	Experiment title: Crystalline Structure of the Metallic State of Oxygen: a Study of the zeta-Oxygen Phase Above 100 GPa by Single Crystal X-ray Diffraction	**Experiment number**: HS2757
Beamline: ID27	**Date of experiment**: from: 22 June 2005 to: 26 June 2005; from 08 October 2005 to: 11 October 2005	**Date of report**: 01/09/2008
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Report:

The main goal of the present study was to determine the crystalline structure of the metallic phase (ζ-O_2) of solid oxygen above 100 GPa. Several experiments have already been done in order to determine the structure of the metallic phase of oxygen. First, Akahama et al. [1] have performed angular dispersive X-ray diffraction on powder of pure oxygen and concluded to an isostructural phase transition at the metallization; the technique used, however, provided limited data at very high pressure due to the low scattering power of oxygen. Moreover, because of the non-hydrostatic condition, the X-ray diffraction peaks recorded were very broad at high pressure and the diffraction patterns were thus difficult to refine. We have carried out energy dispersive X-ray diffraction on oxygen single crystals in helium [2]. The experiments gave access to the orientation matrix of the crystal avoiding any erroneous indexation. The X-ray diffraction peaks from single crystals were intense and sharp even at high pressure. In those experiments, we have highlighted a displacive transition at the metallization; however, only 4 peaks were still present after the transition and we could not refine the structure of the metallic phase. Under the current project, we have performed angular dispersive X-ray diffraction on oxygen single crystals using with helium as a pressure transmitting medium. We used an image plate detector.

Experimental method

Three membrane diamond anvil cells with high X-ray aperture ($2\theta_{max}$ = +/- 37 °, φ = 360°) were loaded with oxygen-helium mixtures at ambient temperature and high pressure (1000 bar). For each sample, a single-crystal of pure oxygen was grown in the middle of the compression volume. The typical size of the sample and the single crystal was 30 μm and 20 μm in lateral dimensions. The optical properties of the oxygen epsilon phase allowed us to choose different orientations for each single crystal. Pressure was measured using the ruby pressure calibrant. 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 (MAR3450). The beam was focussed down to 10 μm in diameter.

Results

X-ray diffraction experiments were performed at ambient temperature for different pressures up to 135 GPa. Despite of the low X-ray diffraction power of oxygen, we could obtain well defined X-ray peaks. Thanks to the microfocussed beam of ID27, we were able to avoid X-ray diffraction arising from the gasket material containing the sample. Methods used to analyze diffraction images and characterize the crystalline structure of the metallic phase were identical to those which made it possible the recent finding of the crystalline structure of the ε phase, comprising clusters of molecules forming O_8 sub-units. The success of the methods used, e.g., the complete determination of the crystalline structure including atomic positions, obviously requires a very precise measurement of the intensity of the observed peaks which first implies the use of good quality single crystals. In the present case, in spite of the displacive nature of the transition observed, the crystals grown at high pressure were all damaged upon solid oxygen becoming metallic in the ζ phase. Also, even at lower pressure, and because of the experimental conditions (beam size 10 microns, single crystal lateral size 20 microns, total sample size 30 microns), only a semi-quantitative determination of diffracted intensities was possible. Nevertheless, the present experiment allowed us to show that the structure of the metallic phase of oxygen is associated with a continuous displacive structural transformation. This transformation take place in the ab plane of the monoclinic C2/m cell of the insulator phase. Although of first-order, the change of crystalline structure at the insulator to metal transition is found to be iso-symmetric. These results are confirmed by Raman measurement performed in our laboratory.

References

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