| ESRF | **Experiment title:** Lattice dynamics of Europium monoxide and Europium sesquioxide by nuclear inelastic scattering and first principles theory | **Experiment number**: HC 1906 |
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| **Beamline**: ID18 | **Date of experiment**: From 13/05/15 to 019/05/15 | **Date of report**: 18/08/15 |
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*The aim of this experiment was to systematically investigate the temperature and thickness dependence on vibrational dynamics and thermodynamic properties of epitaxial EuO films by in-situ nuclear inelastic scattering on* $^{151}Eu$*. Additionally lattice dynamics of polycrystalline* $Eu_2O_3$ *sample was investigated.*


**Report**

The experiment can be divided into two parts, namely thickness dependent and temperature dependent lattice dynamics investigations. The thickness dependent *in-situ* experiments were performed on EuO(001) ultra-thin films grown on yttrium aluminium oxide according to reference [1]. However the growth parameters had to be adapted to achieve films with smooth surface at low coverages – 30ML, 2ML, and 1ML (ML- monolayer). The samples were characterised by *in-situ* reflection high energy electron diffraction (RHEED).

The growth and *in-situ* nuclear inelastic scattering (NIS) were performed at ID18 of the ESRF using the ultrahigh vacuum facility [2] installed in the second experimental hutch. We point out that this is the only place worldwide where thin films and nanostructures can be grown, characterized, and investigated by *in-situ* nuclear resonant scattering methods. We used the 16-bunch filling mode of the ESRF storage ring. The final energy bandwidth of 1.0 meV (at the resonance energy of 21.5 keV) was obtained using a dedicated high-resolution monochromator. The vertical beam size was reduced down to 20 µm using Kirkpatrick-Baez focussing mirror. To suppress the multi-phonon excitations the samples were measured at 100 K.

The temperature dependent lattice dynamics experiment was performed on epitaxial 100 nm thick EuO film grown on Yttria stabilised zirconia (YSZ) according to the growth conditions described by Sutarto et al. [3] in our home lab at ANKA, Karlsruhe. The surface morphology was investigated by *in-situ* RHEED and the stoichiometry was confirmed by *in-situ* x-ray photoemission spectroscopy (XPS). The sample was then capped with 10nm thick Nb layer to prevent further oxidation upon exposure to air. The NIS experiment was performed in the first experimental hutch of the ID18 beamline in He flow cryostat at various temperatures – 30K, 50K, 70K, 90K and at room temperature.

