ESRF	**Experiment title:** Lattice dynamics in lanthanide zirconate pyrochlores for nuclear waste disposal and thermal barrier coatings	**Experiment number:** HC-824
Beamline: ID18	**Date of experiment:** from: 18.05.2013 to: 22.05.2013	**Date of report:** 27.08.2012
Shifts: 12	**Local contact(s):** Dimitrios Bessas (email: bessas@esrf.fr)	*Received at ESRF:*

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Report:

Within the beam time allocated for proposal HC-824 the vibrational properties of six different Eu containing lanthanide zirconates were investigated using nuclear inelastic scattering (NIS) by the ^{151}Eu Mössbauer resonance. Additionally, hyperfine interactions in all compounds were investigated using nuclear forward scattering (NFS).

In order to account for the potential areas of application of this class of compounds, the compounds $Eu_2Zr_2O_7$, $EuNdZr_2O_7$ as well as $Eu_{0.5}Nd_{1.5}Zr_2O_7$ were investigated both in pyrochlore as well as in defect fluoride structure. In particular, the transition of pyrochlore to defect fluoride structure is of special importance in the context of nuclear waste management, since it is accompanied by a substantial increase of amorphization resistance [1,2].

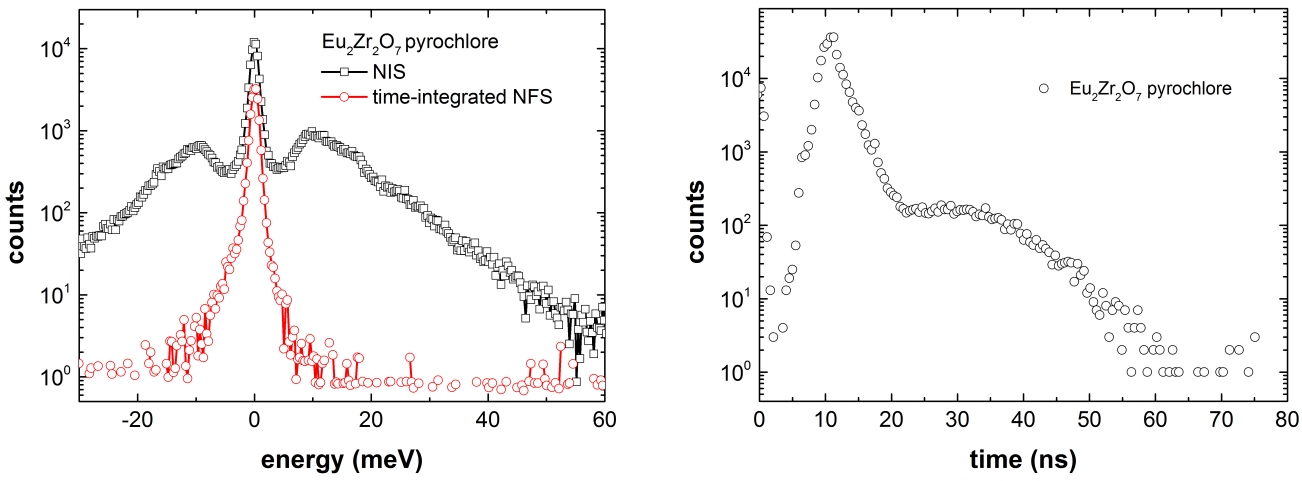

*Fig. 1: Representative NIS (**left**) and NFS (**right**) spectra for $Eu_2Zr_2O_7$ in pyrochlore structure.*

In Figure 1 representative NIS and NFS spectra obtained at room temperature for the pure europium zirconate compound in pyrochlore structure are shown. Concerning NIS measurements an experimental resolution of about 1.4 meV in terms of full width at half maximum (FWHM) could be achieved. Concerning time resolved NFS measurements data obtained about 10 ns after the prompt synchrotron radiation pulse, i.e. electronically scattered radiation, could be used for further data analysis.

While the Eu specific density of phonon states (DPS) is extracted using the Fourier-Log decomposition procedure as implemented in the program DOS [3], NFS data is analyzed using the program MOTIF [4]. Representative examples of data analysis are shown in Figure 2.

Both the structural transition and the substitution of Eu with Nd, which is not shown in this report, have significant impact on the Eu partial DPS and seem to affect mainly low energetic phonon modes.

The quadrupole splitting values as obtained from NFS analysis are comparable to literature data for $Eu_2Zr_2O_7$ in pyrochlore structure [5] and are also consistent with in-house Mössbauer spectroscopic measurements for all compounds. High temperature NFS measurements, which were also performed, will be analyzed with respect to the Goldanskii-Karyagin effect, which is documented for the similar $Eu_2Ti_2O_7$ [5].

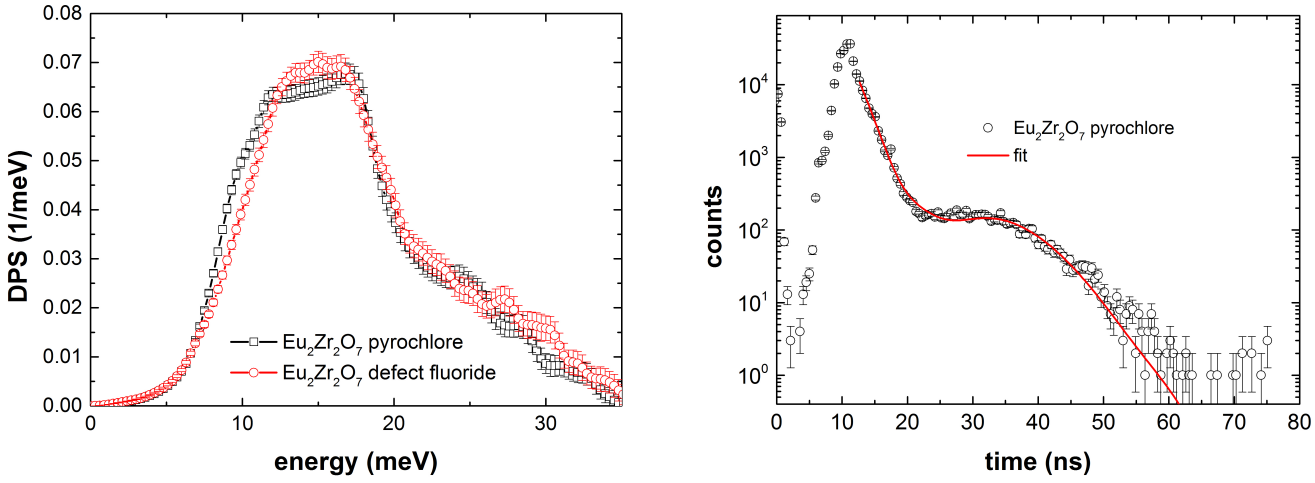

Fig. 2: Representative examples of data analysis. (**left**) Eu specific DPS for $Eu_2Zr_2O_7$ both in pyrochlore as well as in defect fluoride structure obtained using the program DOS. (**right**) The analysis of NFS using the program MOTIF (red line) yields a quadrupole splitting energy of -4.19(4)*Γ_0, where Γ_0 is the natural linewidth of ^{151}Eu, in the case of pyrochlore $Eu_2Zr_2O_7$.

References

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