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► To cite this version:

F. Xu, G. Morard, N. Guignot, A. Rivoldini, Geeth Manthilake, et al.. Thermal expansion of liquid Fe-S alloy at high pressure. *Earth and Planetary Science Letters*, 2021, 563, pp.116884. 10.1016/j.epsl.2021.116884 . hal-03184365

HAL Id: hal-03184365

<https://uca.hal.science/hal-03184365>

Submitted on 31 Mar 2021

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1 **Thermal expansion of liquid Fe-S alloy at high pressure**

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32 **Highlights**

- 33 - Density of liquid Fe-S alloys has been measured under high pressure as a function of
34 temperature by *in situ* X-ray diffraction in multi-anvil press.
- 35 - Thermal expansion of liquid Fe-S alloys has been determined up to 7 GPa and 2200
36 K.
- 37 - Top-down crystallization is the most likely scenario for Fe-FeS cores of planetesimals
38 and small planets.

39

40

41 **Abstract**

42 Local structure and density of liquid Fe-S alloys at high pressure have been
43 determined *in situ* by combined angle and energy dispersive X-ray diffraction experiments
44 in a multi-anvil apparatus, covering a large temperature and compositional range. Precise
45 density measurements collected for increasing temperature allowed us to directly
46 derive the thermal expansion coefficients for liquid Fe-S alloys as a function of
47 composition. In turn, thermal expansion has been used to refine thermodynamic models
48 and to address the crystallization regime of telluric planetary cores by comparing the
49 adiabatic temperature gradient and the slope of the liquidus in the Fe-FeS system.

50 For Fe-S cores of asteroids and small planetesimals, top-down solidification is the
51 dominant scenario as the compositional domain for which the slope of the liquidus is
52 greater than the adiabatic gradient is limited to a narrow portion on the Fe-rich side.
53 However, bottom-up growth of the inner core is expected for S-poor cases, with this
54 compositional domain expanding to more S-rich compositions with increasing pressure
55 (size of the planetary body). In particular, bottom-up crystallization cannot be excluded for
56 the Moon and Ganymede.

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61 1. Introduction

62 Many recent studies highlight a rich diversity in the core structure of terrestrial bodies,
63 largely depending on the core crystallization regime (e.g. Breuer et al., 2015; Dumberry and
64 Rivoldini, 2015; Hauck et al., 2006; Rivoldini et al., 2011; Rückriemen et al., 2018, 2015;
65 Williams, 2009). This, in turn, directly depends on pressure conditions and light element
66 content, which relates to accreting material, planetary differentiation process and planet's size.
67 From an observational point of view, indications on the core structure and dynamics can be
68 derived from geodetic data (static and dynamic gravity field, and rotation) and, when present,
69 from the magnetic field. The presence of an internally generated magnetic field is relatively
70 common in the terrestrial bodies of our solar system. Aside from Earth, active global
71 magnetic fields of internal origin have been detected on Mercury and Ganymede (Kivelson et
72 al., 1997; Ness, 1979). A strong, though now extinct, magnetic field was also present on Mars,
73 as suggested by crustal magnetic field maps (e.g. Acuña et al., 1999, 2001; Connerney et al.,
74 2001) and on the Moon, as evidenced from the remnant magnetism of the lunar crust (e.g.
75 Weiss and Tikoo, 2014). Furthermore, magnetized meteorites indicate that planetesimals and
76 numerous planetary bodies may have had their own self-generated, long-lived fields (e.g.
77 Weiss et al., 2008; Shah et al., 2017; Bryson et al., 2019a). A dynamo operating in the fluid
78 metallic core is the most likely mechanism for generating a planetary magnetic field, and
79 compositional convection driven by core solidification is one of the main power sources for a
80 long standing dynamo (Stevenson, 2003). The strength and lifetime of a magnetic field is thus
81 directly tied to the crystallization process of the core, which can proceed very differently in
82 various planets, depending on composition and size (e.g. Breuer et al., 2015; Rückriemen et
83 al., 2018).

84 As a first order guidance, neglecting complexities arising because of nucleation barriers
85 (e.g. Davies et al., 2019; Huguet et al., 2018), we here assume that solid inner core starts to
86 form when the temperature drops below the melting curve of the core-forming material (the
87 liquidus). In a convecting medium the temperature profile is almost isentropic and the core
88 temperature is generally assumed to follow an adiabat. The relative slopes (dT/dP) of the
89 adiabat and of the liquidus thus determine the style of core solidification (e.g. Breuer et al.,
90 2015; Williams, 2009). If the adiabat is shallower than the liquidus, crystallization occurs at
91 depth, as for the Earth, and the inner core grows bottom-up. On the contrary, if the adiabat is
92 steeper than the liquidus, top-down solidification is expected, in which shallow crystallization
93 occurs and solid material sinks or floats, depending upon density contrast with the liquid,
94 leading to possible compositional stratification (e.g. Dumberry and Rivoldini, 2015; Hauck et
95 al., 2006; Rückriemen et al., 2018, 2015). Although both scenarios don't rule out the
96 development of a dynamo, how and when a magnetic field can be generated in the course of
97 top-down crystallization remains debated (e.g. Breuer et al., 2015).

98 Sulfur (S) is classically considered to be the major light element alloyed to iron (Fe) in
99 the core of small planetary bodies (e.g. Antonangeli et al., 2015; Breuer et al., 2007; Morard
100 et al., 2018; Terasaki et al., 2019; Rückriemen et al., 2015; Weber et al., 2011). The actual S
101 content depends on primordial composition of the planetary forming material and the
102 partitioning of sulfur between silicate and metallic phases, which in turn depends on

103 temperature and pressure conditions, as well as on the oxidation state, during core formation.
 104 For instance, while both Mercury and Mars are thought to have significant S in their bulk
 105 composition, the greater distance from the Sun and the more oxidizing formation conditions
 106 support more significant enrichment of S in the core of Mars than in that of Mercury
 107 (Dumberry and Rivoldini, 2015; Hauck et al., 2006; Rivoldini et al., 2011). Ultimately
 108 differences in the P, T, fO₂ conditions of core formation result in a large variation in the S
 109 abundance in the liquid cores of planets and moons. For instance, S content in the Moon's
 110 core is classically limited to less than 8 wt.% (e.g. Antonangeli et al., 2015; Laneuville et al.,
 111 2014; Rai and Van Westrenen, 2014; Steenstra et al., 2017), while in the case of the Jovian
 112 moons Ganymede and Callisto, the core might have a S concentration greater than the
 113 eutectic concentration (e.g. Scott et al., 2002). As a direct consequence of this composition
 114 diversity, different crystallization scenarios can arise. Indeed S content significantly affects
 115 the difference in the adiabat vs. liquidus relations and density contrast between the
 116 crystallizing solid and the liquid phases (e.g. Breuer et al., 2015; Williams, 2009).

117 Thermal expansion is a central parameter controlling the slope of the adiabat (Stacey,
 118 2005; Williams, 2009), which can be written as

$$119 \quad dT/dP = \alpha(P)T / \rho(P)C_P \quad (1)$$

120 where $\alpha(P)$ is the pressure-dependent thermal expansion, T the temperature at which the
 121 adiabatic gradient is calculated, $\rho(P)$ the pressure-dependent density, and C_P the heat
 122 capacity at constant pressure.
 123

124 According to its definition, thermal expansion can be experimentally derived from the
 125 variation of the density with the temperature (at constant pressure)

$$126 \quad \alpha(P,T) = -1/ \rho(P,T) (d \rho(P)/dT) \quad (2)$$

127
 128 Due to the experimental difficulties in density measurement, neither thermal expansion
 129 measurements over a wide compositional range (0-50 at% S) nor under high pressure have
 130 been performed. Thermal expansion of liquids in the Fe-FeS system has been studied
 131 exclusively at ambient pressure and only for end-member liquids, Fe and FeS (e.g. Assael et
 132 al., 2006; Kaiura and Toguri, 1979; Nagamori, 1969). Results on FeS (Kaiura and Toguri,
 133 1979) are limited to few points covering a very limited T range (~1500-1650 K). Even in the
 134 case of liquid Fe, in spite of the large number of studies, proposed values for thermal
 135 expansion show large discrepancies (e.g.; $11.0 \times 10^{-5} \text{ K}^{-1}$, Hixson et al., 1990; $8.2 \times 10^{-5} \text{ K}^{-1}$,
 136 Nasch and Steinemann, 1995; $13.2 \times 10^{-5} \text{ K}^{-1}$, Assael et al., 2006). Density determinations of
 137 liquid Fe-S alloys under high pressure includes ex-situ sink-float (Balog et al., 2003; Nishida
 138 et al., 2008), X-ray absorption (Chen et al., 2014; Nishida et al., 2011; Sanloup et al., 2000;
 139 Terasaki et al., 2019) and X-ray diffraction (Morard et al., 2018) methods. However, none of
 140 these methods provided sufficient temperature-dependent density data to allow precise
 141 determination of thermal expansion, due to combined difficulties in density measurements
 142 and temperature control at high pressure.
 143

144 As highlighted by Williams (2009), extrapolation of thermal expansion of liquid Fe and
 145 FeS to pressures, temperatures and compositions directly relevant for planetary cores comes
 146

147 with significant uncertainties that limit the reliability of the assessment of the relative slopes
148 of the adiabats and liquidus curves (which also is not so well known). The determination of
149 thermal expansion of liquid Fe-S alloys under the high-pressure conditions existing in
150 planetary cores is thus of fundamental importance for refining thermodynamic models of
151 planetary cores (Morard et al., 2018; Rivoldini et al., 2011; Terasaki et al., 2019) and to
152 constrain adiabatic heat flux in cores of telluric bodies (Silber et al., 2018).

153 In this study, we provide direct determinations of the thermal expansion of liquid Fe-S
154 alloys in the 4 to 9 GPa range, pressures directly relevant for the core of the Moon and
155 Ganymede, using a combined angular and energy dispersive X-ray diffraction (CAESAR)
156 technique (Wang et al., 2004). X-ray diffuse scattering from a liquid sample was analyzed to
157 extract local structure information and also used to determine liquid density at high pressure
158 (Eggert et al., 2002; Morard et al., 2014). To date, this method has been applied in
159 Paris-Edinburgh (PE) cell and diamond anvil cell (DAC) for structural and density studies of
160 liquids under high pressure (e.g. Morard et al., 2018, 2017, 2014; Sanloup et al., 2013;
161 Yamada et al., 2011, 2007). Here we present the nontrivial extension of these measurements
162 to the multi-anvil press. Noticeably, extension of this methodology to the multi-anvil press
163 enables investigations over larger pressure range than achievable in PE experiments and,
164 more importantly, the ability to vary the temperature of a confined liquid with the precision
165 needed for the determination of thermal expansion, refinements not possible in the
166 above-mentioned early PE and DAC studies. Measured thermal expansions are used together
167 with thermodynamic modeling to assess the pressure and compositional dependence of the
168 adiabatic gradients in the liquid Fe-FeS system in the 0 to 10 GPa range and, by comparison
169 with the slope of the liquidus, to discuss the crystallization regime in the cores of small
170 planetary bodies, encompassing conditions of the Moon and Ganymede.

