ESRF	Experiment title: Resonant Auger in La compounds	Experiment number: SI-461
Beamline:	Date of experiment:	Date of report:
ID32	from: 1 st July 1999 to: 10 th July 1999	12 th January 2000
Shifts:	Local contact(s): B. Cowie	Received at ESRF:
21		
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Report:

Resonant spectroscopies are powerful tools for the study of electronic structure of solids. In highly correlated systems, they allow to understand the transitions occuring in the absorption edges, and to probe the degree of localization of selected orbitals. In previous experiments, we had been able to attribute the main structures of the Ce L₃ absorption edge, in mixed-valent compounds, using resonant 3d XPS [1]. Since our measurements, theoritical calculations have confirmed the interpretation of our results [2]. We had also obtained resonant Auger results, at the Ce L₃ edge, showing a Raman-Auger behaviour up to 10 eV above the absorption edge jump : this showed the strongly localized character of the 5d orbitals, in the presence of the 2p core hole [1]. Unfortunatly, these results are difficult to interprete quantitatively :

-First, in mixed-valent compounds, Ce has a ground state consisting in a mixture of $4f^0$ and $4f^1$ configurations. In the excited final state, a $4f^2$ configuration is also allowed. These different electronic configurations give numerous structures in the Auger spectra, often mixed together.

-Second, the number of open shells both in the initial and the final state renders difficult a calculation of the spectra, even in an atomic approach. Moreover, the description of mixed-valent compounds requiers the introduction of hybridization between the 4f states and the conduction band.

For these reasons, we proposed the study of La alloys, where the situation is much simpler : La has a $4f^0$ ground state. The resonant Auger spectra obtained at the La L₃ edge, on LaAl₂ and LaPt, for the L₃M_{4,5}N_{4,5} Auger lines, are presented on the figure. They show the expected behaviour for Raman-Auger lines in a metal, with peaks appearing at roughly constant binding energy, below the absorption threshold. The classical Auger spectrum, at constant kinetic energy appears above the maximum of the L₃ absorption edge white line, at 5486 eV. The two main lines are due to $L_3M_4N_{4,5}$ and $L_3M_5N_{4,5}$ Auger decays.

The most interesting feature in these results is certainly the peak at high kinetic energy, indicated by arrows on the figure. This peak appears at increasing kinetic energy in the rise of the absorption threshold, and resonates before the maximum of the white line. We think that it is due to quadrupolar transitions $2p\rightarrow4f$, occuring in this photon energy range. As a matter of fact, a very localized 4f electron is very efficient to

screen the core hole, and will lower the final energy of the La atom. As a consequence, the Auger decay appears at a higher kinetic energy.

These quadrupolar transitions are less important in $LaAl_2$ than in LaPt. This is certainly due to a conduction band less localized in $LaAl_2$ (made of the La 5d and Al s-p electronic states) than in LaPt (La and Pt 5d electronic states). Obviously, multielectronic effects, like Raman-Auger processes, are more likely if the 2p electron is excited to localized states.

As a conclusion, we showed that quadrupolar transitions occur in the rise of the La L_3 edge. They were detected by resonant Auger, an electronic resonant spectroscopy, much easier to use in metals than inelastic X-ray scattering. We have obtained the same results at the La L_2 edge. Forthcoming calculations should confirm the interpretation of our resonant Auger spectra.



Figure : Top : La L_3 absorption edge in LaAl₂ (left) and LaPt (right). Bottom : Auger spectra obtained for the indicated photon energies in the two compounds.

References :

[1] P. Le Fèvre, H. Magnan, D. Chandesris, J. Vogel, V. Formoso, F. Comin, Phys. Rev. B 58, 1080 (1998).
[2] H. Ogasawara, A. Kotani, P. Le Fèvre, D. Chandesris, H. Magnan, submitted to Phys. Rev. B (1999).