

	<b>Experiment title:</b> 4 <i>f</i> -5 <i>d</i> interaction studied by RIXS at the Ce $L_3$ edge in highly correlated systems	<b>Experiment number:</b> HE748
<b>Beamline:</b> <b>ID12A</b>	<b>Date of experiment:</b> from: 22 Feb 2000                      to: 1 Mar 2000	<b>Date of report:</b> 25/08/00
<b>Shifts:</b> 18	<b>Local contact(s):</b> Andrei ROGALEV	<i>Received at ESRF:</i>
<b>Names and affiliations of applicants</b> (* indicates experimentalists): * <b>C. F. Hague</b> , * <b>J.-M. Mariot</b> , * <b>L. Journal</b> Université. P. et M. Curie, Laboratoire de Chimie Physique, 75231 Paris Cedex 05 <b>J.-P. Kappler</b> IPCMS, Strasbourg <b>G. Krill</b> LURE, Orsay		

### Report:

In order to help in our interpretation of the highly complex behaviour of the RIXS data which we have obtained concerning mixed valent Ce compounds, we performed some preliminary high resolution  $2p3d$  RIXS experiments on La and Nd compounds at the  $L_{2,3}$  edges. Both types of compounds led to interesting results, but here we concentrate on those for La which are of greater relevance to the studies on Ce. Though the latter are the subject of on-going calculations, it has become clear that a proper understanding of the interactions between  $4f$  and  $5d$  states, where the latter are hybridized with  $3d$  or  $4d$  transition metals or  $p$ -like states, calls for comparable work on the neighbouring elements. A good illustration of the usefulness of such studies is provided by comparing the data we have obtained for the  $\text{CeF}_3$  and  $\text{LaF}_3$  ionic compounds.

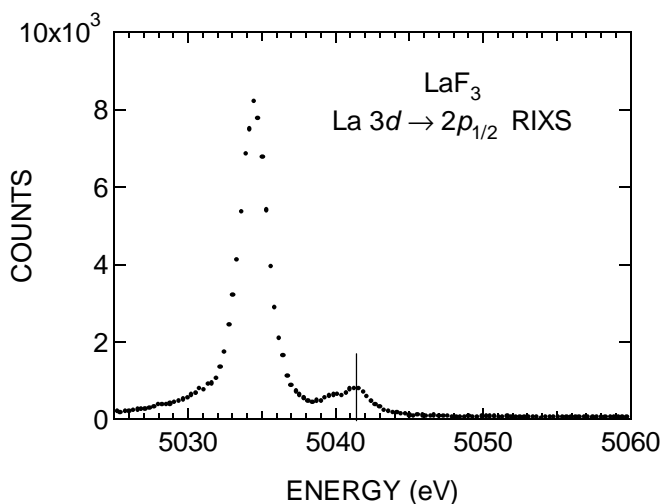
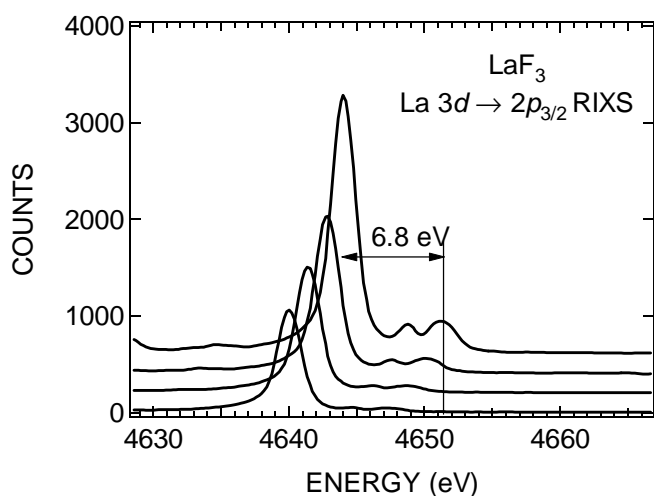
Our earlier experiments on  $\text{CeF}_3$  (see ESRF Highlights 97/98, and Gallet et al 1999) revealed the presence of two low intensity peaks in the Raman regime attributed to quadrupole excitations. The two peaks separated by approximately 3 eV are interpreted as arising from excitations to parallel and antiparallel alignments of the two  $4f$  electrons in the presence of the final-state  $3d$  hole, i.e.,  $4f^1 5d^0 \rightarrow 2p^5 4f^2 5d^0 \rightarrow 3d^0 4f^2 5d^0$  (see Bartolomé et al 1997, 1999 and Gallet et al 1999).

It is striking to note that such a two-peak structure is also observed in the La  $L_3$  RIXS (Fig. 1a). Here the ground state is  $4f^0 5d^0$  so the interpretation is not the same as that given for the structure in Ce. Instead the two-peak structure must be the result of the alignment of a  $4f$  electron whose spin is parallel or anti-parallel to that of the  $3d$  core-hole. It therefore

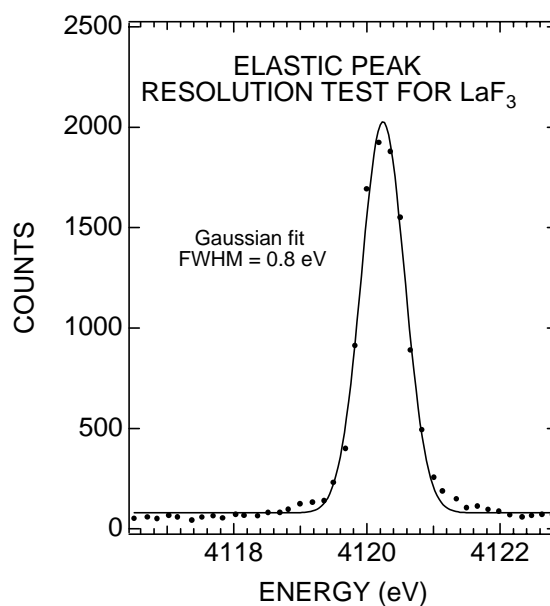
necessarily involves the high probability of a spin flip in the overall excitation-decay process. This in turn means that the spin is not a good quantum number as a consequence of spin-orbit interaction. An atomic multiplet calculation is underway to understand the exact origin of this unexpected structure

We also performed RIXS measurements at the La  $L_2$ -edge in  $\text{LaF}_3$  (Fig. 1b). A quadrupole transition is clearly observed. From XAS experiments it was previously concluded that quadrupole transitions were not observable at the rare earth  $L_2$ -edges (see van Veenendaal et al 1997). We also measured similar structure in a La intermetallic (not shown).

Note that we have been able to improve the total resolving power to  $\sim 5000$  (Fig. 2) which is close to the limit which can be obtained with a two crystal monochromator. We believe that under the present operating conditions our x-ray spectrometer, despite its very compact transportable size, introduces a broadening of  $\sim 0.2$  eV.



**FIG 1a (top panel)**  
**FIG 1b (lower panel)**



**FIG 2**

## REFERENCES

- F. Bartolomé et al., Phys. Rev. Letters **79**, 3775 (1997); Phys. Rev. B **60**, 1162 (1999).  
 J.-J. Gallet et al., Phys. Rev. B **54**, 14238 (1997).  
 J.-J. Gallet et al., Phys. Rev. B **60**, 14128 (1999).  
 M. van Veenendaal et al., Phys. Rev. Letters **78**, 1162 (1997).