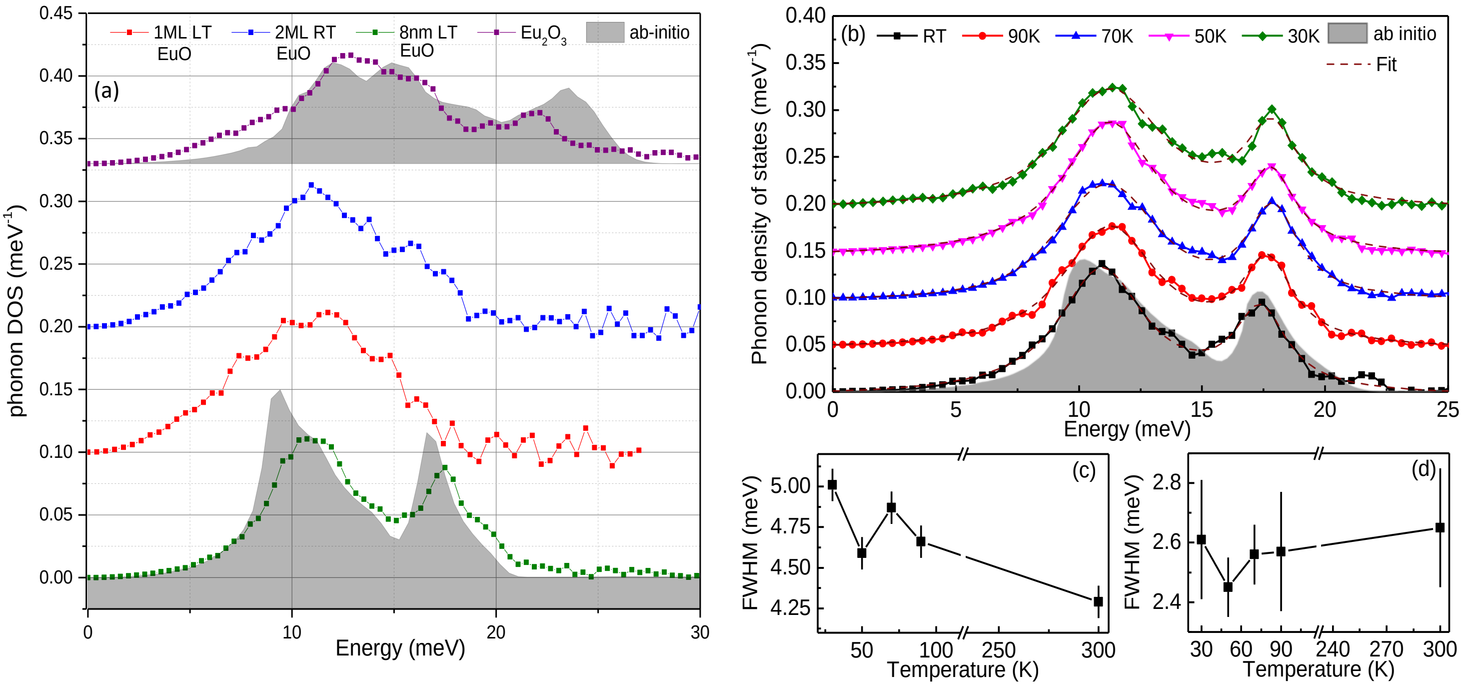


***Figure 1.*** *(a) The partial Eu-projected phonon DOS of EuO obtained from the nuclear inelastic scattering spectra on films with various thicknesses. 2ML film and $Eu_2O_3$ sample were measured at room temperature (RT) while other EuO samples were measured at 100K (LT). (b) The partial Eu-projected phonon DOS of EuO obtained from the nuclear inelastic scattering spectra on 100 nm EuO at different temperatures. The corresponding DOS calculated from first principles is represented by the shaded area. (c) and (d) are the FWHMs as a function of temperature of the voigt peaks at approximately 11 meV and 17 meV respectively.*

The experimental results of the thickness dependent studies show a good agreement between the measured and calculated phonon density of states for the thick film while pronounced deviation from the bulk DOS were observed for the ultrathin films. The minor overall shift in the phonon energies in the bulk like film can be explained by the slightly overestimated lattice parameter used in the calculations. On the other hand, the ultrathin films demonstrate a shift in the spectral mass toward lower energies, which we attribute to an enhanced contribution from the surface modes. Ab-intio calculations for the EuO surface are in progress. This effect opens up new questions related to effects of magnetic ordering on phonons in such ultrathin oxide films.

The temperature dependent studies on bulk-like EuO films served as a complimentary experiment to our results from the inelastic x-ray scattering experiment at ID28 (see Exp Rep. HC1934) where we observed a broadening in certain branches of the dispersion relations. The spectra were fitted with two Voigt profiles to estimate the broadening. As figure 1(c) and (d) suggests, the peak at 11 meV which is mostly contributed by transverse acoustic modes broadens as temperature is lowered while the other peak at 17 meV shows no significant change with temperature. However, such an effect was not predicted by our calculations and we speculate that this broadening originates from the coupling of phonons to spin fluctuations near the Curie temperature (69K) [4]. Further ab-initio calculations for EuO in the antiferromagnetic state that will shed more light on this phenomenon are in progress.

Besides the measurements on EuO, NIS was performed on a pellet made from commercially available high purity $Eu_2O_3$ at room temperature. The obtained phonon DOS is in good agreement with our ab-initio calculations (fig. 1 (a)).

**References**


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