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S. Klotz, Th. Strässle, B. Lebert, M. d'Astuto, Th. Hansen. High pressure neutron diffraction to beyond 20 GPa and below 1.8 K using Paris-Edinburgh load frames. High Pressure Research, Taylor & Francis, 2016, 36 (1), pp.73-78. 10.1080/08957959.2015.1136624 . hal-01282686

HAL Id: hal-01282686
https://hal.archives-ouvertes.fr/hal-01282686

Submitted on 18 Apr 2016

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High pressure neutron diffraction to beyond 20 GPa and below 1.8 K using Paris-Edinburgh load frames

S. Klotz[1], Th. Strässle[2], B. Lebert[1], M. d'Astuto[1], Th. Hansen[3]

[1]IMPMC, CNRS UMR 7590, Université P&M Curie, 4 Place Jussieu, F-75252 Paris, France

[2]Paul-Scherrer-Institut, CH-5232 Villigen PSI, Switzerland

[3]Institut Laue-Langevin, Avenue Jules Horowitz, F-38043 Grenoble France

We describe a method for collecting neutron diffraction patterns simultaneously at high pressure (> 22 GPa) and low temperature (< 1.8 K). The system uses ~ 5-10 mm^3 samples compressed by double-toroidal sintered diamond anvils, with the required forces generated by a Paris-Edinburgh press of 30 kg mass. Technical details are given and diffraction data of ε-iron at 22.6 GPa and 1.79 K are presented.

A large part of neutron diffraction deals with physical phenomena which appear only at very low temperatures close to 0 K, such as many magnetic ordering and quantum critical phenomena. There is a growing interest to investigate these phenomena with high pressure since the interatomic distances, and thus the interaction parameters controlling these phenomena, can be tuned in a continuous and controlled fashion. The simultaneous generation of high pressure and very low temperatures is a well-known problem in neutron scattering, in particular for pressures in the multi-GPa range where the pressure cells are rather massive and therefore need a more complex cryogenic solution.

Here we discuss a method which allows neutron diffraction to at least 22 GPa and 1.8 K using samples of typically 5-10 mm^3. The technique uses a VX5 Paris-Edinburgh high pressure cell of 130 tonnes capacity [1], double-toroidal anvils [2], and cryogenic equipment which was already briefly presented [3]. Figure 1 shows schematically the setup. The pressure cell is attached to, and cooled by, a two-stage closed-cycle refrigerator (CCR) with a base temperature of 3.6 K. A key feature of this cryogenic setup is that during operation the cell is in contact with He-exchange gas inside a sealed Al-container (item 6). This ensures efficient heat exchange and small temperature gradients compared to more conventional CCR solutions where the sample or pressure cell is in vacuum and only cooled/heated by thermal contact to the cold head. A second Al container (item 7) is attached to the first stage of the CCR and acts as a heat shield towards the outer vacuum container (item 8). Temperature changes with the 30 kg mass of the VX5 cell are rather slow using the CCR only, typically 0.1 K/min. A practical modus operandi is therefore to pre-cool the cell with liquid nitrogen injected into the inner Al container through a vent inlet (item 4). This method allows reaching 80 K within approximately 2 hours, including the removal of the remaining liquid and purging with He-gas.

