Liquid-liquid transition in sulfur: structural evolution of the low- and high- density liquids on approaching the critical point

A liquid-liquid transition (LLT) is a first-order transition separating two liquid phases of the same single-component substance. Despite the long-standing and wide interest in this phenomenon, it remains poorly understood and unambiguous experimental realizations are scarce. We reported in 2020 the experimental discovery of a LLT in compressed liquid sulfur in the range 0-2.15 GPa, 450-1040 K [1]. Moreover, we conveyed evidence for a critical point ending the LLT line, never observed in any other system so far. This proposal aims at better understanding the driving mechanisms and order parameter of the LLT in sulfur by studying the nature of the two liquid phases and their structural evolution with pressure and temperature up to the critical point. Using wide-angle x-ray diffraction on sulfur samples inside resistively-heated diamond anvil cells, we will obtain high-resolution pair distribution function as a function of pressure along several isotherms from 700 to 1040 K.

During this beamtime, x-ray diffraction of liquid sulfur was studied in either externally resistively heated and internally resistively heated DACs inside a vacuum chamber, along several isotherms. The DACs were equipped with large culet-size (1 mm) anvils suitable for the low P range of the experiment, and were designed for a wide angle XRD with a total angular x-ray aperture of 100°. In order to obtain a good signal-to-noise ratio at high Q for a low x-ray scatterer like sulfur, the Soller slits developed by ID27 were used to filter out the Compton contribution from the diamond anvils. Experiment was performed at 46 keV in order to obtain diffraction patterns up to 16 $Å^{-1}$. At the beginning of each run, diffraction patterns of the empty cell were collected to obtain the accurate background signal of the experiment.

For the externally resistively heated cells, x-ray diffraction has been performed at 620 K, 700 K and 800 K, and were of excellent quality. Pressure was obtained from the measured lattice parameters of NaCl at the frontier with the sulfur sample, and temperature measure by a thermocouple in contact with the anvils.

The internally resistively heated cells represented a much harder technical challenge, but eventually the setup was successful and experiment made possible at ID27. Diffraction patterns were collected along one isobar at 1.2 GPa during the heating process, up to 1000 K, followed by an isotherm at this given temperature up to 3.2 GPa. Temperature was then increase again at 1075 K where another isotherm was followed to collect diffraction pattern by decreasing the pressure inside the cell up to 1 GPa.

Resulting from the beamtine, excellent resolution structural data for the LDL and HDL of sulfur as a function of P and T were collected. Data are soon going the be processed in order to accurately extract the PDF of the liquid, providing a link between structural variation and density jump across the LLT as a function of T, and valuable information on the driving mechanisms of the LLT.