171

172 **2. Methods**

173 *2.1 In-situ CAESAR measurement under high pressure in a multi-anvil press*

174 High-pressure experiments were carried out *in situ* in the DIA-type multi-anvil apparatus
175 installed on the beamline PSICHE of SOLEIL, France. Starting materials with final
176 compositions of 5, 10, 15, 20, 25 and 36.4 wt.% S, corresponding to 8.4, 16.3, 23.6, 30.4,
177 36.8 and 50 at.%, respectively, were obtained by a dry homogenization of mixtures of Fe and
178 FeS powders in a mortar (without lubricants such as ethanol or acetone). The prepared
179 mixtures were stored in a portable vacuum box to minimize the moisture absorption from air,
180 and kept there until loading right before the experiment. For the high P-T experiments
181 samples were placed in sapphire capsule capped with BN. The pressure assembly consisted of
182 26 mm WC cubes with 4 mm truncation and and pressure was measured *in situ* using the
183 known equation of state of the MgO marker (Tange et al., 2009). Schematic view of the cell
184 assembly is shown in Fig. S1. Trimmed octahedral pressure medium made from B+15 wt.%
185 MgO (B85) was employed to minimize the absorption and diffraction of the sample
186 environment (Xie et al., 2020). B85 gaskets were also used along the X-ray path instead of
187 pyrophyllite gaskets. To remove the water absorbed during fabrication prior to the

188 experiment, MgO and ZrO₂ parts were baked at 1273 K for 1 hour; BN, graphite and B85
189 were kept at 393 K in a vacuum oven for more than 12 hours. Such a tailored cell assembly
190 was proven to be fundamental to the collection of high-quality CEASAR data. High
191 temperature was generated with graphite heaters and monitored with a W₉₇Re₃-W₇₅Re₂₅
192 thermocouple whose junction was set at the position symmetrical to the sample capsule with
193 respect to the center of furnace. Status of cell assembly during the experiments was
194 monitored by X-ray radiography.

195 Energy dispersive X-ray diffraction (EDD) measurements were carried out using a
196 polychromatic X-ray (20-100 keV) focused to 25 μm vertically (FWHM) and collimated to
197 50 μm horizontally, much smaller than the sample dimension (0.8 mm diameter, 0.5 mm
198 thick, Fig. S1). A Canberra SSD Ge detector associated with a XIA multi-channel analyzer
199 with 2048 energy bins was used to acquire EDD covering energies up to 102.4 keV. The
200 energies were calibrated using characteristic fluorescence X-ray lines of Mo, Sn, Ba, Sm and
201 Au. The 2θ angle was calibrated from 5 to 25° using a 7 μm thick Au foil with a precision of
202 ±0.003 °.

203 For all runs, the specimen was first brought to the target pressure and then heated to high
204 temperature while collecting energy dispersive diffraction patterns at a fixed angle (8°). The
205 melting of sample was identified from the disappearance of sharp diffraction peaks during
206 temperature increase, and confirmed by the absence of any sharp peak during CAESAR
207 acquisition. Once complete melting was achieved, CAESAR spectra were collected in order
208 to obtain the structural and density information of the liquid. CAESAR collections were then
209 repeated for increasing temperature, with 100-200 K steps. For each CAESAR scan, EDD
210 were collected every 0.2° over a 2θ angle ranging from 2.5 to 24.1°, allowing acquisition of
211 high quality raw data over a wide Q -range, up to 15 Å⁻¹ ($Q = 4 \pi E \sin\theta / 12.398$, where E is
212 the energy of the X-rays in keV). All measured EDD are combined to form a
213 two-dimensional array of intensities, $\text{Int}(E, 2\theta)$, with each Int value corresponding to a given
214 E and 2θ index. Fig. 1a shows representative raw data collected on Fe-S in this study.
215 CAESAR scan with good counting statistics were acquired with collection times of 5 sec for
216 EDD between 2.5 and 10°, 10 sec for EDD between 10 and 20° and 20 sec for EDD between
217 20 and 24.1° (about 25 minutes in total). The horizontal gaps of the collimating slits were
218 opened gradually during the angular scan from low angle to high angle to control the
219 effective sample volume from which the diffracted X-rays was detected for discriminating
220 sample signal from background scattering caused by surrounding materials. The raw
221 CAESAR data were normalized by collection time and effective sample volume for further
222 treatment. To improve counting statics, raw data were binned utilizing energy data following
223 the method described in Wang et al. (2004) (Fig. 1b).

224 In view of the vertical focusing of the beam, 2D radiographies for alignment and sample
225 observation purposes were recorded by scanning the press in front of the beam (scanning
226 radiography) (Fig. 1c, d).

227

228 2.2 Analysis of the recovered samples

229 After experiments, the recovered cells were mounted in epoxy resin and polished along
230 the plane parallel to the cylindrical axis of sample. The samples were polished sequentially

231 using diamond abrasive disks with 120, 30 μm and diamond paste with 1 μm grain size to
232 obtain well-polished surface. Microstructure of recovered samples was analyzed by a field
233 emission scanning electron microscope (SEM-FEG) (Zeiss Ultra55) at IMPMC, Sorbonne
234 Université, France. The Fe and S contents and the concentrations of potential contaminants
235 (B, N, C, Al, O, W, Re) were determined using electron probe microanalyses (EPMA) at
236 Centre Camparis, Sorbonne Université, France using a Cameca SX-FIVE wavelength
237 dispersive spectrometer (WDS) operating at 15 kV and 300 nA. Our recovered samples show
238 fine dendritic textures of Fe and Fe-S; we therefore used a defocused beam of $\sim 30 \mu\text{m}$ to
239 average the compositions of the quenched liquid. Bulk chemical compositions were obtained
240 by averaging 5-10 measurements. Table 1 reports the obtained average values and
241 corresponding standard deviations.

242

243 2.3 Analysis of diffuse scattering signal

244 The scattering intensity curve, $I(Q)$, is constructed by merging the normalized EDD and
245 removing the background (taking advantage of data collected at multiple 2θ angles over
246 overlapping Q range (Fig. 2a)). The structure factor, $S(Q)$, and the pair distribution function,
247 $g(r)$, are calculated for the fixed Q range of 1-10 \AA^{-1} for all cases. The structure factor, $S(Q)$,
248 is first obtained after subtraction of incoherent scattering ($I_{\text{inc}}(Q)$). The distribution function,
249 $F(r)$, and pair distribution function, $g(r)$, are obtained by the Fourier transformation of
250 structure factor. Following the method detailed in (Morard et al., 2014, 2013), determination
251 of density is based on the assumption that, due to the increasingly strong repulsive
252 component in the interatomic potential, no atoms are located closer than minimal distance,
253 r_{min} . Thus $F(r) = -4\pi r\rho$ for $r < r_{\text{min}}$. Density is, hence, extracted following the minimization of
254 the oscillation in the short distance of the radial distribution function $g(r)$ (through a merit χ^2)
255 (Eggert et al., 2002). Fig. 2b illustrates atomic densities calculated for different values of the
256 minimal distance r_{min} at different temperatures. The value of χ^2 exhibits a well-defined
257 minimum in most cases (r_0 in Fig. 2b), which gives the atomic density ρ_0 for which the
258 relation $F(r) = -4\pi r\rho$ is best-satisfied (Fig. 2c). The position of r_{min} usually corresponds to the
259 base of the first coordination sphere in $g(r)$. Noteworthy for the purpose of the present study,
260 while absolute values of density depend upon r_{min} , density variation with temperature ($d\rho/dT$)
261 is effectively independent of r_{min} (Fig. 2b).

262 Various aspects of the data treatment enter into the assessment of the error bar on the
263 absolute density value, including the selected Q range, the choice of the minimum distance of
264 the first coordination sphere (r_0), along with physical phenomena neglected here such as the
265 self-absorption from the sample (Morard et al., 2018, 2014). In the present data set, the
266 estimated error is $\pm 3 \text{ atoms/nm}^3$ for the atomic density, corresponding to $\pm 220\text{-}270 \text{ kg/m}^3$ for
267 the mass density of the Fe-S liquids (depending on composition).

268

269 2.4 Analysis at constant pressure

270 For analysis at constant pressure, experimental densities have been rescaled to 7 GPa
271 following a Murnaghan formalism:

$$P - P_{ref} = \frac{K_{ref}}{K'} \left[\left(\frac{\rho}{\rho_{ref}} \right)^{K'} - 1 \right] \quad (3)$$

where P is 7 GPa and ρ is the recalculated density at 7 GPa, P_{ref} and ρ_{ref} are our direct obtained experimental data at actual temperatures (Table 1), K_{ref} is the isothermal bulk modulus at P_{ref} and 1900 K, and K' is its pressure derivative. When not otherwise specified, we used the parameterization discussed in (Morard et al., 2018). Both K_{ref} and K' are functions of atomic S content (XS) in the liquid. The isothermal bulk modulus at ambient pressure and 1900 K of Fe-S alloys was obtained following (Chen et al., 2014) according to the relation $K_0 = K_{Fe}^{1-XS} * K_S^{XS}$, assuming an exponential dependence of the bulk modulus of liquid Fe-S alloys with S content, and with $K_{Fe}=76$ GPa and $K_S=1.6$ GPa. The pressure derivative of the bulk modulus (K') was obtained as $K' = K'_{Fe} + XS \cdot 3$, with $K'_{Fe} = 6.5$ (Morard et al., 2018). In this analysis the temperature dependence of bulk modulus was neglected (temperature range of interest < 500 K).