In initial experiments at the Swiss neutron source SINQ at the Paul Scherrer Institute (PSI) we realised that this cryogenic equipment is perfectly adapted to reach much lower temperatures. For this purpose the cell is cooled to a base temperature of typically 4-6 K using the CCR. The inner Al-container is then filled with liquid helium. Figure 1 (right panel) gives the volume of He stored in the Al-container as a function of filling height with respect to the bottom of the inner Al-container (item 6). It is seen that the container can accept approximately 3 litres of helium up to the sample level, and up to 8.5 litres in total. Pumping on the fluid through one of the two vents (diameter 12 mm) leads to a rapid decrease in temperature: the lambda point (2.17 K) is reached within 15 minutes, while 1.8 K is reached within 30 minutes. The He consumption to 2 K is approximately 30-40% and mainly consumed to cool the fluid He itself, not the cell, which has a much lower heat capacity at this temperature. With helium filled up to ~ 20 cm from the bottom of the Al-container, the He-level drops to below the sample/beam height when temperatures below 2 K are reached, i.e. at the point where neutron data can be collected. In fact, even when the sample is below the liquid He level, diffraction data can be collected, although attenuated by approximately 40%. From this moment onward, the He-consumption is very low as a consequence of the CCR still running and hence screening the heat flow from above. With the remaining 3 litres of He, we have kept temperatures below 2 K for 7 hours, with 80% of this time below 1.8 K (base temp. 1.79 K). This period is sufficient for most diffraction experiments, even on small samples.

We have used this setup to investigate the ε-phase of iron which is stable beyond ~ 15 GPa. For this purpose, double-toroidal anvils [2] were used with their profile dimensions reduced by a factor 0.8 compared to previous designs [4], see inset Fig. 2. To generate the required forces we used a VX5 Paris-Edinburgh cell with a capacity of 130 tonnes and a mass of 30 kg. See Ref. [1] for details concerning this type of large volume cell and its application to neutron scattering. Null scattering TiZr gaskets were used with an inner set of encapsulating hemispheres. The solid sample was machined from a rod of pure iron (Goodfellow Ref. 203-947-27, 99.99+% purity) into a roughly 9 mm^3 sphere. No pressure medium was used since all pressure transmitting fluids are solid anyway at our target pressure and temperature conditions. However, the spherical shape of the sample chamber seems to ensure quasi-hydrostatic conditions, judged by the measured onset pressure of the α-ε transition (14.5 GPa) and the pressure where the α-phase disappears (19.5 GPa), in comparison with previous investigations on the effect of non-hydrostaticity on this transition [5]. Pressure values were obtained directly from the measured (refined) unit cell volume and the known equations-of-state of α- [6] and ε-iron [7,8]. Initial tests at 300 K were carried out in 2003 at the PEARL station of the U.K. ISIS facility using D_2O ice VII as a sample. These indicated that using this anvil profile (inset Fig. 2), pressures in excess of 25 GPa can be generated with a force of less than 130 tonnes which is within reach of a VX5 press.

The data reported here were collected at the high-intensity diffractometer D20 [10] at the Institut Laue-Langevin, Grenoble, France, using a wavelength of 1.30 Å, produced by a copper (200) monochromator at a take-off angle of 42°. This instrumental configuration gives maximal neutron flux at reasonable resolution up to 2-theta ~ 60 degree and degraded resolution above. Figure 3 shows a diffraction pattern obtained at 1.79 K and 22.6 GPa, after 10 minutes of beam time. Apart from a scale factor and background, the Rietveld fit to the pattern (line through the data) includes refinements of lattice parameters, isotropic thermal displacement factors as well as preferred orientation. Unavoidably, the strongest reflections are due to the anvil material, polycrystalline diamond. These can be readily included into the fits and pose no serious problem for structural

investigations. The initial room temperature compression to 100 tonne gave patterns of pure ε-Fe (plus diamond) with refined unit cell parameters of a=2.44232(20) Å and c=3.92918(39) Å, i.e. V=20.297(3) $Å^3$, hence a pressure of 21.3 GPa according to the Vinet-Rydberg equation-of-state of Ref. [7] (V_0=11.214 $Å^3$/atom, B_0=163.4 GPa, B_0'=5.38). After cooling at constant load, the refinements of the pattern at 1.79 K shown in Fig. 3 give a=2.43873(22) Å and c= 3.91908(40) Å, i.e. V=20.186(3) $Å^3$. This indicates a pressure of 22.6 GPa using unpublished x-ray synchrotron data of ε-Fe obtained at 15 K [8] and the same type of equation-of-state with V_0=11.207 $Å^3$/atom, B_0=163.6 GPa and B_0'=5.33 [8]. The 13 K difference with our measurements is completely negligible because the thermal expansion of any solid at such temperatures is virtually zero. After decompression at ~ 200 K the anvils were recovered undamaged. A complete analysis of these results including the search for the potential presence of magnetism in ε-Fe will be given separately [11].

