| ESRF | **Experiment title:** Structural changes in $CaSiO_3$ glass at high pressure using X-ray Raman scattering | **Experiment number**: HC-4278 |
|---|---|---|
| **Beamline**: ID20 | **Date of experiment**: from: 27.10.2020 to: 03.11.2020 | **Date of report**: 24.03.2022 |
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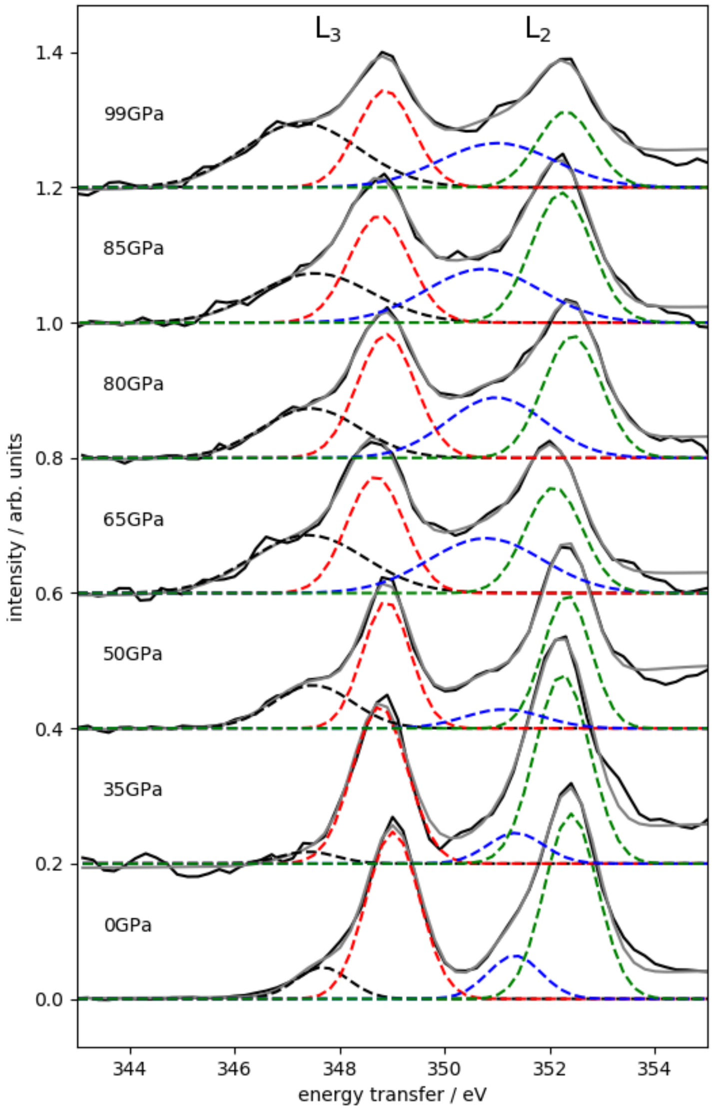
**Report:**

The density and structure of silica melts is extremely important for understanding the crystallization sequences in the Earth's mantle as well as mixing of multi-component melts in the deep Earth. At a previous beamtime (HC-3755) we collected high quality data on $CaSiO_3$ glass up to a pressure of 69 GPa using X-ray Raman scattering (XRS) spectroscopy at the calcium L- and oxygen K-edges. This beamtime is a follow up experiment and was necessary to extent the data set to pressures of the lower mantle.

We conducted high-pressure XRS experiments on pure $CaSiO_3$ glass up to 99 GPa. The high pressure conditions were achieved by Diamond Anvil Cells (DACs). $CaSiO_3$ glass was loaded without a pressure transmitting medium using both beryllium and rhenium gaskets in combination with mini-diamonds. The experiments were performed at beamline ID20-EH3 with an incident energy around 10 keV using the Si(111) monochromator in combination with the Si(311) channel-cut. Oxygen K-edge and calcium L-edge spectra were measured in order to analyze the compaction behavior of oxygen and of the network modifier Ca, respectively. Ca-$L_{2,3}$-edges are shown in Fig. 1 for all pressures accessed in this beamtime.

The analysis of the spectra reveals a consistent increase of both satellite-peaks (see black and blue dashed lines in Fig. 1) of the calcium $L_3$- and $L_2$-edge while the main-peak intensity is decreasing. The main peaks (red and green dashed lines in Fig. 1) are due to spin-orbit coupling whereas the satellite peaks are strongly affected by the crystal field splitting reflecting changes in the local environement of the Ca atoms. The variation of the intensities with pressure are presented in Fig. 2, combining the results of both beamtimes. Moreover, the O-K-edge onset (not shown here) shows a gradual behavior with slight changes in slope at about 30 GPa. In the next step, the results will be compared to ab initio calculations of the XRS spectra based on molecular dynamics simulations in order to interpret them in the framework of structural transitions and pressure-induced coordination changes.



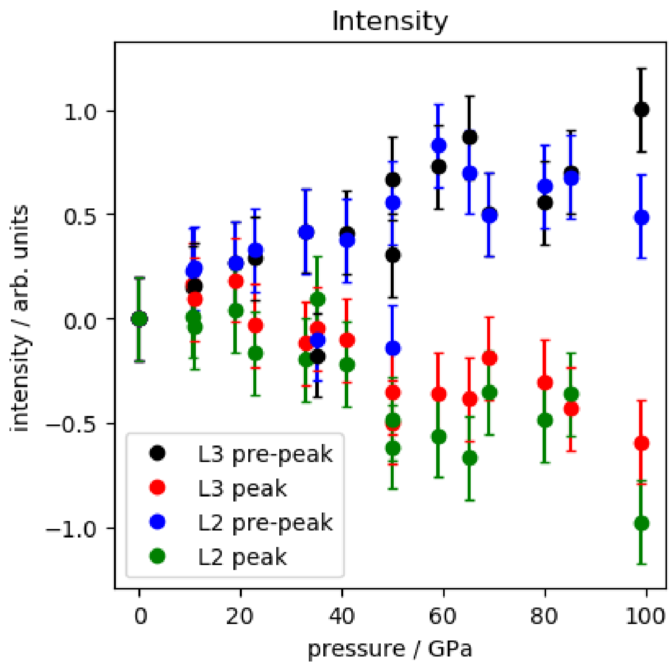




**Fig. 1 (left)**: Measured Ca $L_{2,3}$-edge XRS spectra up to 99 GPa (black) along with a fit of 4 gaussians. The single gaussians are visualized by dashed lines representing the main lines (red and green) and the satellite lines (black and blue).

**Fig. 2 (right)**: Relative change of pre- and main-peak intensity analyzed for calcium $L_3$- and $L_2$-edges (colorcode as Fig. 1).