2.5 Thermodynamic models

To model the thermodynamic properties of liquid Fe-S (for brevity l-Fe-S) alloys we use the non-ideal solution model introduced in Morard et al. (2018) and Terasaki et al. (2019). The excess mixing volume is assumed to be pressure dependent and parameterized by an asymmetric Margules formulation. The two end-member phases of the binary solution model are l-Fe and l-FeS. The volume of the non-ideal solution is:

$$V(\chi_{FeS}, P, T) = (1 - \chi_{FeS})V_{Fe}(P, T) + \chi_{FeS} V_{FeS}(P, T) + \chi_{FeS}(1 - \chi_{FeS}) V_{ex}(\chi_{FeS}, P) \quad (4)$$

where χ_{FeS} is the mol fraction of FeS, V_{Fe} and V_{FeS} are the molar volumes of pure l-Fe and l-FeS, and V_{ex} is the pressure and composition dependent excess mixing volume given by:

$$V_{ex}(\chi_{FeS}, P) = (\chi_{FeS} W_{Fe} + (1 - \chi_{FeS}) W_{FeS}) v(P), \quad (5)$$

where $v(P)$ is the pressure dependent contribution to the Margules parameters. Differently from Morard et al. (2018) and Terasaki et al. (2019) $v(P)$ is not parameterized by empirical expressions, but with a pseudospinodal equation of state (EOS) (Baonza et al., 1995):

$$v(P) = \exp \left[\frac{1}{K'} \left(1 - \sqrt{1 + 2 \frac{K'}{K_0} P} \right) \right] \quad (6)$$

The parameters W_{Fe} , W_{FeS} , K_0 , and K' and the equation of state parameters of l-FeS are estimated from the data of this study as well as from density measurements of Morard et al. (2018) and acoustic velocities of Nasch et al. (1994) and Nishida et al. (2016).

Several recently published EOS of l-Fe (e.g. Komabayashi, 2014; Dorogokupets et al.,

311 2017; Wagle and Steinle-Neumann, 2019) are in relatively good agreement with respect to
 312 the prediction of density and acoustic velocities along isentropes at high pressure and
 313 temperature (Fig. S2a). Nonetheless, quantities derived from the associated thermodynamic
 314 potentials, such as the thermal expansivity and heat capacity that determine the adiabatic
 315 gradient in a convecting liquid core of a planet, show relatively large deviations (Fig. S2b, c).
 316 Therefore, the EOS of l-Fe affects not only the estimation of the temperature in the core but,
 317 together with dependent thermodynamic quantities, also determines the locations at which the
 318 core temperature drops below the liquidus. For this reason we assess here how the EOS of
 319 l-Fe of Komabayashi (2014), Dorogokupets et al. (2017) and Wagle and Steinle-Neumann
 320 (2019) affect the thermodynamic properties predicted by our l-Fe-S model. In the following
 321 we will refer to Model K, Model D and Model W to denote the thermodynamic models
 322 constructed on the basis of our datasets in the Fe-S system at high pressure and high
 323 temperature, which respectively make use of Komabayashi (2014), Dorogokupets et al. (2017)
 324 and Wagle and Steinle-Neumann (2019) for the EOS of l-Fe.

325 For the EOS of l-FeS we use a Vinet equation and the Anderson-Grüneisen formulation
 326 to describe the pressure dependence of the thermal expansivity (α) (e.g. Komabayashi, 2014).
 327 As we could not estimate α and the value of the Grüneisen parameter at reference
 328 conditions ($P_{\text{ref}} = 0.1\text{MPa}$ and $T_{\text{ref}} = 1650\text{K}$) from our experimental data, we assume
 329 $\alpha = 11.8 \times 10^{-5} \text{ 1/K}$ from Kaiura and Toguri (1979) and the isobaric heat capacity
 330 $C_p = 62.5 \text{ J/K/mol}$ (Chase, 1998). The Grüneisen parameter at reference conditions can then
 331 be computed from the estimated EOS parameters with the thermodynamic relation
 332

$$333 \quad \gamma = \frac{K_T V \alpha}{C_p - K_T V T \alpha^2}, \quad (7)$$

334 where K_T is the isothermal bulk modulus and V is the molar volume. The EOS parameters
 335 and Margules coefficients estimated from the experimental data for the three different models,
 336 using different EOS of l-Fe, are given in Table S1.

337 The liquidus at sulfur concentration lower than that of the eutectic is parameterized
 338 following Buono and Walker (2011) (Eq. 7-9). This procedure neglects the small amount of
 339 S that can be dissolved in solid Fe and requires only the knowledge of the Gibbs energy of
 340 l-Fe and stable solid Fe phases together with the Margules parameters required to describe the
 341 non-ideal behavior of the Fe-S system. To obtain a thermodynamically consistent description
 342 we compute the liquidus according to the l-Fe EOS' of Komabayashi 2014 and Dorogokupets
 343 et al. 2017 together with the EOS' of the relevant solid phase provided by those authors
 344 (Model K and Model D).

345 Using those EOS' the Margules parameters are then estimated from experimental
 346 melting data. Here we make use of the experimental data at 1bar (e.g. Waldner and Pelton,
 347 2005), 3 GPa (Brett and Bell, 1969), 6 GPa (Buono and Walker, 2011) and 10 GPa (Chen et
 348 al., 2008). With the knowledge of the Margules parameters and with the EOS' of l-Fe and
 349 solid Fe, the liquidus can then be computed at the required pressures. The estimated values of
 350 the Margules parameters are given in Table S2.

352

353 3. Results

354 The experimental data obtained here are summarized in Table 1 and described more in
355 detail in the following subsections.

356

357 3.1 Recovered sample and chemical compositions

358 A representative cross-section of the recovered cell is shown in Fig. 3a. All of the
359 samples (except FeS end member) exhibited homogeneous dendritic textures (Fig. 3c-g),
360 characteristic of completely molten samples. The compositions of the recovered samples
361 determined by EPMA marginally deviate from the starting compositions of the initial powder
362 mixtures, showing a systematic increase by 0.7-1.8 at.% in the S content (Table 1). Potential
363 causes include Fe exsolution from the liquid as an oxide, or simply a deviation from the
364 expected 1:1 molar ratio in the FeS starting material. Possible chemical contamination by B,
365 N, C, Al, O, W and Re was carefully checked. No Al contamination from sapphire capsule
366 was ever observed for any of the samples. Some of the runs on samples with high melting
367 temperature (low S content) showed B and/or N contamination from the BN cap (Fig. S1).
368 For runs for which we experienced a thermocouple failure, often in relation to sample leaking
369 as observed by *in-situ* radiography, W and Re were often detected in the analysis of the
370 recovered experiments. In this paper we only present analysis of data from samples that show
371 no contamination.

372

373 3.2 Structure of liquid Fe-S alloys

374 Fig. 4 shows examples of the structure factors, $S(Q)$, and derived pair distribution
375 functions, $g(r)$, of liquid Fe-S alloys at high pressure and high temperature. The shape of $g(r)$
376 is characterized by two peaks located at approximately 2.35-2.55 Å and 4.7 Å corresponding
377 to the distance of the first and second coordination spheres (CS), respectively. The sharpness
378 of these features decrease with increasing S content, and a third local maximum around 6.5 Å
379 is visible only for alloys with 31.7 at.% or less (the $g(r)$ become less structured with
380 increasing S content). The first peak position of $g(r)$, r_1 , was observed to monotonically
381 decrease with increasing S content (Table 1, Fig. S3), which agrees quite well with previous
382 report on liquid Fe-S (Shibazaki and Kono, 2018). The progressive reduction of r_1 with S
383 content can be rationalized by considering that the partial pair distribution functions
384 systematically show Fe-S bonds (~ 2.2 Å) shorter than Fe-Fe bonds (~ 2.5 Å) (Morard et al.,
385 2018). As mentioned, with increasing S content, the first and second peaks of $S(Q)$ become
386 broader, and oscillation of the $g(r)$ becomes less pronounced (Fig. 4a, b). The second
387 oscillation in the $g(r)$ seems to vanish at 51.2 at.% S.

388 For a fixed S concentration, the $S(Q)$ and $g(r)$ do not show major changes with
389 temperature (Fig. 4c, d). On a qualitative ground, this indicates that the temperature does not
390 significantly modify the structure of liquid Fe-S alloys over the investigated range, other than
391 for the increasing bond length.

392

393 3.3 Density and thermal expansion of liquid Fe-S alloy

394 All densities determined in this study are reported in [Table 1](#). In selected cases we
395 repeated the measurements in more than one heating cycle (e.g. MA40), showing a
396 remarkable consistency. Also, using the nominal starting composition, the densities derived
397 from data obtained in an experiment with leakage of the molten sample (MA58), yielded
398 values compatible with those from more reliable data (MA66) obtained in the absence of
399 leakage. The systematic consistency of our results proves the validity of our data collection
400 strategy and data treatment (e.g. range of Q , value of r_{\min}). As such, pressure and temperature
401 derivatives of density, as its variation with S content, are highly reliable (see also discussion
402 in section 2.3).

403 Previous density data for liquid Fe-S alloys under high pressure includes results obtained
404 by ex-situ sink-float (Balog et al., 2003; Nishida et al., 2008), in situ X-ray absorption (Chen
405 et al., 2014; Nishida et al., 2011; Sanloup et al., 2000; Terasaki et al., 2019) and X-ray
406 diffraction (Morard et al., 2018) methods. The reported values display large discrepancies,
407 however, even when the same method was used. Overall, the derived thermodynamic model
408 from this study is compatible with densities determined here, although the agreement is less
409 good for alloys with moderate S content (18 at.% or below) ([Fig. S4](#)). Of the parameters
410 entering into the density modeling, the weight of thermal expansion is small, in particular
411 when compared to that of compressibility. The most direct way to assess thermal expansion is
412 thus considering the density evolution with increasing temperatures as from equation (2).
413 Density determination for liquid Fe-S alloys with different S content at 7 GPa over an
414 extended temperature range in this study is summarized and compared with thermodynamic
415 Model W ([Fig. 5](#)).

416 For each composition, density decreases approximately linearly with increasing
417 temperature (see section 2.4, equation 2). Aside from the already mentioned difference in the
418 absolute density values noticeable for the alloys with low-S content, the thermodynamic
419 model, is in good agreement with the experimentally observations for all the liquid alloys.
420 Within the scatter of the data, the temperature derivative of the density doesn't show a clear
421 compositional dependence ([Fig. S5](#)). As such, we cannot discriminate between results from
422 thermodynamic Model K, Model D and Model W (see section 2.5), which equally well
423 account for the measurements. Similarly, the experimentally derived thermal expansion
424 shows values scattered in the range $6\text{-}13 \times 10^{-5} \text{ K}^{-1}$ ([Fig. 6](#)), in overall agreement with the
425 results of the three thermodynamic models, but higher than previous models (Morard et al.,
426 2018; Terasaki et al., 2019), in particular for S-rich samples.

427 To a large extent, the variability in our thermodynamic models reflects the spread in the
428 values of thermal expansion of liquid Fe at ambient pressure (e.g. Williams, 2019). The
429 density of model W has the strongest dependence on temperature for liquid Fe at ambient
430 pressure ([Fig. S6](#)), with a thermal expansivity ($14.1 \times 10^{-5} \text{ K}^{-1}$ at 1900 K) in agreement with
431 Assael et al., (2006), while the thermal expansivity of Model K and Model D are smaller
432 (respectively 9.0 and $9.3 \times 10^{-5} \text{ K}^{-1}$ at 1900 K) and in good agreement with the values of
433 Hixon (1990).

