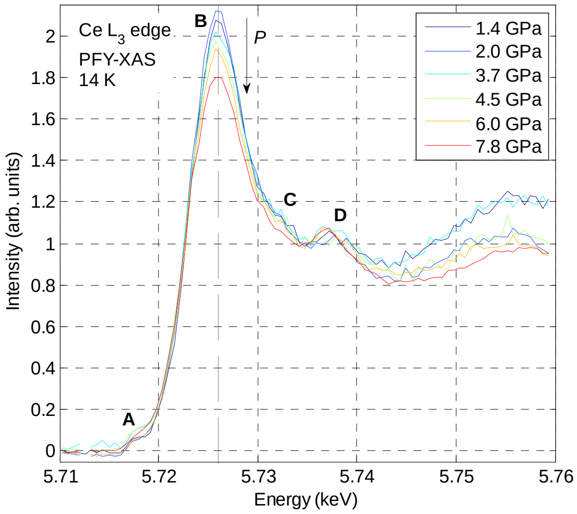

Figure 2: P-dependence of the Ce L_3 edge measured by RIXS in the PFY mode at the Ce $L_{\alpha1}$ line.

Earlier estimation of the Ce valence under pressure at 300 K using standard x-ray absorption spectroscopy (XAS) at the Ce L_3 edge reveals a jump of the valence under pressure [3]. However, the result accuracy can be seriously questioned considering the modest resolution of XAS. Furthermore, the equation of state of $CeCu_2Si_2$ at 300 K does not show any anomalies in this pressure range [4]. This suggests that the valence changes are small at room temperature. In contrast, one expects these to be larger close to $P_v(T)$ line which calls for new measurements combining high pressure and low temperature.

We report on the first investigation of the valence properties of Ce at high pressure at low temperature in $CeCu_2Si_2$ by resonant inelastic x-ray scattering. The measurements were carried out at the ID-26 beamline. A thin single crystal of $CeCu_2Si_2$ was loaded with silicone oil as pressure transmitting medium in a diamond anvil cell (DAC) prepared for transmission geometry. To optimize the photon throughput, the cell was equipped with a pair of 2 mm thick perforated diamonds capped by a 500 micron thick diamonds. Such a configuration yields a gain in transmission of several orders of magnitude in the considered energy range compared to standard pressure setups. For low temperature measurements, the cell was mounted on a liquid He cryostat. The pressure dependence of the Ce L_3 edge was measured using the beamline RIXS spectrometer in the so-called partial fluorescence yield (PFY) mode at the Ce $L_{\alpha 1}$ line. This technique yields absorption spectra essentially free from lifetime broadening effects, a crucial feature to unravel the Ce valence properties [5]. To that end, the RIXS spectrometer was equipped with a spherically bent Ge(331) analyzer which both selects the emitted photons energy in a narrow window around the Ce $L_{\alpha 1}$ maximum and focuses the reflected beam onto the detector. The absorption spectra are then measured by recording the detector intensity variation while scanning the incident energy around the Ce L_3 edge.

We focus on the region of the phase diagram close to P_v as indicated on Fig. 1. Figure 2 shows the Ce L_3 spectral changes at 14 K with increasing pressure. Unfortunately, time lacked to repeat the series at 300 K. The absorption spectrum consists of several features in the near edge region which reveals the Ce mixed valent character. By comparison with our early works, one can assign features A, B and D to different final state (in presence of the core hole) with varying f electron count, namely $4f^2$, $4f^1$ and $4f^0$. Peak B decreases markedly with increasing pressure while the intensity of peak D seems to be slightly reinforced. No clear variation of peak A can be observed. Finally, peak C could be related to the same sub-set of final states as the peak B for it shows a similar behavior with pressure although on a smaller scale.

Preliminary analysis of the data demonstrates a substantial increase of the Ce valence at high pressure and low temperature in $CeCu_2Si_2$. This is likely caused by a decrease of the f electron count from mainly a $4f^1$ towards a $4f^0$ configuration. The observed changes are consistent with the behavior of other Ce heavy fermion compounds at 300 K. To get a full picture of the valence transition in $CeCu_2Si_2$ and to understand how this correlates to the superconducting properties, we intend to pursue this study by new measurements at room temperature and high pressure in the same sample. Comparison with $CeCu_2Ge_2$ would be instructive to highlight the interplay between the electronic and structural degrees of freedom.

References

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