ESRF	**Experiment title:** Oxygen in the laser-heated diamond cell: High Temperature Equation of State, Melting Curve, and Phase Diagram	**Experiment number**: HS2853
Beamline: ID 27	**Date of experiment**: from: 14 Feb 2007 to: 20 Feb 2007	**Date of report**:
Shifts: 18	**Local contact(s)**: Mohamed Mezouar	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Laura Robin Benedetti*

Daniele Antonangeli*

Chantel Aracne*

Daniel Farber*

Report:

In order to improve understanding of the phase diagram of oxygen, the third most abundant element in the solar system (after hydrogen and helium), and further, in order to assess previous speculative results *(Benedetti, 2001)* suggesting the presence of a new phase of oxygen quenched from the high P-T conditions of laser heating in a diamond cell, in HS2853 we performed simultaneous x-ray diffraction and laser heating of samples of elemental oxygen in a diamond anvil cell.

Laser heating experiments always involve careful sample preparation because the technique itself requires a delicate balance between thin, well insulated samples (to promote heating and to minimize temperature gradients within the x-ray volume) and thicker samples (to increase x-ray diffraction signal). Both sides of this balance were important to this experiment because while oxygen (Z=8) is a weak x-ray scatterer, there is also a high perceived risk of chemical reaction between hot oxygen and the materials used to contain it (including the carbon of the diamond anvil). We prepared our samples using a novel sample geometry: Inside the Re sample chamber a 10-15 micron thick by 50-60 micron diameter cryogenic sample of oxygen was encased radially by a gold ring (for chemical inertness), and the oxygen-gold sample was sandwiched between two single crystals of Al_2O_3, each 15 microns thick. Samples prepared this way maintained high purity and were quite successful at stably reaching high temperatures. During single-sided laser heating, we measured similar temperatures on both sides of the sample, indicating that not only was our geometry successful from the point of view of allowing the sample to heat, but that the oxygen was also thin enough that temperature gradients within the x-ray volume are small.

The phase diagram of oxygen has previously been studied to temperatures < 900 K. *(Weck, 2002),* and at 16 GPa, the melting temperature of oxygen is only 638 K *(Young, 1987)*. Therefore, to access the hot solid phase by laser heating (minimum measurable temperature ~1300 K) we need to perform laser heating experiments at very high pressures. In this experimental run, we laser heated oxygen at several pressures between 30 and 60 GPa. Our initial interpretation of the experimental data is that *in situ* laser heating might not be useful for pressures below 45 GPa : the oxygen sample rapidly absorbs the laser and goes directly to the fluid phase -- so it is not necessarily possible to put a lower bound on the melting temperature at these pressures.

However, above this pressure, we were able to heat the solid oxygen, and we have succeeded in measuring the melting temperature at at least one pressure. (Two more heating cycles are still being analyzed.) During laser heating at 55 GPa, temperature increased significantly as soon as two lasers were used to heat the sample. (Figure 1) This strong heating coincided with loss of diffraction of oxygen (Figure 2) and also with changes in wavelength-dependent emissivity. Therefore, we interpret the melting temperature to be between the two temperature regimes: T_{melt} = 1850 K, consistent with extrapolation of the

previously measured melting curve (Figure 3). We expect further data analysis to reduce the error bars on the temperature measurements and also the melting temperature.

In summary, we have demonstrated a sample preparation technique for oxygen samples that results in high-purity, stably-heating samples, and we have used such samples to measure the melting temperature of oxygen at 55 GPa. In future work, we will use this technique to prepare higher pressure samples in order to define the melting curve throughout the epsilon-oxygen stability field (P<100 GPa). Further, when the melting points are higher, we will be able to also get thermal equation of state information from the laser-heated solid data. Finally, while we never clearly saw the diffuse scattering that indicates positively the presence of fluid oxygen, because our samples heat well and stably, we hope to observe it in future runs if we collect diffraction for longer periods of time.