434

435 4. Discussion

436 4.1 Slope of adiabat and core solidification

437 As already mentioned, the dynamic of crystallization in liquid planetary cores can be
438 evaluated by comparing the adiabatic temperature gradient (slope of the isentrope) and the
439 slope of the liquidus, which, in turn, depend upon pressure and chemical composition of the
440 studied cores.

441 The adiabatic gradient in a liquid core is calculated according to the relation (1) for the
442 three thermodynamic models, with densities benchmarked against measurements. Specific to
443 liquid FeS, we use $C_p=62.5$ J/K/mol from NIST at 1bar (Chase, 1998). Assuming a different
444 value of 40 J/K/mol (Kaiura and Toguri, 1979) does not significantly change the resulting
445 densities, slightly worsening the agreement with respect to experimental results, and making
446 the isentropes moderately steeper.

447 The slopes of the adiabats obtained at selected pressures as a function of sulfur content
448 are illustrated in Fig. 7 and compared with the slope of the liquidus (see section 2.5). It should
449 be noted that the S concentrations considered here are always below the eutectic, where
450 denser almost pure solid Fe is the phase crystallizing from the Fe-S liquid.

451 Observation of our different models highlights the dispersion depending on the chosen
452 equation of state for liquid Fe. Nonetheless, on a qualitative ground, we note that the
453 compositional domain for which the slope of the liquidus is larger than the adiabatic gradient
454 is limited to a narrow portion on the Fe-rich side for all the pressure considered here (0-10
455 GPa).

456 On one hand, due to decreasing thermal expansion and increasing density, the slope of
457 the adiabat decreases with increasing pressure. At the conditions of Moon's (5.2 GPa) and
458 Ganymede's (10 GPa) center, the adiabat only moderately depends on S content, as a
459 consequence of the density reduction that overcomes the effect on C_p (Fig. S2). On the other
460 hand, the slope of the liquidus shows a significant decrease with increasing S content. The net
461 effect is that, for Fe-S cores of small to middle size bodies (0 to 10 GPa) top-down
462 solidification is the most likely scenario for wide range of S contents. Only for S-poor cases
463 bottom-up inner grow is possible, like in the case of the Earth.

464 However, uncertainties on the relevant thermodynamic quantities resulting from the
465 liquid equation of state of Fe and thermoelastic properties of l-Fe-S alloys do not allow
466 derivation of quantitative conclusions on the crystallization regime of planetary cores as a
467 function of composition. For instance, assuming Model W, crystallization will always occurs
468 top-down because the adiabatic gradient is always steeper than the liquidus over the pressure
469 range considered here (Fig. 7c). Conversely, assuming the Model K or Model D, adiabatic
470 gradients and the slope of the liquidus cross each other at S concentrations that increase with
471 increasing pressure (Fig. 7a-b).

472 Paleomagnetic studies of chondritic and small-body achondritic meteorites have revealed
473 a large diversity of magnetic field records (e.g. Weiss et al., 2020). Convection in the cores of
474 differentiated, or at least partially differentiated asteroids, planetesimals, or meteoritic parent
475 bodies in general, may have generated dynamo magnetic fields responsible for the
476 magnetization of the overlying silicate rocks (Gattacceca et al., 2016; Weiss et al., 2008).

477 While heating from short-living radioactive isotopes is generally considered to be responsible
478 for the early dynamo, core solidification is expected to produce compositionally-driven
479 dynamo activity, at a later stage with the timing dependent on the S concentration of the core
480 and the radius of the body (Neufeld et al., 2019). At low pressures within an asteroid,
481 isentropes are steeper than liquidus, irrespective of the S content for both models (b) and (c),
482 while the adiabatic gradient is below the slope of the liquidus for alloys up to 4 at.% S
483 according to model (a) (Fig. 7). Top-down crystallization thus remains the most likely
484 scenario, but bottom-up cannot be firmly excluded for very low S content. Generation of a
485 magnetic field during top-down core crystallization could therefore be relatively common in
486 differentiated planetesimals in the early age of the solar system.

487 Lunar core formation models based on metal/silicate partitioning of siderophile elements
488 support a Moon core containing up to 10 at.% S (Rai and Van Westrenen, 2014; Stenstra et
489 al., 2017). Assuming a fully molten core in the binary Fe-S system, geodetic constraints argue
490 for a larger amount of S, between 16 and 40 at.%, depending on the core radius (Morard et al.,
491 2018). The presence of a solid inner core, made of pure Fe, brings estimates down below 10
492 at.% (Antonangeli et al., 2015). At about 5 GPa, the pressure at the center of the Moon, the
493 adiabatic gradient and slope of the liquidus might cross at about 3 or about 9 at.% S,
494 depending on the EOS of l-Fe (Fig. 7). Accordingly, if the Moon does not have a solid inner
495 core today, it will have most likely crystallized top down. On the contrary, if the existence of
496 a solid inner core, as suggested by some seismological studies (Weber et al., 2011), is
497 confirmed, its crystallization regime remains uncertain. That an initial bottom-up scenario,
498 may have evolved into a top-down regime due to the progressive enrichment in S of the
499 liquid portion of the core upon crystallization of pure Fe, as hypothesized to explain the early,
500 now extinct Moon magnetic field (e.g. Laneuville et al., 2014), remains an appealing
501 possibility.

502 Compositional convection is considered a fundamental element to explain Ganymede's
503 present-day dynamo (e.g. Rückriemen et al., 2018). In turn, compositional convection is
504 strongly linked to core differentiation and solidification. Recently, significant efforts have
505 been dedicated to model dynamics and magnetic field generation under the notion that
506 crystallization occurs at the top of Ganymede's core (Breuer et al., 2015; Rückriemen et al.,
507 2018, 2015). Our results suggest this is not necessarily the case, and that crystallization might
508 proceed bottom-up for S content below about 10 at%. Indeed, the solution space for
509 bottom-up crystallization might increase up to 7-11 at.% S, again depending on assumed
510 liquid Fe equation of state, at 10 GPa, central pressure of Ganymede's core (Fig. 7). As both
511 bottom-up and top-down crystallization can power a core dynamo and generate a magnetic
512 field, although via different mechanisms, independent constraints on S abundance in
513 Ganymede's core are needed to assess its thermo-chemical history.

514

515 **5. Conclusions**

516 We performed a structural investigation of liquid Fe-S alloys at high pressure over a
517 large temperature range and compositional domain. Derived densities have been used to
518 refine thermodynamic models of the thermo-elastic properties of the liquid Fe-FeS system

519 and, in particular, to put constraints on modeled thermal expansion at conditions directly
520 relevant for the core of small planetary bodies.

521 Comparison of the adiabatic gradient with the slope of the liquidus, is used to discuss the
522 crystallization regime of the core of planetary bodies in the range 0-10 GPa as a function of
523 sulfur concentration, with specific emphasis to the case of asteroids and small planetesimals,
524 the Moon, and Ganymede. Our results show that the compositional domain for which the
525 slope of the liquidus is larger than the adiabatic gradient is limited to a narrow portion on the
526 Fe-rich side for all the pressures considered here, implying that top-down crystallization is
527 likely a widespread phenomenon. However, bottom-up crystallization is still possible for
528 S-poor cases, and the compositional domain for which an inner core would grow bottom-up
529 increases with increasing pressure. On the basis of our experimental results and
530 thermodynamic modeling, a bottom-up scenario cannot be excluded to occur at the pressures
531 of the core of the Moon and, all the more so, of Ganymede. Improved constraints on the
532 liquid Fe EoS are needed to better discriminate possible scenarios.

533

534 **Acknowledgments**

535 The authors wish to thank Nicolas Dumesnil, Charlotte Georget and Philippe Rosier for
536 their help with machining of the cell assembly parts. We also thank Tomoo Katsura for the
537 access to the MDX machining equipment. We acknowledge Clemens Prescher, Silvia
538 Boccato and Bin Zhao for their assistance in some of the experiments. We thank Imène
539 Estève for her help with sample analysis by SEM, and Michel Fialin and Nicolas Rividi for
540 their help during microprobe analysis. Femtosecond laser micromachining at the Institut de
541 Minéralogie de Physique des Matériaux et de Cosmochimie (IMPMC), Paris, has been
542 developed and realized by the “Cellule Project” with the financial support of ANR
543 2010-JCJC-604-01. The Scanning Electron Microscope (SEM) facility at IMPMC is
544 supported by Région Ile de France grant SESAME 2006 N°I-07-593/R, INSU-CNRS,
545 Institute de Physique (INP)–CNRS, University Pierre et Marie Curie–Paris 6, and by the
546 French National Research Agency (ANR) grant ANR-07-BLAN-0124-01. This work was
547 supported by the European Research Council (ERC) under the European Union’s Horizon
548 2020 research and innovation Programme (grant number 724690). The work of AR was
549 financially supported by the Belgian PRODEX program managed by the European Space
550 Agency in collaboration with the Belgian Federal Science Policy Office. The authors
551 acknowledge synchrotron SOLEIL for the provision of beamtime under proposals 20170506,
552 20171307, 20181797 and 20190551.

553

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750 Table 1 Chemical composition of quenched samples, pressure-temperature conditions, and
 751 measured densities and position of the first sharp peak r_1 . The density scaled to a constant
 752 pressure of 7 GPa is also shown.

