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Equation of State of Liquid Iron under Extreme Conditions

Yasuhiro Kuwayama,^{1,2,*} Guillaume Morard,^{3,4} Yoichi Nakajima,^{5,6,*} Kei Hirose,^{1,7} Alfred Q. R. Baron,⁶ Saori I. Kawaguchi,⁸ Taku Tsuchiya,² Daisuke Ishikawa,^{6,8} Naohisa Hirao,⁸ and Yasuo Ohishi⁸

¹*Department of Earth and Planetary Science, The University of Tokyo, 113-0033 Tokyo, Japan*

²*Geodynamics Research Center, Ehime University, 790-8577 Ehime, Japan*

³*Sorbonne Université, Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, IMPMC, Museum National d'Histoire Naturelle, UMR CNRS, 7590 Paris, France*

⁴*Université Grenoble Alpes, Université Savoie Mont Blanc, CNRS, IRD, IFSTTAR, ISTERre, 38000 Grenoble, France*

⁵*Department of Physics, Kumamoto University, 860-8555 Kumamoto, Japan*

⁶*Materials Dynamics Laboratory, RIKEN SPring-8 Center, 679-5148 Hyogo, Japan*

⁷*Earth-Life Science Institute, Tokyo Institute of Technology, 152-8550 Tokyo, Japan*

⁸*SPring-8, Japan Synchrotron Radiation Research Institute, 679-5198 Hyogo, Japan*

*To whom correspondence should be addressed. E-mails: kuwayama@eps.s.u-tokyo.ac.jp (Y.K.); yoichi@kumamoto-u.ac.jp (Y.N.)

Abstract

The density of liquid iron has been determined up to 116 GPa and 4350 K via static compression experiments following an innovative analysis of diffuse scattering from liquid. The longitudinal sound velocity was also obtained to 45 GPa and 2700 K based on inelastic x-ray scattering measurements. Combining these results with previous shock-wave data, we determine a thermal equation of state for liquid iron. It indicates that the Earth's outer core exhibits 7.5–7.6% density deficit, 3.7–4.4% velocity excess, and an almost identical adiabatic bulk modulus, with respect to liquid iron.

Main text

32 Iron is the sixth most abundant element in the universe and the main component of dense
33 metallic cores of planets. This is not only true for the Earth, but also for Mercury and Mars,
34 which are expected to have partially molten cores [1,2]. Density (ρ) and longitudinal sound
35 velocity (V_P) (equivalent to bulk sound velocity, V_Φ , in a liquid) are the primary observables
36 of the Earth's liquid outer core [3]. Therefore, laboratory measurements of these properties
37 at high pressure are of great importance to understand Earth's and other planets' core
38 composition and behavior.

39 While determination of density for crystalline materials under high pressure and
40 temperature (P - T) is relatively straightforward by *in situ* x-ray diffraction (XRD), it is still
41 challenging for disordered materials. Although XRD is potentially applied up to 100 GPa
42 and high temperature, analytical methods to extract ρ from a diffuse XRD signal, which is
43 characteristic of a liquid, are not yet well established; a recent study [4] concluded that a
44 conventional technique to analyze the diffuse signals gives a liquid density with uncertainty
45 exceeding more than 10%. Improvement of the diffuse scattering analysis is therefore
46 necessary. In particular, the density of liquid iron has not been reported at high pressure
47 based on static experiments.

48 V_P is also a key property to understand liquid behavior as it is related to compressibility,
49 thermal expansivity, the Grüneisen parameter (γ), etc. In particular, it is an important quantity
50 for constructing an equation of state (EoS) of a liquid when combined with density data.
51 Previously, the V_P of liquid iron was obtained only to 5.8 GPa by ultrasonic measurements
52 in a multi-anvil apparatus [5]. This is much lower than the pressure range of the Earth's core.
53 Moreover, the structure of liquid iron may be different above 6 GPa [6], indicating that
54 measurements are needed to higher pressures to understand the core.