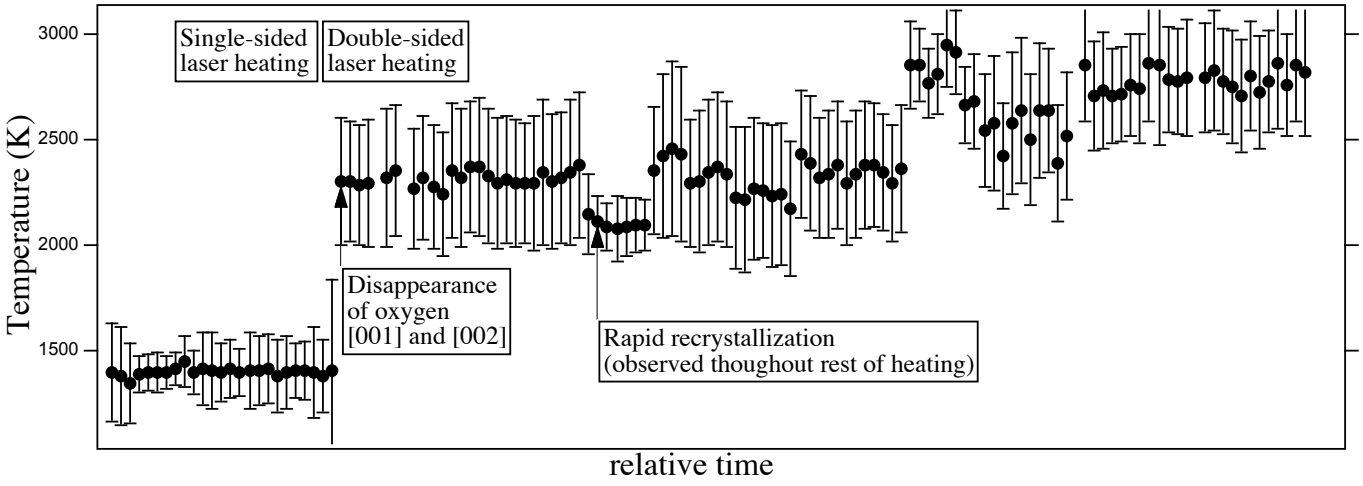

Figure 1. Temperature measurement throughout the heating cycle at 55 GPa.

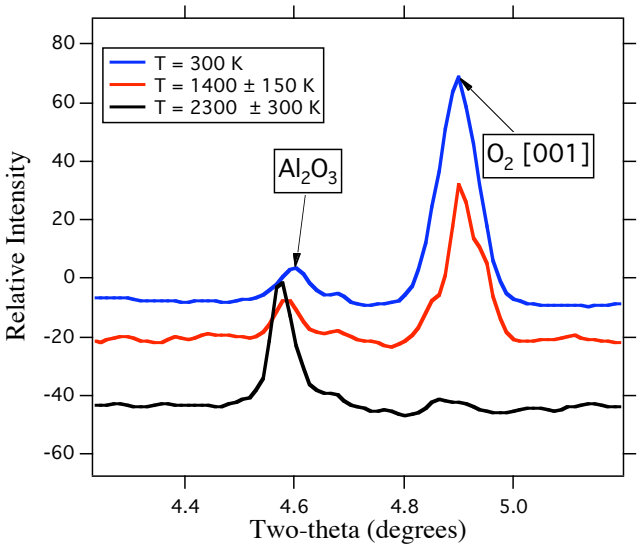

Figure 2. X-ray diffraction of oxygen in the region of the [001] reflection at three temperatures: 300 K (blue, top), 1400 K red, middle), and 2300 K (black, bottom). The [001] reflection of oxygen disappears between 1400 and 2300 K.

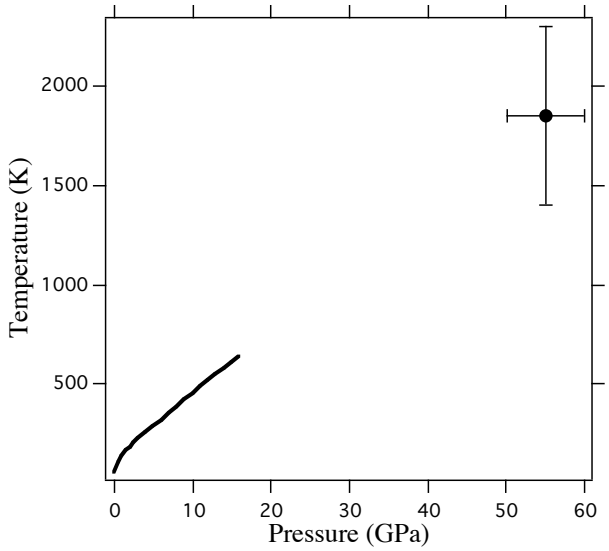

Figure 3. Melting curve of oxygen. This measurement (solid circle) is in reasonable agreement with the lower pressure data (solid line) of Young. (*1987*) We expect further data analysis to reduce our errors.

References

Benedetti, L. R., *Ph. D. Thesis,* University of California, Berkeley (2001).

Weck, G., ESRF report HS-1842 (2002).

Young, D. A., *et al.* , "Diatomic melting curves to very high pressure," *Phys. Rev. B.,* **35**(10): 5353 (1987).