ESRF	**Experiment title:** Density of phonon states of ultrathin europium metallic films from *in-situ* ^{151}Eu nuclear inelastic scattering	**Experiment number**: SI-2364
Beamline: ID18	**Date of experiment**: from: 15/08/11 to: 29/08/11	**Date of report**: 18/02/12
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Report:

In this experiment we studied the density of phonon states (DOS) of single crystalline epitaxial Eu films exhibiting either the native for this metal *bcc* crystal structure or the characteristic for the heavy lanthanides hexagonal close-packed (*hcp*) crystal lattice.

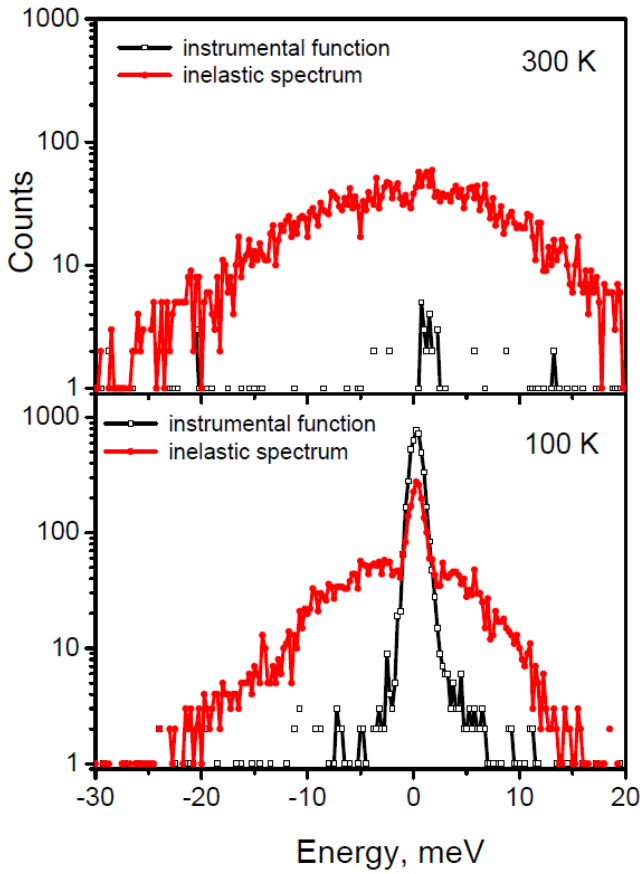

Fig. 1 Spectra of nuclear inelastic absorption obtained upon 1.1 nm thick *hcp* Eu film at room temperature (top graph) and at 100 K (botom graph).

The experiment was performed at ID18 of the ESRF using the ultrahigh vacuum facility [1] 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.

Europium films with various thicknesses ranging from 1.1 nm up to 60 nm were grown epitaxialy on Nb buffered Al_2O_3 following an elaborated procedure [2]. Up to a film thickness of about 6 nm Eu grows in the characteristic for heavy lanthanides *hcp* structure. Above this thickness the film relaxes into the native for Eu *bcc* lattice. The films were grown from high-purity Eu metal enriched in 95% to ^{151}Eu. The film structure was controlled by reflection high-energy electron diffraction (RHEED) immediately after the growth, as well as after the end of the inelastic experiment. The vacuum during the sample growth and the experiment was equal or better

than $5x10^{-10}$ mbar.

The experiment was performed during 16-bunch filling mode of the ESRF storage ring operation. 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 beamsize was reduced down to 20 μm using Kirkpatrick-Baez focussing mirror

Our previous experiments on thick Eu [3] and Sm [4] films demonstrated that measurements at room temperature are not informative. Due to the relatively low Debye temperature of these metals the multiphonon excitations represent a significant part of the measured signal at room temperature. Fig. 1 shows the spectrum of nuclear inelastic absorption derived from 1.1 nm thick Eu film at room temperature and at 100 K. It is directly seen from the raw data that the room temperature spectrum is featureless and the phonon DOS cannot be derived. Cooling down of the film to 100 K is already enough to extract the single-phonon term and to calculate the phonon DOS of the film.

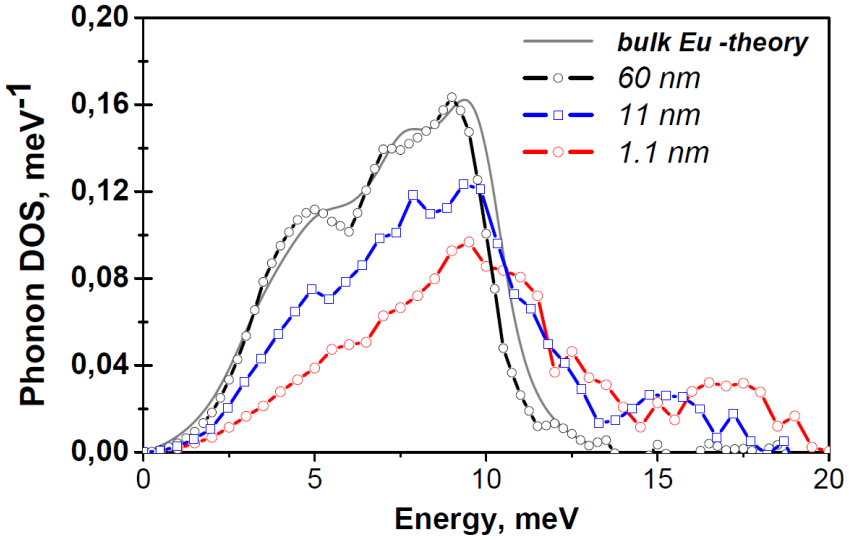

Fig. 2. Phonon DOS for selected Eu films derived from the spectra of inelastic absorption measured at UHV and at 100 K.

Fig 2 shows the density of phonon states for selected Eu films with various thicknesses derived from the inelastic spectra measured at 100 K. The 60 nm thick film (black line and open circles) shows features very close to those of calculated bulk Eu (solid grey line) from first principles [3]. The observed small shift towards lower energy is attributed to strain within the film.

Quite surprisingly, reduction of the film thickness leads to suppression of the phonon modes at lower energies and enhancement of the states at high energies (Fig. 2, Eu film with thickness 11 nm). This behaviour contradicts with the reported in the literature phonon softening in thin films (see e.g. [5, 6]). Moreover, further reduction of the film thickness to 1.1 nm results in even more pronounced hardening of the phonon spectrum (Fig. 2, Eu film with thickness 1.1 nm), i.e. this behaviour has a systematic character.

The observed shape of the phonon DOS is determined by three factors: (1) the ultrathin Eu film exhibits *hcp*, instead of the native for europium *bcc* lattice; (2) the presence of free surface, i.e. broken translational symmetry, and (3) the large misfit parameter (~ 14%) between the lattice constants of Eu layer and bulk Eu. It seems that the high compressive strain plays a decisive role on lattice dynamics of the ultrathin Eu films. To get a detailed understanding and to disentangle quantitatively between different contributions more systematic investigation is necessary, as well as a comparison with the results from the on-going *ab-initio* calculations.

References:

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