Run	S content (at.%)	Pressure (GPa)	Temperature (K)	Density (kg/m ³)	Density corrected to 7 GPa (kg/m ³)	r_1 (Å)
MA84	51.2±0.4	6.5	1700	4486	4521	2.374
		6.8	1800	4427	4440	2.375
		6.8	1900	4332	4345	2.376
		6.5	2000	4286	4319	2.379
		5.7	2100	4222	4312	2.378
MA40	38.3±0.5	6.5	1280	5619	5659	2.440
		6.4	1400	5526	5578	2.442
		6.4	1500	5511	5562	2.442
		6.7	1320	5619	5642	2.446
		6.6	1310	5619	5654	2.446
		6.9	1500	5573	5577	2.440
		6.0	1600	5464	5545	2.446
MA44	31.7±0.9	6.1	1790	5348	5420	2.439
		7.1	1415	5944	5938	2.450
		6.9	1500	5880	5889	2.447
		6.8	1610	5800	5817	2.451
		7.0	1710	5720	5721	2.454
MA47	25.1±0.3	6.9	1800	5656	5666	2.454
		7.5	1600	6211	6171	2.485
		7.5	1705	6160	6119	2.485
		7.8	1790	6137	6077	2.489
		7.9	1900	6084	6018	2.486
		8.4	2050	6059	5956	2.484
MA82	18.1±0.4	8.3	2020	6043	5951	2.486
		5.6	1500	6308	6416	2.502
		5.3	1600	6291	6423	2.512
		5.4	1700	6188	6310	2.515
MA66	9.1±0.5	5.2	1800	6171	6309	2.520
		6.8	1850	6746	6759	2.541
		7.1	1950	6711	6704	2.553
		7.4	2050	6647	6621	2.555
		7.7	2155	6569	6524	2.554
MA58	8.4*	8.1	2250	6520	6451	2.554
		5.5	1825	6717	6880	2.537
		5.4	1960	6599	6766	2.541
		4.7	2075	6487	6703	2.554

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754 § The uncertainty of ± 3 atoms/nm³ estimated for the used protocol in data analysis ([Morard](#))

755 [et al., 2013](#)), reflects into an error of 220, 230, 240, 250, 260 and 270 kg/m³ for 51.2, 38.3,
756 31.7, 25.1, 18.1 and 9.1 at.% S content, respectively

757 * Starting composition; analysis of the recovered sample was not possible due to the leak of
758 the sample at further high temperature.

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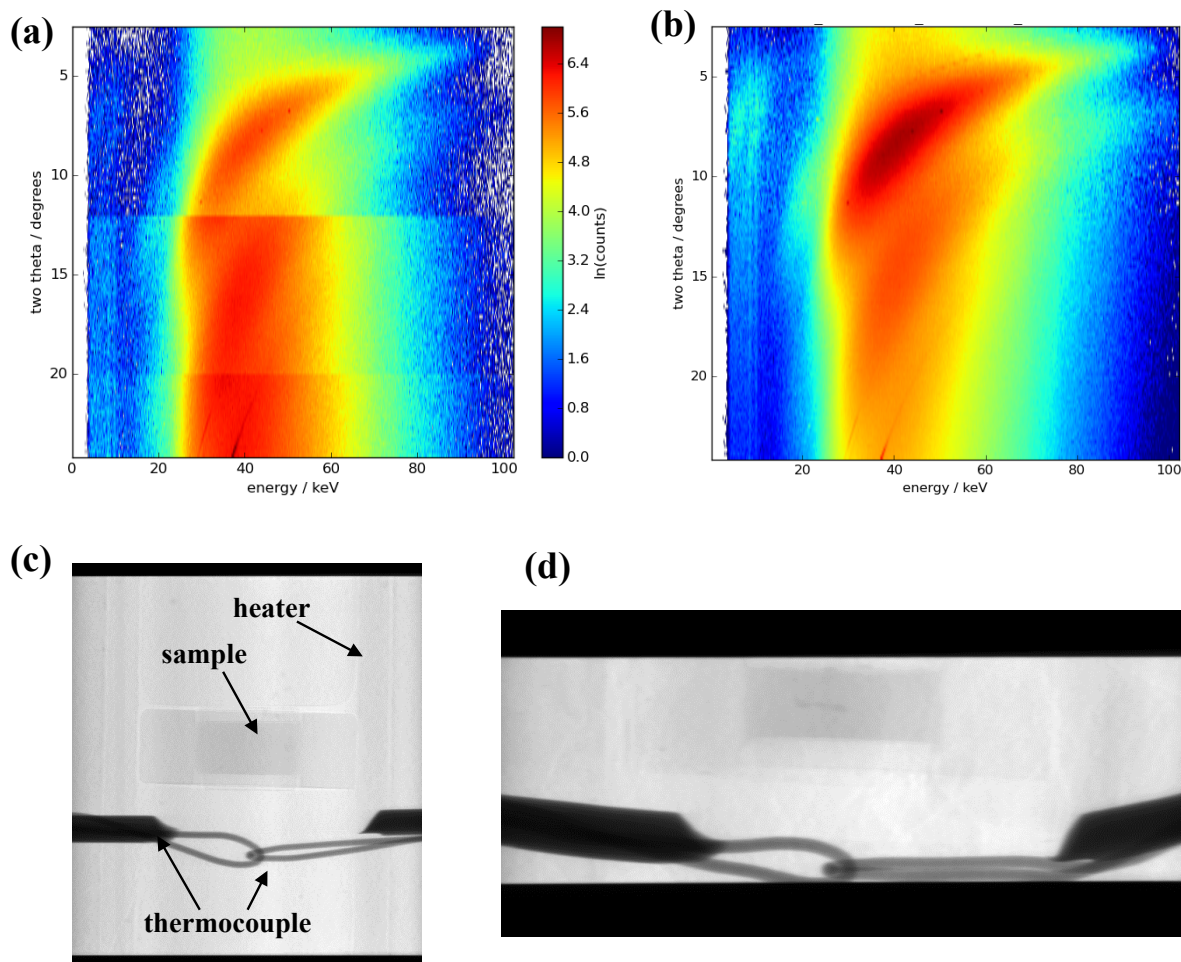
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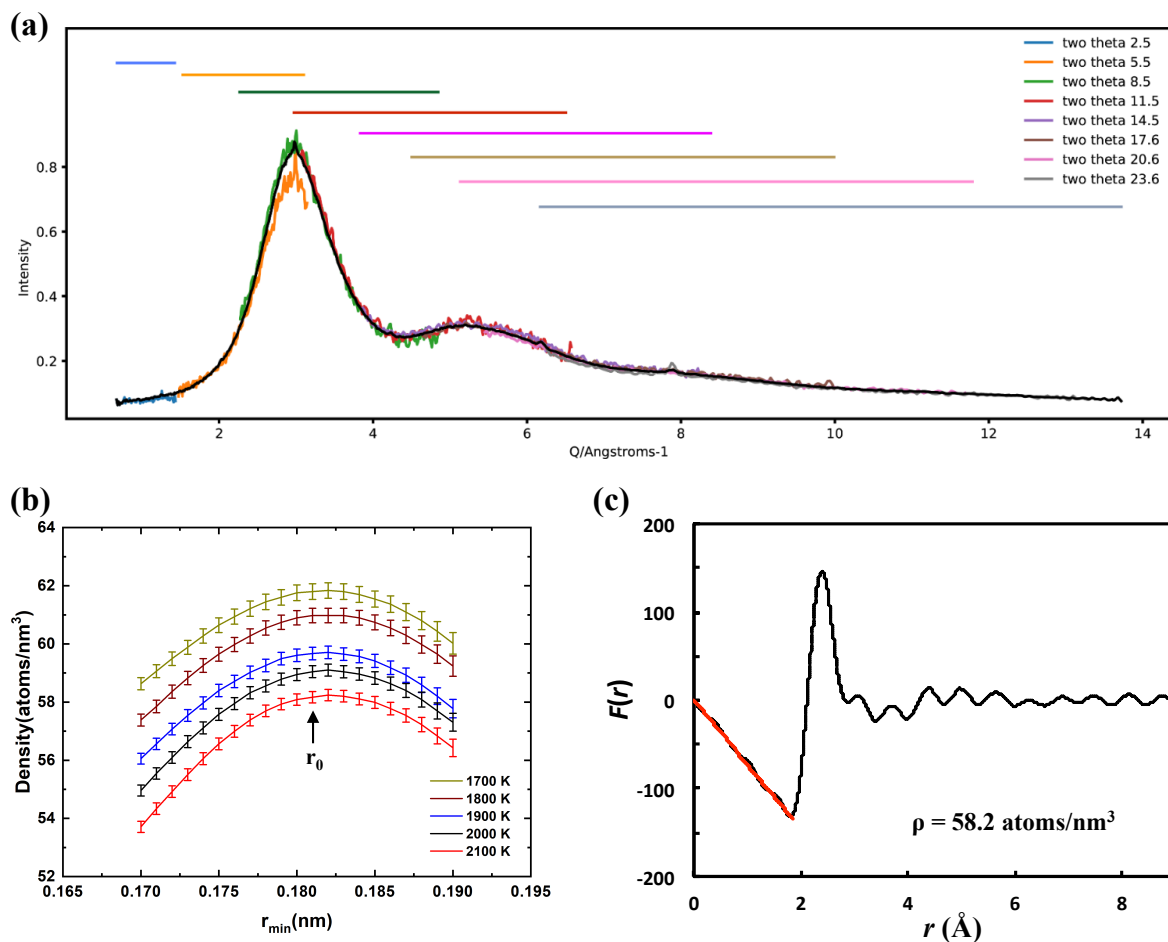
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768 **Fig. 1** (a, b) Two-dimensional CAESAR plot of diffraction intensities collected on liquid
769 FeS (run MA84). Raw 2D data (EDD spectra as a function of angle) (a) and data after
770 normalization for acquisition time and binning (b). The discontinuities visible in (a) for
771 $2\theta=12^\circ$ and 20° are due to the change of acquisition time. (c, d) Images of cell assemblies
772 obtained by X-ray radiography before compression (a) and at 6.3 GPa (c) after reaching to the
773 target load.