It should be admitted that iron's large scattering length and simple structure produce high intensity diffraction patterns. Nevertheless, the method described here might be applicable for a large number of materials where relatively low resolution is required at high scattering angles, e.g. for examples studies with a focus on magnetism. With the ever increasing performance of neutron focussing techniques the method described here might become more widespread in the future.

Acknowledgments. We are grateful for beamtime [12] and other resources provided by ILL for these experiments, as well as the ISIS Facility for beamtime in initial test measurements in 2003. Blair Lebert acknowledges a PhD grant from the French excellence initiative under the MATISSE program and support from the LLB-SOLEIL PhD grant program. We thank X. Tonon, Claude Payre and Alain Daramsy (ILL) as well G. Hamel (IMPMC) and J. White (PSI) for their help during the experiments.

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Figure captions

Figure 1 : Left: High pressure cell in its cryostat. Right: Helium volume as a function of filling level. (1) : Pressure cell, (2) He-capillary, (3) CCR cold head, (4) vent, (5) Indium-seal, (6) inner Al container, (7) 30 K Al-heat shield, (8) outer vacuum container. Filling levels are measured from the bottom of the inner Al-container.

Figure 2 : Pressure-load curve for a 6 mm^3 sample of iron and ice VII, at 300 K. Pressure values were obtained using equations of state from Refs. [6,7] (iron) and Ref. [9] (ice VII). Lines are guides to the eye. The inset shows the cross section of the anvil profile used in the experiments with dimensions given in mm.

Figure 3 : Diffraction pattern of ε-iron at 22.6 GPa and 1.79 K. The line is a Rietveld fit to the data (dots), the difference curve is given below. Upper tick marks indicate ε-Fe peak positions, lower tick marks those of diamond from the anvils. The pattern represents raw data, i.e. no background was subtracted. The accumulation time is 10 minutes.