55 In this study, we measured the density of liquid iron at pressures up to 116 GPa and 4350
56 K via static compression using a laser-heated diamond-anvil cell (LH-DAC). This is close to
57 conditions at the top of Earth's core. A new analytical method was applied to derive ρ from
58 diffuse x-ray scattering signals, as this is key to precise determination of liquid density under
59 pressure. We also obtained the V_P of liquid iron to 45 GPa by inelastic x-ray scattering (IXS)
60 measurements in the LH-DAC. From our new data combined with previous shock-wave data
61 [7,8], we obtain the P - T - ρ - V_P - γ relation for the Earth's entire outer core conditions (136–
62 330 GPa, 4000–5400 K) based on the Mie-Grüneisen EoS. The earlier shock compression

63 experiments measured the ρ , V_P , and γ of liquid iron only between 278 and 397 GPa along
 64 the Hugoniot path that intersects the melting curve of iron around 270 GPa [7]. The
 65 temperature in shock experiments is not well determined, being dependent on the model of
 66 internal energy of liquid iron. We therefore do not employ the temperature data reported in
 67 the shock experiments.

68 We collected angle-dispersive XRD spectra using a brilliant x-ray beam at BL10XU,
 69 SPring-8 [9] (see Supplemental Material [10], Sec. 1). Strong diffuse scattering signals from
 70 molten iron were found in the XRD spectra collected at about 100–400 K above its melting
 71 point (Fig. S1 in [10]). A background is subtracted (based on the measurement just below
 72 the melting point), and the result is converted into the structure factor $S(Q)$, where Q is the
 73 momentum transfer. Transformation of $S(Q)$ gives the distribution function $F(r)$ and the
 74 radial distribution function $g(r)$ (r , radial distance) [Fig. 1] (Eqs. S6 and S7 in [10]).

75 The density of the liquid can, in principle, be determined from the slope of $F(r)$ for r
 76 smaller than the inter-atomic spacing, where $F(r) = -4\pi\rho r$ and $g(r) = 0$. However, the
 77 transformation from Q to r requires integration over $Q \rightarrow \text{infinity}$. Experimental limits on
 78 the Q range result in oscillations in $F(r)$ and $g(r)$ that lead to large uncertainty in the
 79 determination of density if not corrected. An iterative analytical procedure originally
 80 developed in [11] has been applied for liquid density determinations at high pressure [12,13],
 81 but it often fails [4]. Indeed, it modifies $S(Q)$ from experimentally observed one, losing
 82 information from raw data.

83 In this study, we have developed an innovative analytical method in which the observed
 84 $S(Q)$ is extended beyond Q_{\max} (the maximum Q in experimental data) so that the
 85 corresponding $g(r)$ is physically reasonable; $g(r) = 0$ for $r < r_{\min}$ region (r_{\min} , the distance
 86 between the nearest neighboring atoms). We extend $S(Q)$ by;

$$87 \quad S_{\text{extend}}(Q) = \begin{cases} S(Q) & (Q \leq Q_{\max}) \\ 1 - \frac{1}{Q} \int_0^{r_{\min}} \left\{ 4\pi r \rho + \frac{2}{\pi} \int_0^{Q_{\max}} f(Q) \sin(Qr) dQ \right\} \sin(Qr) dr & (Q > Q_{\max}) \end{cases}, \quad (1)$$

88 where $f(Q) = Q\{S(Q) - 1\}$ (see Supplemental Material [10], Sec. 2). The transformed
 89 quantities $F_{\text{extend}}(r)$ and $g_{\text{extend}}(r)$ are calculated from the $S_{\text{extend}}(Q)$. However, there are several
 90 unknowns in the procedure, including a scale factor for background, s , normalization of $S(Q)$,
 91 α_N , as well as desired ρ and r_{\min} . If incorrect values are used for them, the difference between
 92 calculated $F_{\text{extend}}(r)$ and expected $F(r) = -4\pi\rho r$ (or between calculated $g_{\text{extend}}(r)$ and

