ESRF	**Experiment title:** **Study of dense liquid alkali metals**	**Experiment number**: HD 570
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Shifts: 15	**Local contact(s)**: Gaston Garbarino	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

* G. Weck, CEA/DAM Bruyères-le-Châtel, France
* P. Bouvier, INPG - MINATEC LMGP, Grenoble, France
* M. Mezouar, ESRF, Grenoble, France

Report:

The alkali group elements are considered as textbook examples of free-electron metals because of the single s electron in the valence band. However, when these metals are subjected to compression these simple systems exhibit unexpected complexity. One of the interesting point is the large number of extremely complex crystal structures at high pressure. Indeed, in the case of Na one of these structures contains more than 500 atoms in the unit cell[1]. This could be correlated with the astonishing behavior of the melting temperature pressure dependence. For Li, at pressure between 40 and 70GPa the melting line is below room temperature; and for Na this region is reduced from 116 to 120GPa just above room temperature. The complex phase diagrams of these "simple" metals suggests extraordinary liquid states at extreme conditions and has implications for other "simple" metals. Previously, a possible link between the maximum in the melting line and liquid–liquid phase transition (LLPT) was suggested[2]. Also, changes in the short-range order of molten Na at high pressure have been correlated to the corresponding solid phase transitions. We intended to explore the P-T diagram (200K<T<300K) of the alkali metal samples following isochoric paths in order to obtain quantitative structure factor, radial distribution function and density of liquid alkali metals up to 120 GPa by angular dispersive X ray diffraction (ADXRD). These unique high pressure and low temperature studies imply the use of a synchrotron source able to provide focused high brilliant and stable beam with the additionally cryogenic and multichannel collimator facilities.

Experimental method

Three membrane diamond anvil cells were equipped with diamond anvils with culet size ranging from 600μm of 75μm for this experiment. The cells were loaded with Sodium in the

glove box available in the ESRF Chemistry lab. We introduced a small grain of KBr to be able to determine the pressure[3]. The excellent conditions of the glove box (extremely low values of oxygen and water in the glove box) assured clean loaded samples for the experiment. We performed angular dispersive X-ray diffraction with a monochromatic beam of energy E = 33 keV at the ID27 beamline. The X-ray diffraction images were collected with an on-line image plate detector (MARCCD). The beam was focussed down to 4*4 μm^2.
At the beginning of the experiment an important effort was dedicated to minimize the "parasitic" diffuse scattering that reached the detector and polluted the signal from the sample. We aligned the multichannel collimator and we assured (using a cell with liquid Argon at 1GPa) that the measured total scattering signal was consistent with the published data. This procedure was established as a protocol for the next diffuse scattering experiments.

Results

In the second cell equipped with 600μm culet size diamonds and an external heater we were able to increase pressure up to 1GPa at room temperature (Na in the solid phase), then start heating at constant pressure up to 250C (Na in the liquid phase) and finally increase pressure again up to 10GPa. We followed extremely carefully the liquid-solid transition in order to determine if there is a density jump across the transition. In figure 1 we present the obtained total diffuse scattering signal of Sodium at 1GPa and 200C.
A third cell was prepared with 75μm culet diamonds, loaded with a grain of KBr as pressure marker and Rhenium gasket. It is important to mention that in this loading the hole where the sample was located has approximately 30μm. In this case, we increased pressure up to 100GPa to reach the region where the melting line has a negative slope and we increased temperature to melt the sample. The diamonds were polluted with a thin Rhenium layer (less than 100μm) that has diffraction peaks in the same Q range where the diffuse scattering from the Sodium is expected so we were not able to detected any signal from the liquid.
We also developed some independent tests on other liquid alkaline metals, noble gases and sulfur to show the feasibility of the method.

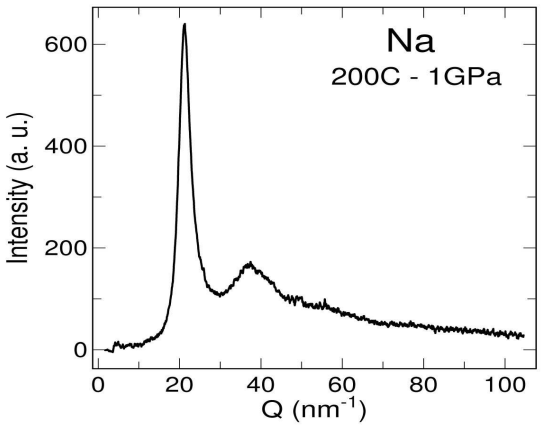

Fig. 1 Total diffuse scattering of liquid Sodium obtained at 200C and 1GPa at ID27 using the MCC during the experiment.

[1] E. Gregoryanz , L. Lundegaard, M. McMahon, C. Guillaume, R. Nelmes, M. Mezouar, *Science* **320**, 1054 (2008).
[2] S. Falconi, L.F. Lundegaard, C. Hejny, M.L. McMahon, *Phys. Rev. Lett.* **94**, 125507 (2005)
[3] A. Dewaele, A. B. Belonoshko, G. Garbarino,et al, *Phys. Rev. B* **85**, 214105