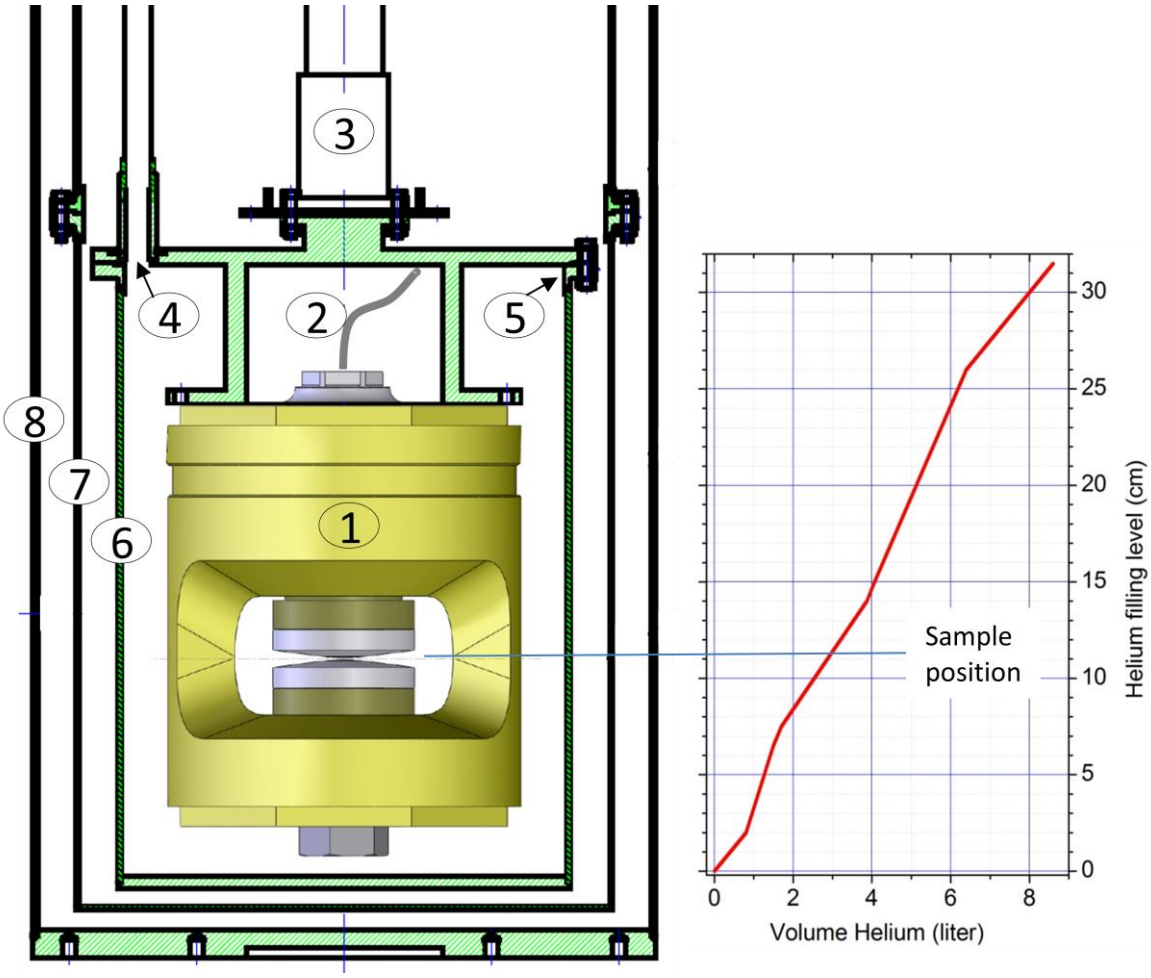
3
4
2
5
8
7
6
1
Sample
position
Helium filling level (cm)
0
5
10
15
20
25
30
0
2
4
6
8
Volume Helium (liter)

Figure 1

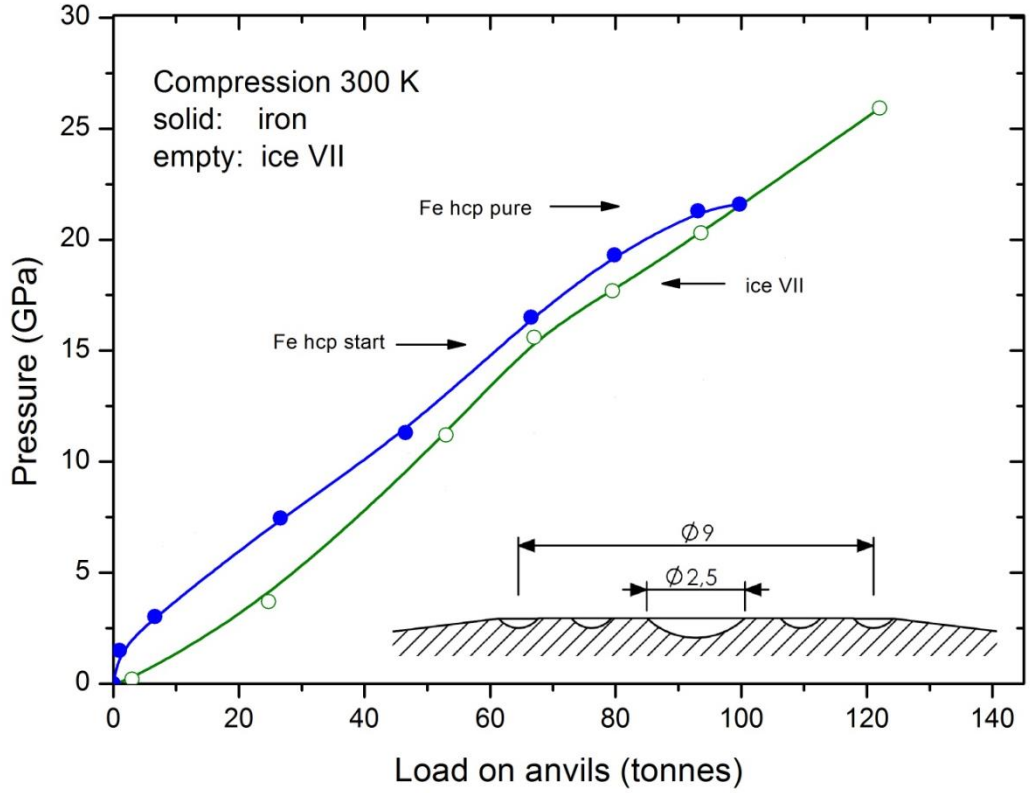
Compression 300 K
solid: iron
empty: ice VII
Fe hcp pure
ice VII
Fe hcp start
Ø9
Ø2,5
Pressure (GPa)
Load on anvils (tonnes)
0
5
10
15
20
25
30
0
20
40
60
80
100
120
140