93 expected $g(r) = 0$ at $r < r_{\min}$ will be larger compared to that calculated for true values. For
 94 instance, if input ρ includes error $\Delta\rho$ as $\rho_{\text{input}} = \rho_{\text{true}} + \Delta\rho$, $F_{\text{extend}}(r)$ calculated from $S_{\text{extend}}(Q)$
 95 involves an additional term given by;

$$96 \quad \frac{2}{\pi} \int_{Q_{\max}}^{\infty} \left\{ \int_0^{r_{\min}} -4\pi r \Delta\rho \sin(Qr) dr \right\} \sin(Qr) dQ, \quad (2)$$

97 (see Supplemental Material [10]). We sought the best $S_{\text{extend}}(Q)$, as well as s , α_N , ρ , and r_{\min} ,
 98 by minimizing χ^2 (see [11]) given by;

$$99 \quad \chi^2(s, \alpha_N, \rho, r_{\min}) \equiv \int_0^{r_{\min}} \{g_{\text{extend}}(r)\}^2 dr. \quad (3)$$

100 We searched for the minimum χ^2 in wide ranges of s , α_N , ρ , and r_{\min} ; 1 ± 0.5 for s (s is expected
 101 to be within 1 ± 0.05 , since the fluctuation in incident x-ray intensity was less than 5%), $\pm 50\%$
 102 from the value obtained by the Krogh-Moe and Norman's method [14,15] for α_N , $\pm 50\%$ from
 103 the density of solid iron at the P - T condition of an experiment for ρ , and between 0.15 and
 104 0.30 nm for r_{\min} (0.30 nm is larger than the first peak position in $g(r)$). For $r_{\min} < 0.15$ nm, a
 105 small sub-peak appeared between $r = 0.15$ nm and the dominant peak in $g(r)$. The existence
 106 of such sub-peak is unreasonable, because liquid iron is expected to be a simple monoatomic
 107 liquid.

108 In practice, the shape of artificial oscillations in $F_{\text{extend}}(r)$ and $g_{\text{extend}}(r)$ are also affected
 109 by Q_{\max} in Eq. 1 [16,17]. Therefore, we calculated χ^2 with changing Q_{\max} from the
 110 experimental limit to $\sim 30 \text{ nm}^{-1}$ that corresponds to the position at the end of the first dominant
 111 peak in $S(Q)$. The calculation shows that there is a unique Q_{\max} at $\sim 70 \text{ nm}^{-1}$ which minimizes
 112 χ^2 for each experimental data. When Q_{\max} is smaller than 65 nm^{-1} , the calculated χ^2 is larger
 113 than the minimum χ^2 by more than two orders of magnitude. For run #1 (Table I), for example,
 114 we find a unique set of these parameters that minimizes χ^2 ; $s = 1.0052$, $\alpha_N = 4.522$, $r_{\min} =$
 115 0.194 nm , and $Q_{\max} = 72.0 \text{ nm}^{-1}$, giving $\rho = 85.26 \text{ atoms/nm}^3$ (7.91 g/cm^3) [Fig. S9]. Figure
 116 2 shows $F_{\text{extend}}(r)$ and $g_{\text{extend}}(r)$ calculated from $S_{\text{extend}}(Q)$ for run #1, indicating that our
 117 procedure successfully reduced the oscillations in $F(r)$ and $g(r)$ at $r < r_{\min}$ that are mainly
 118 caused by a lack of data beyond experimental Q and inaccurate s and α_N . The uncertainty in
 119 ρ estimated from the difference between calculated $F_{\text{extend}}(r)$ and expected $F(r) = -4\pi\rho r$ is
 120 found to be less than $\sim 1\%$ (Table I). Note that our procedure does not modify $S(Q)$ at $Q \leq$
 121 Q_{\max} , in contrast to previous iterative analytical procedures.

