ESRF

Experiment title:

Synthesis of the first xenon nitride: Determination of the structure and stoichiometry

Experiment number:

HC-3086

Beamline:

ID27B

Date of experiment:

from: March 16th 2017 to: March 19th 2017 (5 shifts)

from: May 20th 2017 to: May 23th 2017 (4 shifts)

Date of report:

30/08/2017

Shifts:

9

Local contact(s):

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Objectives

Both N_2 and Xe are inert under ambient conditions and hardly interact with other elements. Under pressure, the properties of both elements drastically change and their chemistry becomes richer. The XeF_2 solid [1] as well as the Xe_3O_2 and Xe_2O_5 [2] compounds have been formed at high pressure and have shown to be composed of covalent bonds. Recently, we have produced the first xenon nitride with indications of N-N and Xe-N single covalent bonds [3], similar to theoretically predicted XeN_6 solid [4]. The aim of this proposal was to characterize this newly-synthesized Xe-N compound by X-ray diffraction in order to determine its structure and stoechiometry. Pressure and temperatures in the range 150 GPa – 200 GPa and 1500 – 3000 K, respectively, were generated in a laser-heated diamond anvil cell so as to enter the stability domain of the Xe-N solid. During sample decompression, X-ray diffraction measurements at regular pressure steps were attempted so as to obtain the compound’s equation of state as well as its stability domain.

Experimental method

Three membrane diamond anvil cells were equipped with anvils of 70 (2x) and 50 µm in culet size. All cells were loaded with a Xe-N_2 mixture, with xenon concentrations of 20, 15 and 5 mol%. A gold particle was loaded along with the sample and its equation of state was used to determine the pressure inside the experimental chamber. While chemical insulation of the sample was previously shown not to be necessary, one of the samples (with 20 mol% Xe) had a thin Al_2O_3 layer deposited on both diamond anvils and acted as a thermal insulator. Double-sided YAG laser-heating was performed with the on-line setup of ID27, allowing black-body radiation temperature measurements. Angular dispersive powder X-ray diffraction was performed, using a monochromatic beam of energy E = 33 keV, between each pressure step as well as before and after laser-heating. The X-ray diffraction images were collected with an on-line image plate detector (MARCCD). The beam was focussed down to ~ 3x3 μm^2.

Previously to the beamtime, all samples were dynamically compressed to 70 GPa. This rapid compression prevented the formation of the $Xe(N_2)_2$ van der Waals compound, previously established to be extremely stable [5] and hinder the synthesis of the Xe-N compound. In doing so, amorphous Xe and N_2 (see Fig. 1 a)) were observed before laser-heating.

Results

The results obtained are summarized in Table 1. The first sample (20 mol% Xe) was compressed to 170 GPa, pressure at which it was laser-heated to 2300 K for a few minutes. As shown in Fig. 1, laser-heating of the amorphous $Xe+N_2$ mixture resulted in many new diffraction lines. While some diffraction lines match $Xe(N_2)_2$ or the high pressure and high temperature phase of Al_2O_3, others fit with the previously-observed Xe-N covalently-bonded solid. Decompression of the sample was then attempted, but after a 5 GPa pressure decrease one of the diamonds gave away, allowing the sample to escape the experimental chamber.

Table 1: Summary of the three experimental runs performed on the $Xe-N_2$ system at high pressure and high temperature.

Run	Concentration (mol% Xe)	Insulating layer	Laser-heating pressure (GPa)	Laser-heating temperature (K)	Chemical reaction
1	20	Al_2O_3	170	2300	Yes
2	5	None	131	2500	Yes
3	15	None	205	-	-

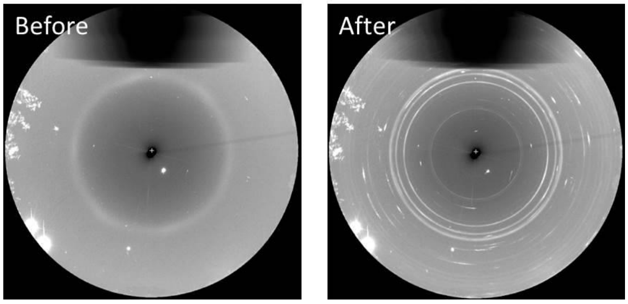

Figure 1: X-ray diffraction image plates of a $Xe-N_2$ sample (20 mol% Xe) at 170 GPa. (Left) As seen, before laser-heating the sample appears amorphous and does not show diffraction rings characteristic of the $Xe(N_2)_2$ van der Waals compound. This was observed from all dynamically-compressed samples. The diffraction spots belong either to the diamond anvils or the thin Al_2O_3 layer. (Right) Image plate after laser-heating the sample at 2300 K at 170 GPa. Diffraction rings appear and some match those previously observed and assigned to a new Xe-N compound. Other diffraction rings belong to $Xe(N_2)_2$ and the high pressure and high temperature phase of Al_2O_3.

The second sample, of greater nitrogen concentration (5 mol% Xe) and without an insulating layer on the diamond anvils, was laser-heated to 2500 K at 148 GPa. As shown in Fig. 2, diffraction rings similar to those observed from the first sample appeared. Again, some of them correspond to the diffraction lines of $Xe(N_2)_2$ or the Xe-N compound, but others match cg-N. The synthesis of a pure form of nitrogen, cg-N, not found in the previous run, indicates the Xe-N compound to be of nitrogen concentration between 80 and 95 mol% N_2. Once again, a diamond anvil broke when attempting sample decompression.

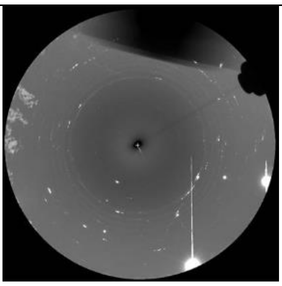

Figure 2: X-ray diffraction image plate of a $Xe-N_2$ sample (5 mol% Xe) at 148 GPa after laser-heating at 2500 K. Slightly textured, the diffraction rings match those expected from $Xe(N_2)_2$, cg-N as well as the Xe-N solid.

The last sample was prepared with a 15 mol% Xe concentration. To favor the synthesis of the Xe-N compound instead of $Xe(N_2)_2$, pressure was increased to 205 GPa before laser-heating. However, one of the anvils ruptured during heating, releasing the pressure and therefore the sample. A deeper analysis of the data obtained during the three experimentals runs will be necessary to solve the structure of the Xe-N compound.

References

[1] M. Kim, M. Debessai and C.-S. Yoo, Nat. Chem. **2**, 784 (2010).
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[4] F. Peng *et al*. PRB **92**, 094104 (2015).
[5] D. Laniel *et al*. PRB **94**, 174109 (2016).