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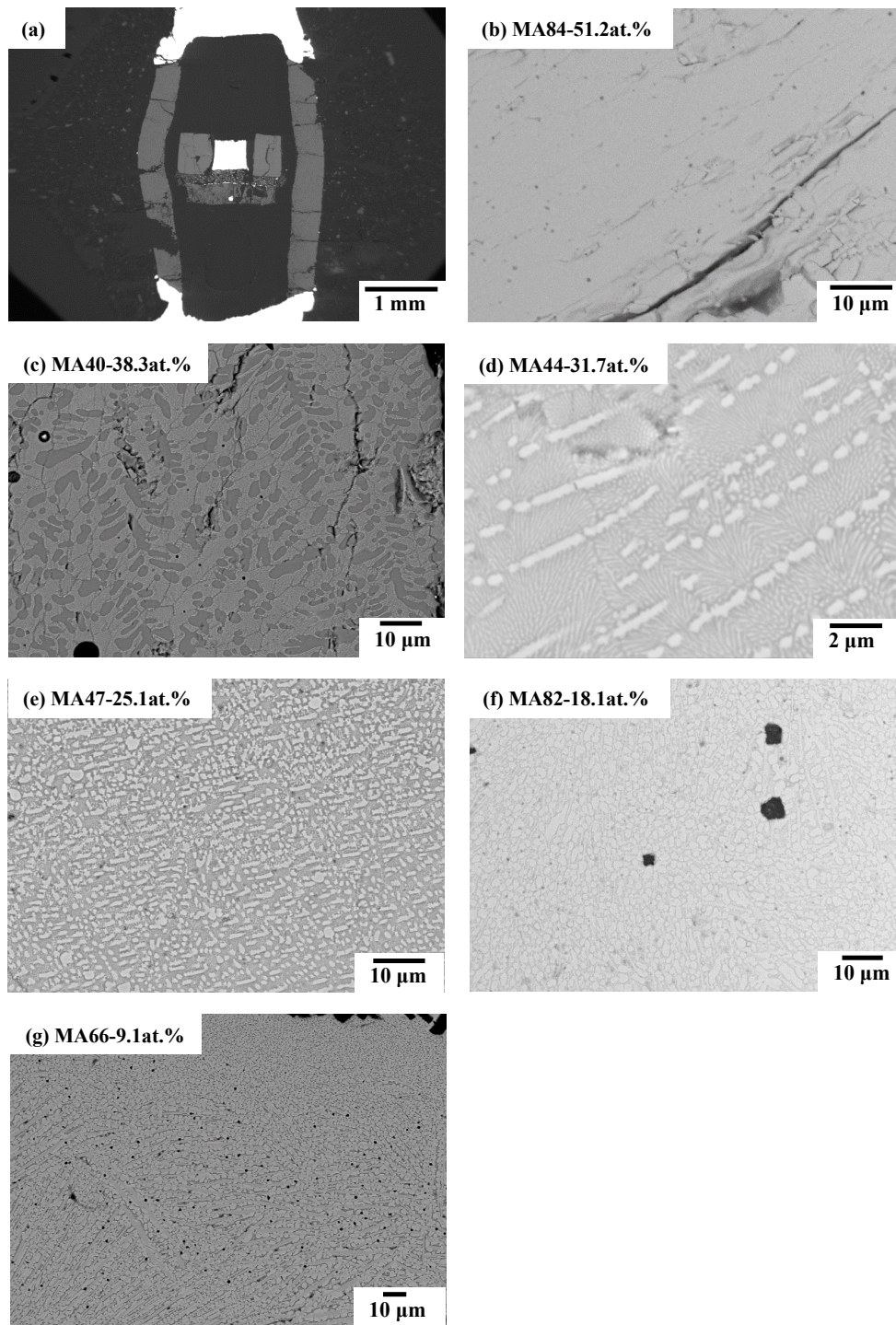
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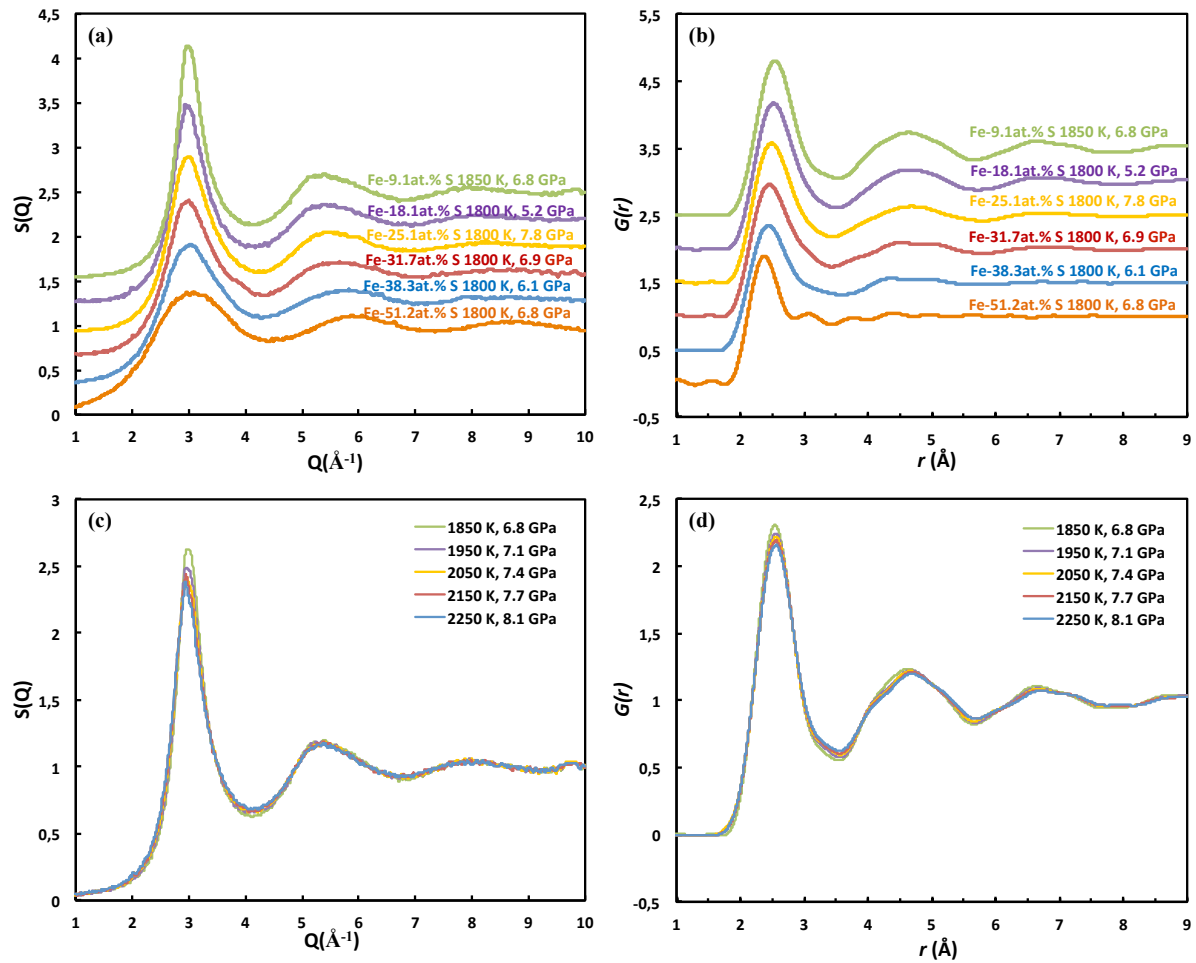
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Fig. 2 Examples of raw CAESAR data treatment process. (a) Normalized scattering intensity plotted as a function of Q . Color-coded horizontal bars and overlapping $I(Q)$ patterns illustrate the Q range covered and $I(Q)$ derived from the EDX spectra at corresponding 2θ . (b) Atomic densities calculated for different values of the minimal distance r_{\min} . Error bars indicate the value of merit χ^2 for each r_{\min} (see Morard et al. (2013) for more details). Arrow points out the local minimum of χ^2 , providing atomic density and r_0 for this data set. We stress that the temperature dependence of density is effectively independent of r_{\min} . (c) Comparison of the distribution function $F(r)$ with the function $-4\pi r \rho_0$ (dashed lines) calculated using density obtained by χ^2 minimization (d) (2100 K).



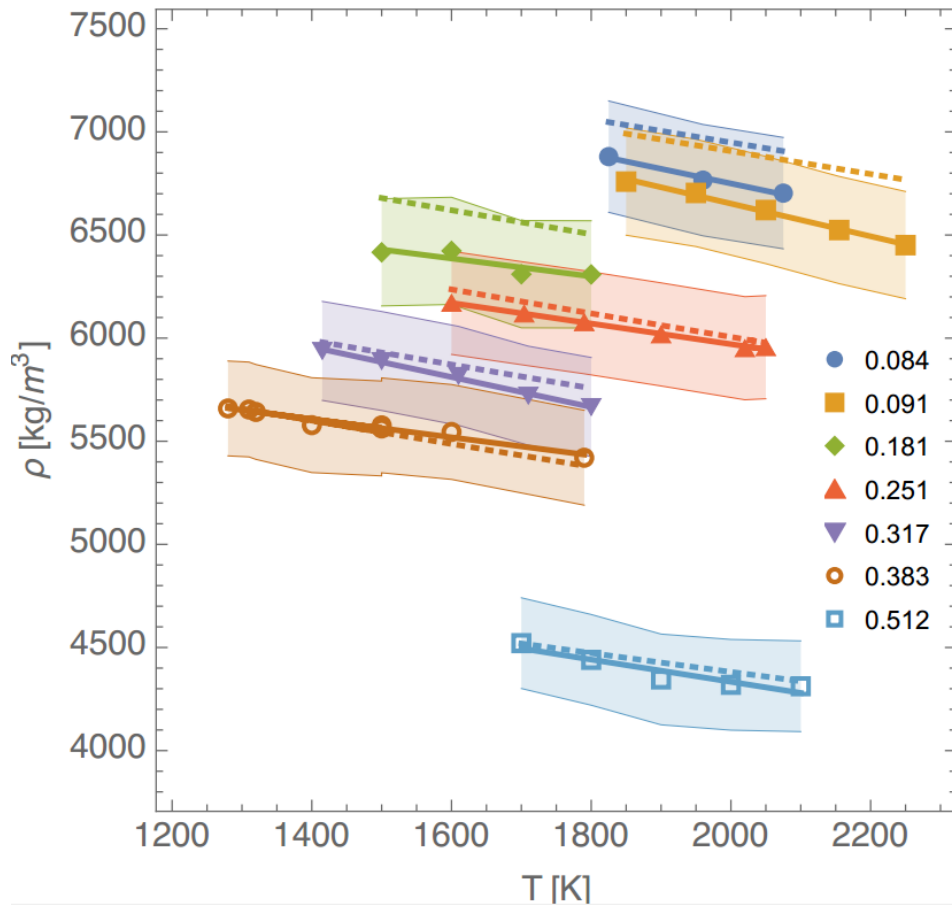
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Fig. 3 Backscattered electron image of recovered samples. (a) An overall view of the recovered cell after run MA47. (b-g) Microstructure of the recovered samples of the indicated runs. In (c-g) brighter areas correspond to Fe-rich portions of the sample, while darker areas correspond to S-rich portions.



