ESRF	**Experiment title:** Structure determination of superconducting modulated phases of sulphur.	**Experiment number**: **HS2982**
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Report:

The room temperatures structures of S have been studied in detail in the megabar range[1] and were considerably revised[2]. In contrast, the structural behaviour below 300 K is virtually unknown under pressure due to the difficulties associated with high-pressure diffraction studies at cryogenic conditions especially for complex structures. We have performed x-ray powder diffraction in a He-cooled cryostat to explore the phases of S at pressures between 5 and 135 GPa and low temperatures ranging from 300 K down to 15 K. Our goal was to map out the stability fields of the common S8 allotrope, the recently discovered spiral chain structure (S-III) and the incommensurately modulated phase (S-IV)[3]. However, during the course of this work we discovered a surprising transformation to an amorphous form having liquid-like properties and a broad range of kinetic stability. In situ x-ray diffraction data demonstrates that an amorphous state of sulphur is observed from 36 GPa at temperatures below 300 K to at least 100 GPa at 40 K[4].
The amorphous diffraction signal was extracted by substracting the solid crystalline state background signal taken at the nearest P-T point. The resulting spectra were then subject to intensity normalization by the form factor and scaled using the Krogh-Moe and Norman method in order to get the structure factors (Fig.1) and the radial distribution functions. Density of the amorphous state is then extracted using a modified version[5] of the method described by Eggert et al.[6] for the study of liquid water and liquid argon. The values obtained for amorphous sulphur at high pressure fall on the room temperature equation of state of sulphur S-III at ambient temperature. The transition from crystalline S-I to the amorphous phase is accompanied by major volume collapse. As such, the amorphization can be understood as a pressure-induced thermodynamic

process.

If compressed further at low temperatures, sulphur recrystallizes into high quality single crystals of the spiral chain structured phase, a newly discovered phase, or the incommensurate superconducting phase depending on pressure and temperature. This first documented case of such effects in an elemental solid could be a common scenario for simple systems at extreme conditions.

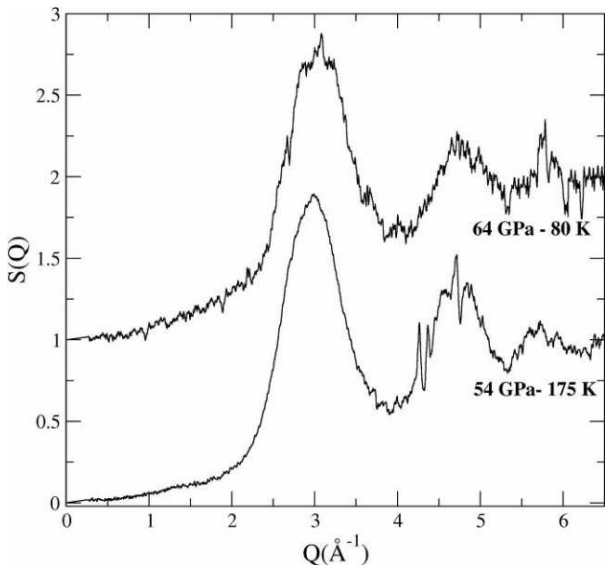

Fig. 1. Structure factors of amorphous S under pressure.

Also, using first-principle calculations, we show that a one dimensional charge-density wave (CDW) instability is responsible for the formation of the incommensurate modulation of the atomic lattice observed during this experiment in the high-pressure phase of sulfur between 83 and 153 GPa[7].

Publications:

These experiments have provided material for publications in three papers[4,5,7], two of them being submitted yet.

References:

1- Degtyareva et al., Phys. Rev. B 71, 214104 (2005)
2- Crapanzano et al., Nat. Mater. 550, 4 (2005)
3- Degtyareva et al., Nat. Mater. 4, 152 (2005)
4- Gregoryanz et al., submitted.
5- Sanloup et al., to be submitted.
6- Eggert et al., Phys. Rev. B 65, 174105 (2002)
7- Degtyareva et al., submitted.