Experimental X-ray Studies of Liquid Polymorphism

Frédéric DATCHI (Sorbonne Université, CNRS, MNHN, FR),
Mohamed MEZOUAR (ESRF, FR),
Rajaji VINCENT (Sorbonne Université, CNRS, MNHN, FR),
Max GERIN (ESRF,FR),
Laura HENRY (SOLEIL, FR),
Martin DEMOUCRON (ESRF, FR),
Lakou PETIT DIT DARIEL (Sorbonne Université, CNRS, MNHN, FR),
Ciprian G. PRUTEANU (Edinburgh Univ, UK).

Abstract

The liquid-liquid transition (LLT) is a rare and intriguing phenomenon is which a single-component liquid transforms into another one via a first-order transition. Due to their counterintuitive nature, LLTs have intrigued scientists for many years and challenged our perception of the liquid state, for which the notion of polymorphism was long considered impossible. Since the early 1970's, LLTs have been predicted from computer simulations of several systems, and heavily debated in the case of water. So far though, experimental evidence remain scarce and often controversial as they mostly concern supercooled, i.e. metastable, liquids. Meanwhile, our understanding of the LLT remains quite primitive, and there is no theory able to predict whether a given system will exhibit this phenomenon.

Clear, in situ evidence of a LLT in the stable liquid domain have so far been obtained in two cases, phosphorus [1-3] and sulfur [4]. In both cases, this was established by investigating the high-pressure, high temperature liquids using synchrotron x-ray sources. By combining several x-ray-based techniques, important information may be accessed, such as the liquid structure, its density and density-contrast images of phase coexistence which can firmly establish the occurrence of a LLT.

In this talk, I will present results obtained by our consortium from recent experiments at the ID27 beamline of ESRF and PSICHE beamline of SOLEIL. Both beamlines have made large efforts to develop and implement instrumental and analytical tools for the investigation of disordered systems under high pressure [5,6]. After reviewing these techniques, I will present and discuss our experimental data obtained on two systems, realgar (As_4S_4) [7] and As-P alloys.

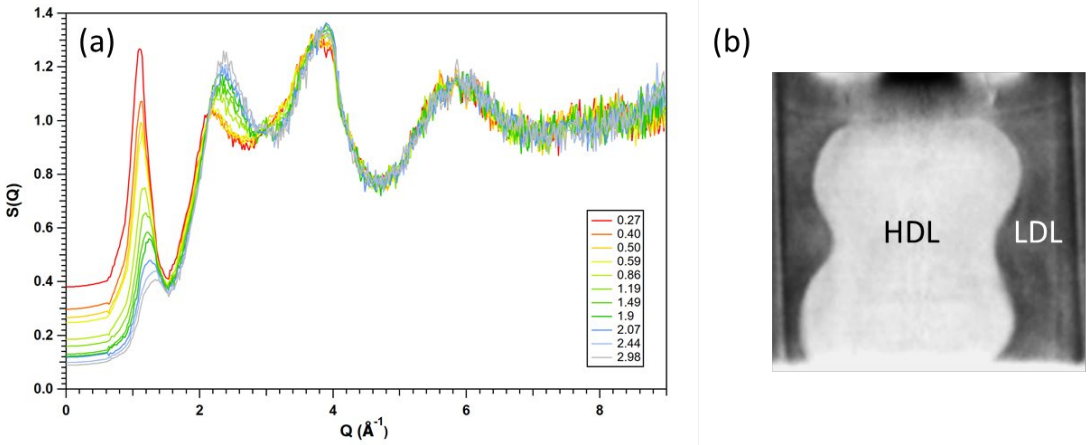

Figure 1: (a) Structure factor of liquid As_4S_4 as a function of pressure extracted from x-ray diffraction data collected at ESRF ID27. (b) Slice of a 3D tomography made at SOLEIL PSICHE, of a As-P liquid sample inside the Paris-Edinburg press, showing the coexistence of two liquids with low (LDL) and high (HDL) densities.

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