122 The hard sphere model (HSM) [18] has often been applied for the structure of a single-
123 component liquid. For example, Ikuta *et al.* [4] determined the density of liquid aluminum
124 up to 6.9 GPa on the basis of fitting the HSM to experimentally obtained structure factor
125 $S(Q)$. The HSM, however, does not match the $S(Q)$ of liquid iron observed in this study at
126 relatively large Q [Fig. S2], indicating that the structure of liquid iron at high pressure is
127 more complex than the HSM. Similar discrepancies between observed $S(Q)$ for liquid metals
128 and the HSM have been reported [4,19]. The densities of liquid iron obtained on the basis of
129 the HSM are smaller than those determined by the present analyses by ~ 3 atoms/nm³ (~ 3 –
130 3.5%) for all of runs #1–11.

131 The sound velocity, V_P , of liquid iron was determined from IXS spectra collected at
132 BL43XU, SPring-8 [20,21] (Table I, see Supplemental Material [10], Sec. 3). The molten
133 state of a sample was confirmed by the absence of XRD peaks from solid iron, before and
134 after the IXS measurements. The IXS spectra included three peaks in the present scanned
135 energy range [Fig. 3(a)]; Stokes and anti-Stokes components of the longitudinal acoustic
136 phonon mode from the sample, and a quasi-elastic contribution near zero energy transfer.
137 We determined the V_P of liquid iron between 16 and 45 GPa from dispersion relations [Fig.
138 3(b)]. Note that the V_P of liquid iron is not sensitive to temperature [22–24]. While the
139 structure of liquidus crystal of iron changes from face-centered-cubic (fcc) to hcp at ~ 100
140 GPa, it does not likely affect the P – V_P relation for liquid since both are close-packed
141 structures and such effect is not found in the present P – ρ data [Fig. 4(a)].

142 Now we have both P – T – ρ and P – T – V_P data for liquid iron from the present study, in
143 addition to the P – ρ – V_P – γ relation from previous shock compression experiments [8]. From
144 these data, we obtain the P – T – ρ – V_P – γ relation across the Earth’s entire outer core conditions,
145 based on the Mie-Grüneisen EoS (see Supplemental Material [10], Sec. 4) [Figs. 4(a) and
146 4(b)]. We do not employ the ρ at 1 bar [25] nor the V_P determined below 5.8 GPa [5], but,
147 even so, our EoS reproduces these data well, suggesting that a possible structural change in
148 liquid iron below 6 GPa [6] has only a small impact on ρ and V_P .

149 In order to compare liquid iron properties with seismological observations [26], we
150 calculated the isentropic T profiles using γ determined in this study (see Supplemental
151 Material [10], Sec. 4), considering three different model temperatures at the liquid/solid core
152 boundary (inner core boundary, ICB) ($T_{\text{ICB}} = 5000$ K, 5400 K, and 5800 K) [3] [Fig. 4(c)].

153 Compared to the ρ , V_P , and adiabatic bulk modulus (K_S) of liquid iron calculated along the
154 isentrope with $T_{ICB} = 5400$ K, the Earth's liquid outer core exhibits low ρ by 0.99–0.81 g/cm³
155 (7.5–7.6%) [Fig. 4(d)] and high V_P by 0.43–0.29 km/s (4.3–3.7%) [Fig. 4(e)]. Such ρ deficit
156 is about 1% smaller than the previous estimates of 8.4–8.6% [8] that was based on the EoS
157 determined by a combination of the shock-wave data [7] including their uncertain T estimates
158 and the 1-bar data. In contrast, the observed K_S of the outer core is almost identical to that of
159 liquid iron [Fig. 4(f)]. Note that the K_S of liquid iron is not sensitive to temperature.