Figure 2

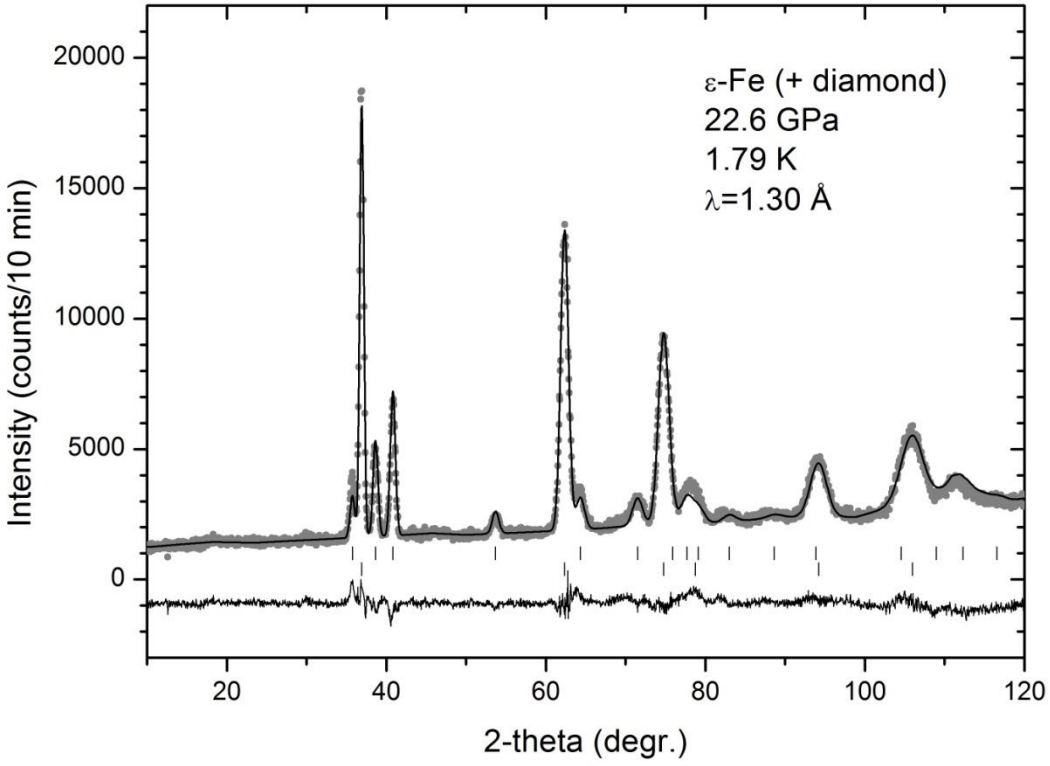
ε-Fe (+ diamond)
22.6 GPa
1.79 K
λ=1.30 Å
Intensity (counts/10 min)
2-theta (degr.)
20000
15000
10000
5000
0
20
40
60
80
100
120

Figure 3