



	<b>Experiment title:</b> <b>Sm valence and closure of the correlation gap in Kondo insulator SmB<sub>6</sub></b>	<b>Experiment number:</b> HC-4723
<b>Beamline:</b> ID24	<b>Date of experiment:</b> from: 15/02/2022 to: 21/02/2022	<b>Date of report:</b> 03/04/2023
<b>Shifts:</b> 18	<b>Local contact(s):</b> O. Mathon, R. Torchio	<i>Received at ESRF:</i>
<b>Names and affiliations of applicants</b> (* indicates experimentalists): <b>C. Strohm*</b> DESY Department of Photon Science Notkestrasse 85 DE - 22607 HAMBURG <b>F. Duc*</b> LNCMI Laboratoire National des Champs Magnétiques Intenses 143 avenue de Rangueil FR - 31400 TOULOUSE Cedex 04 <b>B. Galaup*</b> LNCMI Laboratoire National des Champs Magnétiques Intenses 143 avenue de Rangueil FR - 31400 TOULOUSE Cedex 04 <b>E. Mijit</b> LNCMI Laboratoire National des Champs Magnétiques Intenses 143 avenue de Rangueil FR - 31400 TOULOUSE Cedex 04		

### Report:

At ambient conditions, SmB<sub>6</sub> is a mixed valence compound with an average Sm valence ( $v_{Sm}$ ) of about 2.6, and behaves like a correlated poor metal [1,2]. However, with the decrease of temperature, it shows a metal-insulator (semiconductor) transition [2]. The narrow gap opening at low temperatures was understood as Kondo lattice behaviour [3], and SmB<sub>6</sub> became the subject of intense research over the years as a prototype Kondo insulator. Temperature dependent x-ray absorption spectroscopy (XAS) measurements have shown that  $v_{Sm}$  reduces upon cooling, indicating an intimate correlation between valence fluctuations and formation of an insulating state in SmB<sub>6</sub>. Besides, the narrow band-gap is known to decrease linearly under pressure, leading to its closure around 5.5 GPa [3]. Ambient pressure resistivity measurements in intense pulsed magnetic fields up to 148 T have shown a minimum at about 89 T [4]. The field induced resistivity minimum was attributed to the closure of the gap, even though the detailed mechanism is not entirely clear.

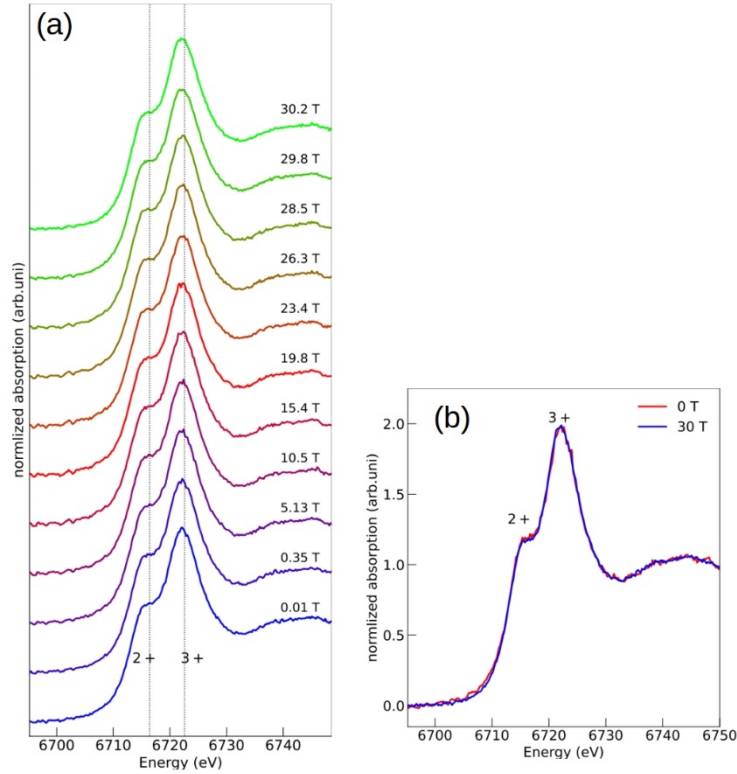
The main **objective** of experiment HC4723 was to confirm previous results and to further investigate the field induced closure of the resistivity gap by direct valence measurements using XAS. The idea was to reduce the critical field lower than 30 T by tuning the gap under high pressure, and perform field dependent XAS measurements at low temperatures (within the insulating phase) to observe a possible increase of  $v_{Sm}$ , as the signatures of field induced gap-closure. A second objective was to probe the magnetic state of the sample via XMCD.

### Datasets:

We measured field dependent XAS measurements at 1.6 GPa. Sm L<sub>3</sub>-edge XAS data have been collected along the rising and falling slopes of 30 T field pulses, at several different temperatures starting from 100 K down to 60 K (with a 10 K step, approximately). An exemplary set of field dependent XAS obtained at 77 K is reported in Fig. 1 (a). Two XAS data measured at lowest (0 T) and highest (30 T) field values are compared in Fig. 1(b), showing nearly identical spectral features.