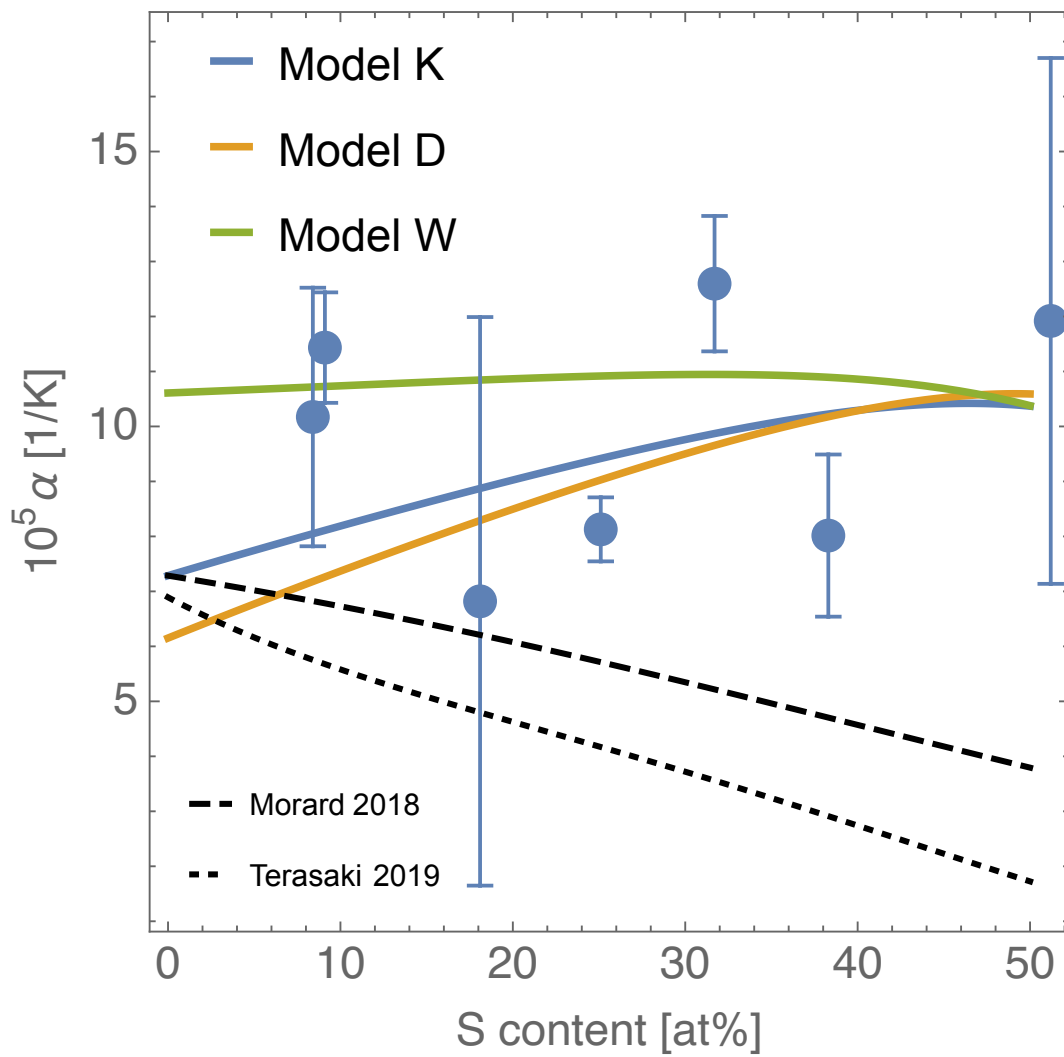
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Fig. 4 Examples of structure factor, $S(Q)$, and pair distribution function, $g(r)$, of liquids Fe-S alloys at high pressure and high temperature. (a) $S(Q)$ and (b) $g(r)$ as a function of S content. For clarity, $S(Q)$ and $g(r)$ are shown with a vertical offset of 0.3 and 0.5 in (a) and (b), respectively. Oscillations at ~ 4.4 and 5.0 Å visible in (b) for S concentration larger than 31.7at.% are not real features and arise from spurious signal due to the limited Q range in the corresponding structure factors. (c) $S(Q)$ and (d) $G(r)$ of Fe-9.1at.%S as a function of temperature (MA66).



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Fig. 5 Temperature dependence of the density of liquid Fe-S alloys at constant pressure (7 GPa). Linear fit of our data (solid line) and temperature derivative according to the thermodynamic Model W (dashed lines). Legend: atomic S concentration. Hatched areas indicate uncertainties on absolute density values (± 3 atoms/nm³, see section 2.3). Reported liquid compositions are those determined by chemical analysis of quenched and recovered samples but for the experiment on Fe-S_{0.084} for which we considered the composition of the starting material.



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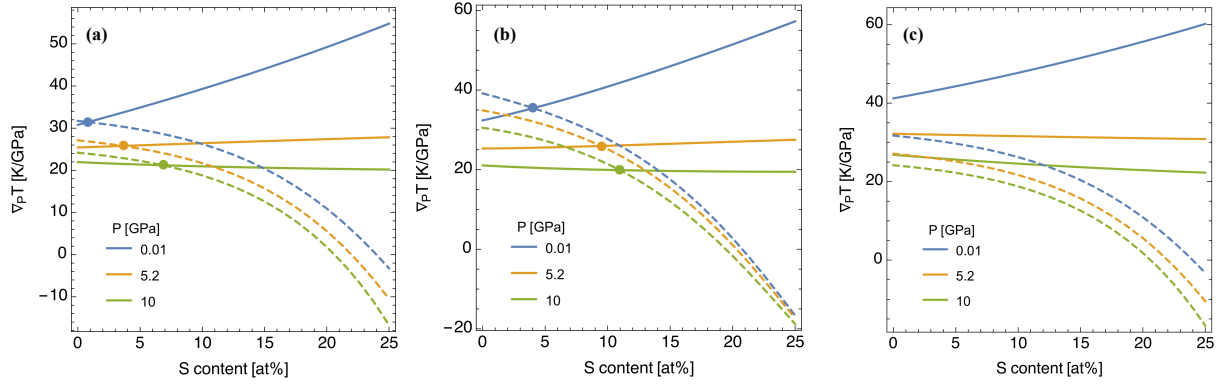
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830 **Fig. 6** Thermal expansivity as function of sulfur content at 7 GPa and 2000 K. Dots are
 831 estimations from experiments while colored lines are outcomes of thermodynamic models -
 832 Model K (blue), Model D (orange), and Model W) (green). Please refer to section 2.5 for
 833 details on the thermodynamic models and Table S1 and S2 for the used parameters. Black
 834 lines are results from literature thermodynamic models (dotted - Terasaki et al., 2019; dashed
 835 - Morard et al., 2018).

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842 **Fig. 7** Adiabatic gradient (solid lines) and slope of the liquidus (dashed lines) as a function of
 843 the S content at 0.01 GPa, 5.2 GPa, and 10 GPa, corresponding to core pressure of
 844 planetesimals, Moon and Ganymede, respectively, for the Model K (a), Model D (b), and
 845 Model W (2019) (c). Note that for the Model W the EOS' of solid Fe from Komabayashi
 846 (2014) has been used to compute the slope of the liquidi. For other differences in the models,
 847 please refer to section 2.5 and to Table S1 and S2 for the used parameters.

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851 Supplementary material for:

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853 **Thermal expansion of liquid Fe-S alloy at high pressure**

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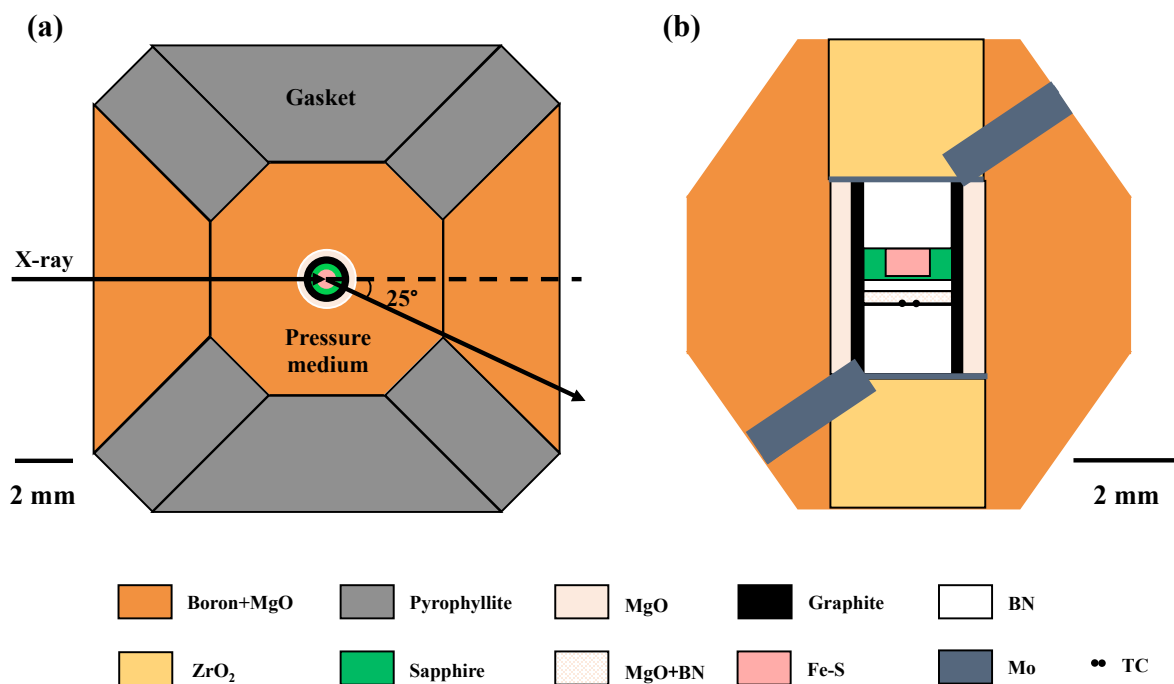
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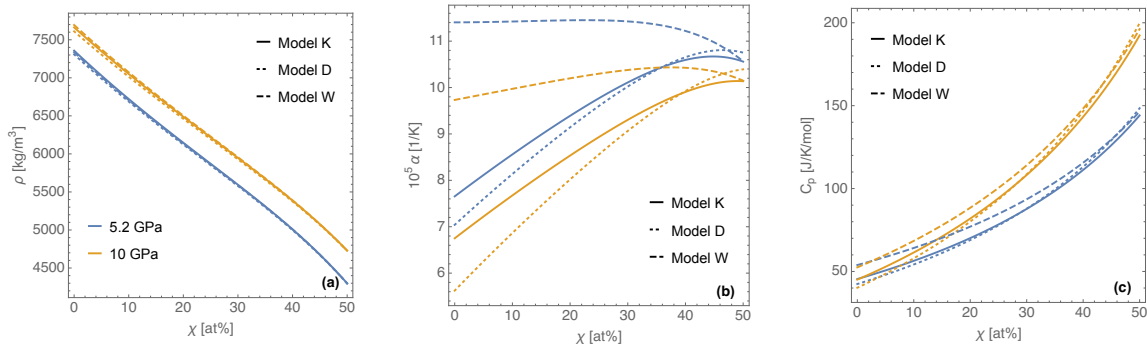
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Fig. S1 Schematic illustration of the experimental design. (a) A top view of the diffraction geometry through the gasket and cell assembly. (b) A cross-section of cell assembly used in this study. Temperature was monitored with a W₉₇Re₃-W₇₅Re₂₅ thermocouple (TC) whose junction was indicated by black dots.

