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UCRL-TR-218876

FY05 LDRD Final Report Mapping Phonons at High-pressure

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February 13, 2006

Disclaimer

Auspices Statement

This work was performed under the auspices of the U. S. Department of Energy (DOE) by the University of California, Lawrence Livermore National Laboratory (LLNL) under Contract No. W-7405-Eng-48. The project (04-ERD-106) was funded by the Laboratory Directed Research and Development Program at LLNL.

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In order to shed light on the intriguing, and not yet fully understood fcc-isostructural $\gamma \rightarrow \alpha$ transition in cerium, we have begun an experimental program aimed at the determination of the pressure evolution of the transverse acoustic (TA) and longitudinal acoustic (LA) phonon dispersions up to and above the transition.

γ-Ce Crystals of 60-80 μm diameter and 20 μm thickness were prepared from a large ingot, obtained from Ames Lab, using laser cutting, micro-mechanical and chemical polishing techniques. Three samples with a surface normal approximately oriented along the [110] direction were loaded into diamond anvil cells (DAC), using neon as a pressure transmitting medium. The crystalline quality was checked by rocking curve scans and typical values obtained ranged between one and two degrees. Only a slight degradation in the sample quality was observed when the pressure was increased to reach the α-phase, and data could be therefore recorded in this phase as well. The spectrometer was operated at 17794 eV in Kirkpatrick-Baez focusing geometry, providing an energy resolution of 3 meV and a focal spot size at the sample position of 30 x 60 μm^2 (horizontal x vertical, FWHM).

Eight to ten IXS spectra were typically recorded per phonon branch. Figure 1 reports the pressure dependence of the LA[100] branch in the γ-phase for pressures of 1, 4 and 6 kbar, together with previous inelastic neutron scattering (INS) results [1] at ambient pressure. A clear decrease of the phonon energies with increasing pressure is observed for 1 and 4 kbar, whereas the phonon energies increase again at 6 kbar, still well within the stability field of the γ-phase. Figure 2 reports the LA dispersion along all three main symmetry directions at 6 kbar (γ-phase) and 8 kbar (α-phase), together with the INS results at ambient conditions. Besides the already discussed unusual behaviour along the [100] direction, the pressure evolution of the two other longitudinal branches in the γ-phase is quite different. The LA [110] branch displays a downward bending near the zone boundary (ZB), whereas the phonon energies at low reduced momentum transfer remain close to the ones at

room pressure. In contrast to this, the LA [111] branch does not display any pressure dependence.

The LA phonon energies in the α-phase at 8 kbar are systematically higher than the corresponding lower pressure phonon energies, consistent with the higher density of the α-phase and the expected larger elastic constants. We note, however, substantial changes in the lattice dynamics along the [110] direction. While the phonons between ξ=0.4 and 0.6 show a large energy increase with pressure, the phonon energy decreases at the zone boundary, thus leading to a pronounced overbending of the branch. The shape of the LA phonon branches in the α-phase are close to those measured in thorium at ambient conditions [2] while the γ-phase phonon dispersion resembles fcc metastable lanthanum [3]. This behaviour might be a signature of substantial changes in the Fermi surface topology, leading to significant changes in the electron-phonon coupling mechanism. A Born-von Karman fit to the phonon dispersion is currently being performed in order to quantify the changes in the force constant matrix.

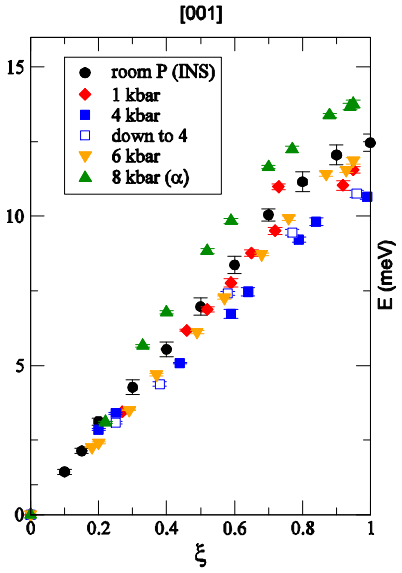

Figure 1

Evolution of the LA [001] branch with pressure.

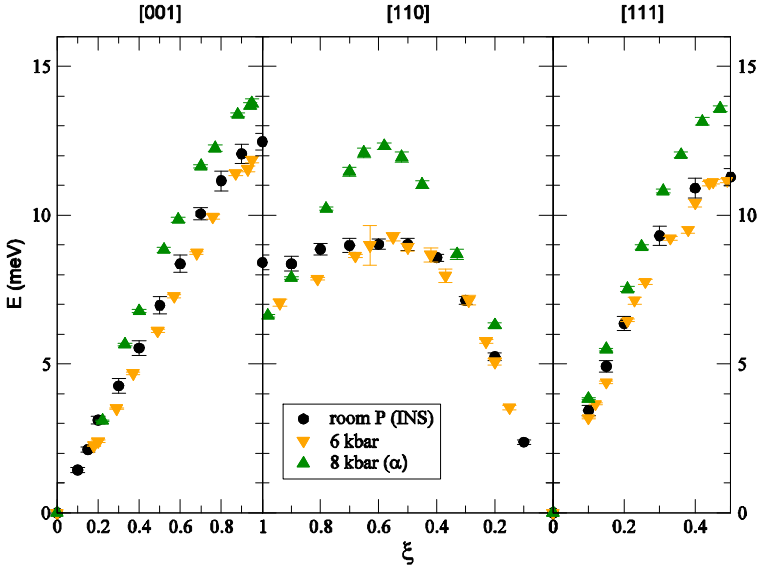

Figure 2

Full set of LA phonon branches at room conditions, 6 kbar and 8 kbar.

The present study was limited to the longitudinal dynamics. This was partly due to the limited time, but as well due to the fact that the samples had a slight miscut (angle between the surface normal and the [110] direction). This in conjunction with the limited angular opening of the DAC (30 degrees), this prevented us from measuring the TA phonons. We are in the process of preparing the next batch of samples with the goal of reducing the miscut angle to less than two degrees, thus

allowing us to focus on the pressure evolution of the transverse dynamics, most notably the TA[111] branch.

References

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[2] *"Phonon spectrum of thorium."*, R.A. Reese et al.; Phys. Rev. B **8**, 1332 (1973).
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