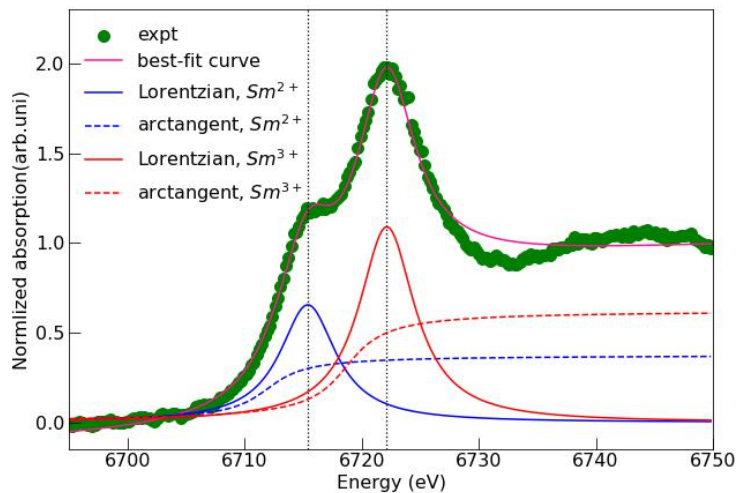
### Data analysis:

As one can see from In Fig. 1 (a,b), the white line range (6710-6730 eV) of individual Sm L<sub>3</sub>-edge XAS spectra of SmB<sub>6</sub> consists of two components, typical of mixed valence compounds. The shoulder like peak at the lower energy side (~ 6715 eV) corresponds to the divalent Sm<sup>2+</sup> component (with 4f<sup>6</sup>5d<sup>1</sup> configuration), while the strong



**Fig. 1: (a) Sm L<sub>3</sub>-edge XAS data measured along a rising slope of a 30 T field pulse, P=1.6 GPa, T=77 K. (b): Comparison of XAS spectra measured at 0 and 30 T**

peak at the high energy side (~ 6722 eV) corresponds to the trivalent Sm<sup>3+</sup> component (with 4f<sup>5</sup>5d<sup>1</sup> configuration). In order to quantitatively estimate  $\nu_{Sm}$ , the spectral weights of Sm<sup>2+</sup> and Sm<sup>3+</sup> components have been evaluated by a peak fitting procedure, as show in Fig.2. Each spectral component was modeled by the sum of a Lorentzian and arctangent functions to mimic the peak shape and corresponding edge jump, respectively. Average valence was then calculated based on the wait of the Sm<sup>2+</sup> and Sm<sup>3+</sup> peaks (peak area of Lorentzian functions). This fitting procedure was repeated (using a batch fitting script) for all the XAS data measured along the field pulse.



**Fig. 2: An exemplary fit for the data measured at P=1.6 GPa, T=77 K, H=0 T.**

## Results:

Following above mentioned procedures, we obtained  $\nu_{Sm}$  for all field values along each field pulse. An exemplary  $\nu_{Sm}$  vs. field plot is given in Fig. 3. We can see from Fig. 3 that average Sm valence remained constant (within the error bar), in magnetic fields up to 30 T. In fact, this not surprising if we look back again at Fig. 1(b), which shows negligible spectral difference for lowest (0 T) and highest (30 T) field values.

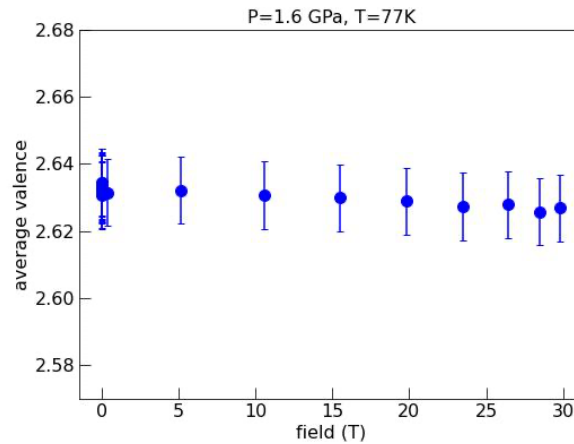


Fig. 3: Average Sm valence as a function of field, obtained from the peak fitting procedures.

### Conclusions:

We did not find significant valence change in pulsed magnetic field up to 30 T, at different

### XMCD:

We attempted to obtain XMCD data, but unfortunately the quality was not sufficient to detect the small expected signal. One possible explanation is a combination of (i) the enhanced spatial coherence of the EBS, (ii) a periodic slope error of the (old) horizontal mirror, and (iii) small (periodic) variations in source position. This ultimately leads to a failure of the normalization procedure of the spectra image and also in the evaluation of difference spectra.

We understood that ID24 is planning to replace the mirror in order to mitigate the issue. At the same time ID24 offered support to attempt to exploit the data through correlation with a (temporally) large set of  $I_0$  acquisitions. This numerical approach is based on the assumption of a periodic temporal pattern or modes in the variation of the source position.

We thank the local contacts and ID24 staff for their excellent support during this successful experiment.

### References:

- [1] M. Mizumaki et al., J. Phys. Conf. Ser 17, 2009.
- [2] A. Mentel et al., Phys. Rev. Lett 22, 1969.
- [3] V. V. Moshalkov et al., JMMM 47&48, 1985
- [4] J. C. Cooley et al., Phys. Rev. B 52, 1995