


**Experiment title:**
*Correlation between hybridisation and resonant Raman scattering from Cerium compounds in the M<sub>5</sub> region with final state 4p hole*
**Experiment**
**number:**

HE546

**Beamline:**

ID12B

**Date of experiment:**

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**Date of report:**

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**Shifts:**

15

**Local contact(s):**

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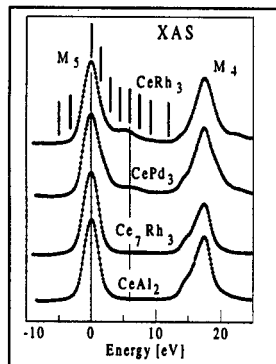
**Report:**
**Aim of the experiment**

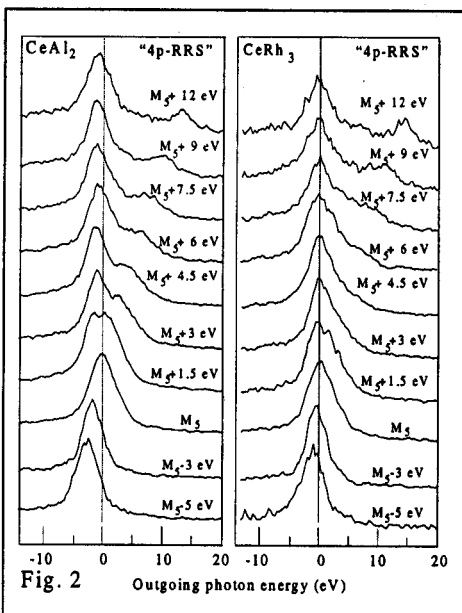
The aim of the experiment was a first exploration, in different Ce intermetallics, of the behaviour of Ce resonant Raman scattering with M<sub>5</sub> excitation and 4p final hole. The process is  $3d^{10}4f^n \rightarrow 3d^9 4f^{n+1} \rightarrow 3d^{10} 4f^{n+1} 4p^5$  in a simplified notation where n is the 4f occupation in the ground state and the final state configuration interaction (CI) with the  $4d^8 4f^{n+2}$  is neglected. We will call this process 4p-RRS [1]. The experiment was motivated by a hint we obtained previously in the comparison between a  $\gamma$ -like i.e. a poorly hybridised system (Ce<sub>7</sub>Rh<sub>3</sub>) and an  $\alpha$ -like compound i.e. a strongly hybridised system (CeRh<sub>2</sub>). We noticed that the 4p-RRS spectra with excitation in the satellite about 6 eV above M<sub>5</sub> are sensitive to the change from  $\gamma$  to  $\alpha$ -like systems. This motivated a general assessment of this effect by exploring the 4p-RRS in a variety of Ce compounds ranging from  $\alpha$  to  $\gamma$ -like and with a variety of excitation energies in each compound. This work has been done in the present experiment.

**Results and Discussion**

We have measured the 4p-RRS in the following samples going from more  $\alpha$ -like to more  $\gamma$ -like: CeRh<sub>3</sub>, CeNi<sub>2</sub>, CePd<sub>3</sub>, Ce<sub>7</sub>Rh<sub>3</sub> and CeAl<sub>2</sub>. For space reasons we report only a selection of the data which is sufficient to show the main conclusions.

The M<sub>4,5</sub> absorption curves (XAS) are reported in figure 1, where the vertical bars indicate the excitation energies used in the Raman experiment. The broad bandwidth used for the XAS of figure 1 and for all the 4p-RRS was dictated by the need of having a reasonable counting rate in the emission spectra. In figure 2 we give a set of 4p-RRS spectra in the





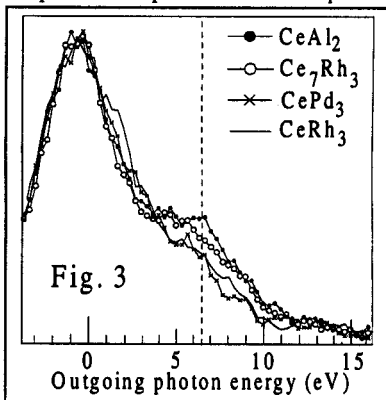
two extreme cases of  $\text{CeAl}_2$  and  $\text{CeRh}_3$ . The results show the following points supported also by the 4p-spectra of the other compounds.

- The behaviour of  $\alpha$ -like and  $\gamma$ -like systems is very similar in the  $M_5$  region from below threshold to about 2 eV above threshold. The same happens also with excitation far above threshold (typically 12 eV).

- A clear difference is seen when the excitation is between about 3 eV and 7 eV above  $M_5$  i.e. in the satellite region of the  $\alpha$ -like XAS. This is probably due to the spread of the multiplet splitting, at this excitation, in strongly hybridised systems. Thus we arrive to the new conclusion that this is a general behaviour and that only this excitation energy range is useful to distinguish between systems having different ground state properties.

With the excitation energy in the 6 eV satellite an even more interesting result has been obtained in the comparison between the different compounds. This is shown in Fig 3 where four compounds are compared. The sensitivity of the 4p-RRS to the change of the 4f population is considerably stronger in the  $\gamma$ -like region than in the  $\alpha$ -like region of the parameters space. In fact the 4p-RRS

distinguishes rather well between two  $\gamma$ -like compounds ( $\text{Ce}_7\text{Rh}_3$ ) and  $\text{CeAl}_2$  whereas the difference is very small in other bulk sensitive spectroscopies such as XAS. In this case the 4p-RRS acts as an "amplifier" of the effect of tiny changes of the 4f occupation in the region just below unity. This is a theoretically challenging result that candidates the 4p-RRS as a new bulk sensitive tool in the study of poorly hybridised Ce systems.



#### References

- [1] A. Tagliaferri *et al.*, Phys. Rev. B60, 5728 (1999).