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Experiment title:
Low temperature equation of state of epsilon-iron

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Names and affiliations of applicants (* indicates experimentalists):

Dewaele Agnès*

Garbarino Gaston*

Report:

The electronic and magnetic properties of high pressure phases of iron are still debated. Alpha-Fe is ferromagnetic up to 12 GPa at ambient temperature. Experimental data suggest that epsilon-Fe is paramagnetic at 300K, while DFT-GGA theoretical calculations predict a lower energy for different antiferromagnetic orderings at 0K [1]. The agreement between the calculated Equations of State (EoS) for these antiferromagnetic phases and the measured EoS at 300K is relatively poor [1]. Recently, the use of a theoretical method which aims at a better description of electronic correlation effects, DMFT, yielded a good agreement between calculated and measured EoS for paramagnetic epsilon-Fe at 300 K [2]. This is a strong hint that epsilon-Fe is paramagnetic at 300K, as suggested by experiments. However, the magnetic state of epsilon-Fe at low temperature is unknown.
Experimental and theoretical evidences for an electronic topological transition (ETT) in epsilon-Fe around 40 GPa have been published recently [3]. It is expected that this transition has an effect on thermal expansion and lattice parameters c/a ratio evolution at low temperature.
The aim of this proposal was to measure a reference equation of state for epsilon-Fe at low temperature, to provide a test for future calculations of the magnetic state of this phase in the ground state, and to see a possible signature of the predicted ETT on the thermodynamic behaviour of epsilon-Fe at low temperature.

We have carried out three experiment at 15K in a helium cryostat. The pressure was measured using a SrB_4O_7:Sm2+ luminescence gauge, and cross-checked with a gold luminescence gauge. The volume was measured using angular-dispersive X-ray diffraction. The conditions of the experiments are summarized in **Table 1**.

Name	culet diameter (µm)	Pressure range (GPa)	T (K)
FeBT_1	400	2-35	15
FeBT_2	150	29-94	15
FeBT_3	150	25-64	15

Table 1: Conditions of the three experimental runs.

The measurements of *V* vs. *P* for alpha-Fe and epsilon-Fe and *c/a* vs. P for epsilon-Fe at 15K are plotted in **figure 1**. The low pressure volume measured for alpha-Fe is compatible with previous data at 300K [1] and thermal expansion measured at room pressure [4].
The 15K EoS for epsilon-Fe is very close to the 300 K EoS measured previously [1]. A fit of the 15 K *P-V* points for epsilon-Fe with a Rydberg-Vinet EoS yields the folowing parameters: V_0=11.207 A^3/at, K_0=163.6

GPa, K'$_0$=5.33 (fixed), to be compared with V_0=11.177 A^3/at, K_0=168.4 GPa, K'$_0$=5.33 at 300K [1]. It can be noted that the EoS at 15K diverges (as the 300K EoS) from the calculated EoS for the AFM-II magnetic order, which suggests that the magnetism at 15K does not differ from the magnetism at 300K (which is likely to be paramagnetic). Surprisingly, the 15K volume around 20 GPa is not clearly lower than the 300K volume, which could be related to pressure gradients in the pressure chamber, which leads to small pressure errors, or elastic strains after phase transformation. The 15K bulk modulus, 163.6 GPa, is lower than expected if a simple Mie-Güneisen-Debye model is used to model the thermodynamic behaviour of epsilon-iron (173 GPa, if the value of 168.4 GPa at 300K is assumed). This is the expected trend if the predicted ETT occurs [3]. However, such a small (5%) difference can be due to metrology issues (in particular, pressure gradients in the pressure chamber).

At 300K, the *c/a* ratio decreases with increasing pressure [1,3]. Two trends have been distinghished in Ref. [3]: below 40 GPa, a fast decrease, above 40 GPa, a slower decrease, with a small disconituity between these two trends (see **figure 1**, inset, red points). This has been interpreted as a signature of an ETT around 40 GPa. At 15 K, the *c/a* ratio decreases with *P*, with an almost constant slope between 20 and 100 GPa. No discontinuity can be clearly seen in the data. However, the progressive divergence of the *c/a* at 15K from the *c/a* at 300K, which is larger than the data scatter, could be a signature of the ETT. Our data suggest that the thermal expansion along the *c* axis becomes higher than the thermal expansion along the *a* axis above 40 GPa. This should be compared with expected behaviour of thermal expansion below the Debye temperature taking into account the ETT.

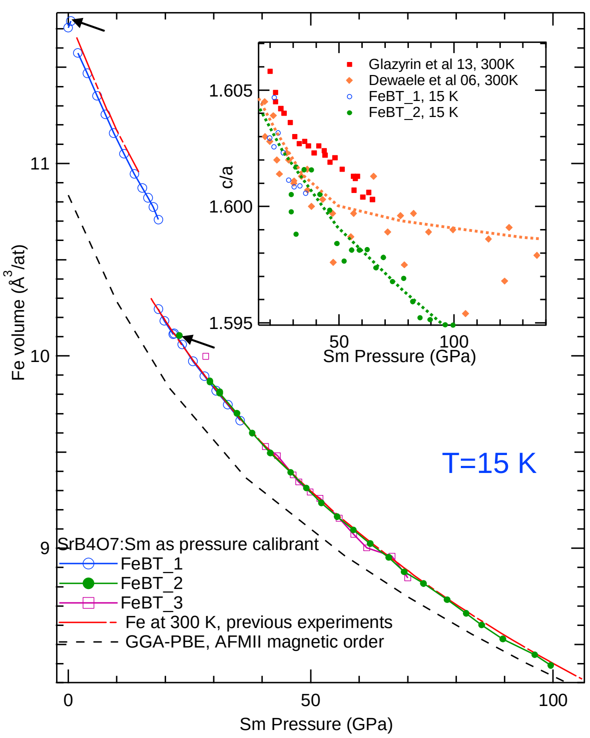

Figure 1: Iron volume, measured at 15 K, in three experimental runs. The pressure has been measured using a $SrB_4O_7:Sm^{2+}$ pressure gauge. It is compared with a fit of 300K data (in red, ref. [1]). The arrows point to the two ambient temperature measurements. Inset: plot of the *c/a* lattice paremeters ratio in epsilon-Fe, vs. pressure. The data for the run FEBT_3 have not been represented, due to their large scatter due to data acquisition difficulties. The dashed lines are guide to the eyes representing the trends for 300K (in orange) and 15 K (in green) measurements.

References:

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