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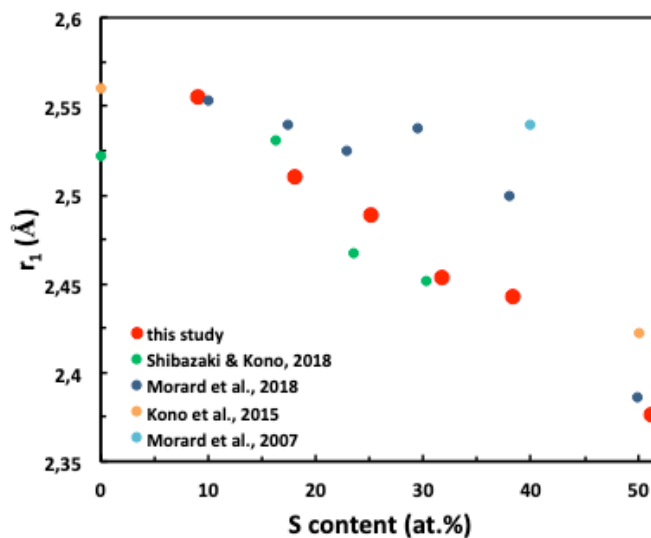
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896 **Fig.S2.** Density (a), thermal expansivity (b), and iso-baric heat capacity (c) as a function of
897 molar sulfur fraction X at 5.2 GPa and 10 GPa and 2000K for the thermodynamic Model K,
898 Model D and Model W. Model K, Model D and Model W differ in the reference EOS for
899 liquid iron (Model K uses l-Fe EOS of Komabayashi (2014), Model D uses l-Fe EOS of
900 Dorogokupets et al. (2017), Model W uses l-Fe EOS of Wagle and Steinle-Neumann (2019)).
901 Parameters of the thermodynamic models are in Table S1 and S2.

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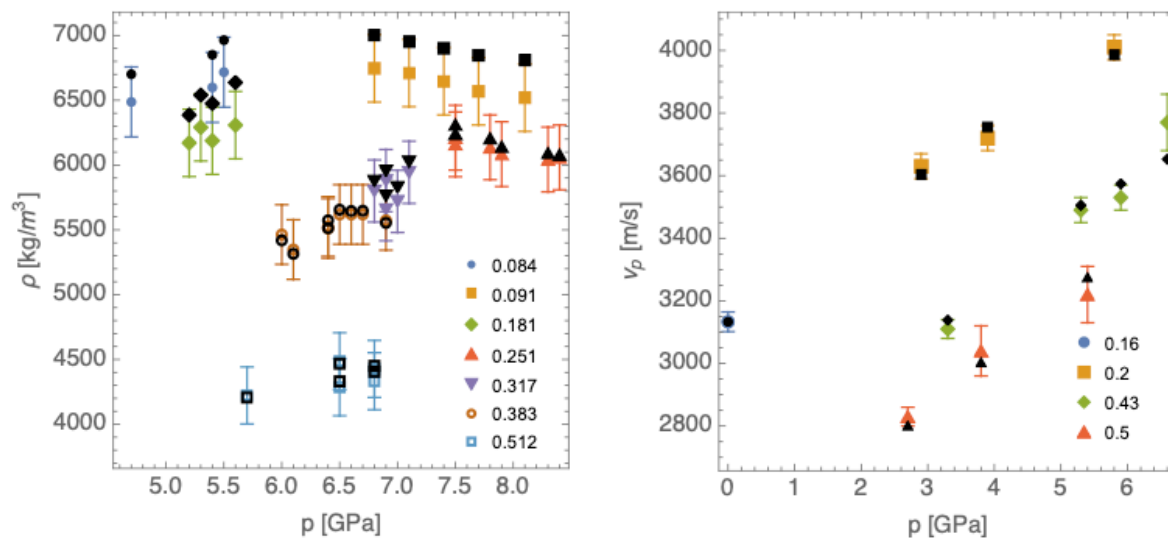
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Fig.S3. First peak position (r_1) as a function of the S content. Previous results from Shibazaki et al. (2018) at 3-5 GPa, Morard et al. (2018) at 2-5 GPa, Kono et al. (2015) at 1-6 GPa, and Morard et al. (2007) at 3-17 GPa, were also shown for comparison. For clarity only one value is plotted for each of the considered composition (r_1 for a given S content only moderately depends on pressure and temperature over the P-T range covered by individual studies).

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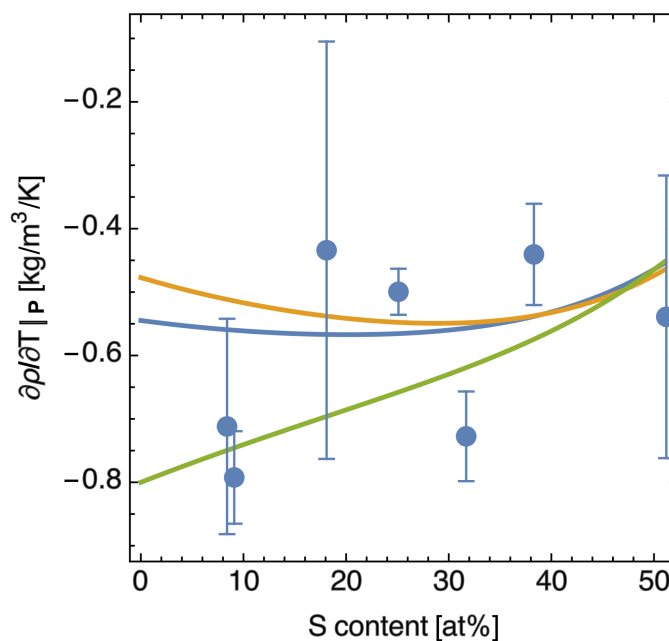
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Fig. S4. Measured densities (this study) and acoustic velocities (Nasch et al. 1994 and Nishida et al. 2016) (colored symbols) at varying temperature and predicted values according to the thermodynamic Model W (black symbols) under the same condition. Different symbols corresponds to different S content. For details on the thermodynamic model, please refer to section 2.5 and to Table S1 and S2 for parameters.

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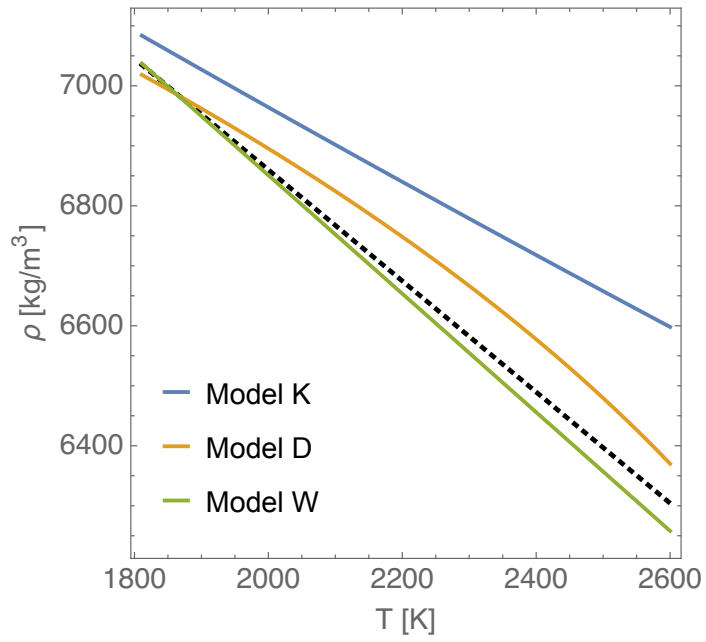
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931 **Fig. S5.** Temperature derivatives at the constant pressure (7 GPa) of density as a function
932 of S content. Dots are the slopes of the linear fit to the experimental data (see Fig. 6) with
933 uncertainties at 2σ . Colored lines are outcomes of thermodynamic Model K (blue), Model D
934 (orange) and Model W (green). Model K, Model D and Model W differ in the reference EOS
935 for liquid iron (Model K uses l-Fe EOS of Komabayashi (2014), Model D uses l-Fe EOS of
936 Dorogokupets et al. (2017), Model W uses l-Fe EOS of Wagle and Steinle-Neumann (2019)).
937 Parameters of the thermodynamic models are in Table S1 and S2.

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Fig. S6. Density as a function of temperature for liquid iron at ambient pressure. Colored lines are outcomes of our thermodynamic models (see section 2.5 and Table S1 and S3 for details). Dashed black line is a fit to data from Assael et al. (2006), and preferred fit according to Williams (2009).

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Table S1. EOS parameters for l-FeS and Margules parameters estimated from the experimental data of this study and from density measurements of Morad et al. (2018) and acoustic velocities from Nishida et al. (2016) using the EOS of l-Fe of Komabayashi (2014), Model K, Dorogokupets et al. (2017), Model D and Wagle and Steinle-Neumann (2019), Model W. $P_{\text{ref}} = 0.1\text{MPa}$, $T_{\text{ref}} = 1650\text{ K}$ and for the l-FeS end-member $\kappa = 1.4$, $\alpha = 11.8 \times 10^{-5} 1/\text{K}$, and $C_p = 62.5\text{ J/K/mol}$.

Thermodynamic model	l-Fe EOS	V [cm ³ /mol]	K_T [GPa]	K_T'	γ	δ_T	W_{Fe}	W_{FeS}	B_0 [GPa]	B'
Model K	Komabayashi 2014	24.25	13.22	6.36	0.68	0.62	-9.627	-3.435	3.45	2.14
Model D	Dorogokupets 2017	24.26	13.12	6.38	0.68	0.52	-9.481	-3.528	3.44	2.10
Model W	Wagle 2019	24.26	13.07	6.44	0.68	0.61	-9.627	-3.393	3.44	2.23

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Table S2. Margules parameters required to compute the Fe-S liquidus (see Eq. 9-11 Buono et al. 2011) for the EOS' of l-Fe of Komabayashi (2014) and Dorogokupets et al. (2017). For the Model W the EOS' of solid Fe from Komabayashi (2014) has been used to compute the slope of the liquidi.

Thermodynamic model	l-Fe EOS	$W_{H,Fe}$ [kJ/mol]	$W_{S,Fe}$ [kJ/mol/K]	$W_{V,Fe}$ [kJ/mol/GPa]	$W_{H,FeS}$ [kJ/mol]	$W_{H,FeS}$ [kJ/mol/K]	$W_{H,FeS}$ [kJ/mol/GPa]
Model K	Komabayashi 2014	53.70	0.029	-3.82	25.34	0.0	-2.95
Model D	Dorogokupets 2017	69.28	0.043	-2.78	27.3	0.0	-3.48

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