160 Seismology gives the density difference between the liquid and solid core at the ICB;
161 $\Delta\rho_{ICB} = 0.55\text{--}0.82$ g/cm³ [26–28]. Our results show that liquid iron is less dense than
162 hexagonal-close-packed (hcp) iron [29] by $\Delta\rho_{melting} = 0.32$ g/cm³ at 330 GPa and its melting
163 point of 6230 K [30]. This is larger than the previous estimates of 0.12–0.22 g/cm³ [31,32]
164 and approximately half of the observed $\Delta\rho_{ICB}$. Therefore, the remaining 0.23–0.50 g/cm³
165 (corresponding to 1.9–4.1% of the liquid core density at the ICB) should be attributed to a
166 compositional difference between the outer and inner core ($\Delta\rho_{comp}$).

167 Since the solubility of oxygen (O) in solid iron is negligible [33,34], oxygen has been
168 widely considered as a main light element in the Earth's core, in order to account for $\Delta\rho_{comp}$
169 [33]. Our revised density EoS of liquid iron indicates a smaller $\Delta\rho_{comp}$ that can be explained
170 with only 1.6–3.8 wt% O in the outer core and none in the inner core. This gives the upper
171 bound for oxygen concentration in the liquid core. However, 1.6–3.8 wt% O is not high
172 enough to explain the ρ deficit of the outer core with respect to liquid pure iron [35],
173 suggesting that oxygen is not a predominant light element in the core. While the light
174 elements in the core have not yet been identified [3], this study revealed that the ρ deficit is
175 constant at 7.5–7.6% and the V_P excess is also almost constant at 4.3–3.7% over the entire
176 outer core [26], which strongly constrains its possible compositional range.

177 The EoS is a fundamental macroscopic characteristic of a material. Our new analytical
178 procedure to derive liquid density from diffuse x-ray scattering signals can be applied to any
179 amorphous materials and may be used to explore the EoSs of other liquids. In addition, this
180 work demonstrates that a combination of high-pressure density and velocity data enables a
181 precise determination of the EoS. These data can now be obtained not only for solids but for
182 liquids via XRD and IXS measurements even at extreme high P - T conditions by using LH-
183 DAC techniques. For further understanding the nature of amorphous materials under

184 compression, future technical developments to use higher energy x-rays are necessary to
185 collect diffuse signals in a wider Q range.

186

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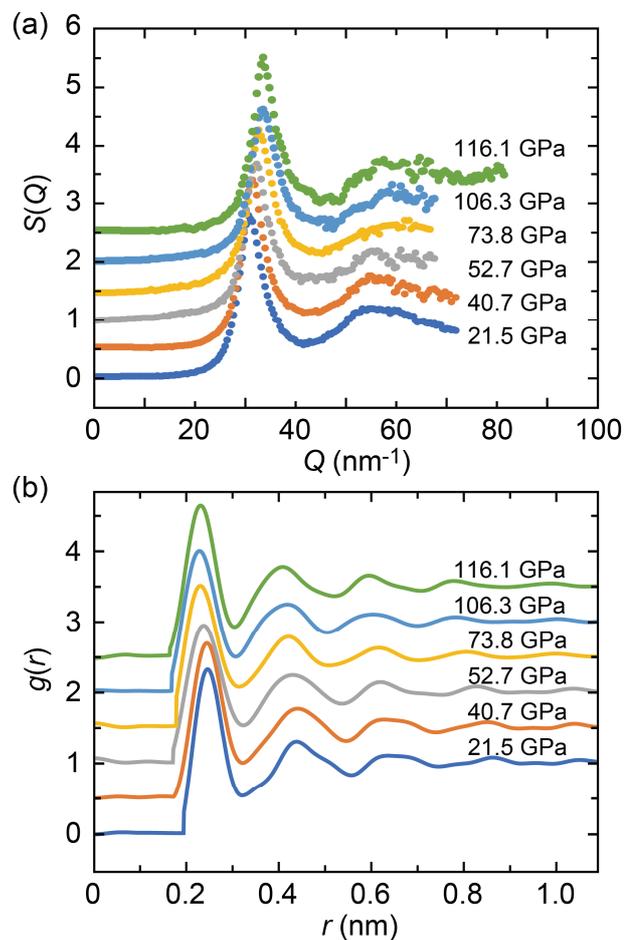
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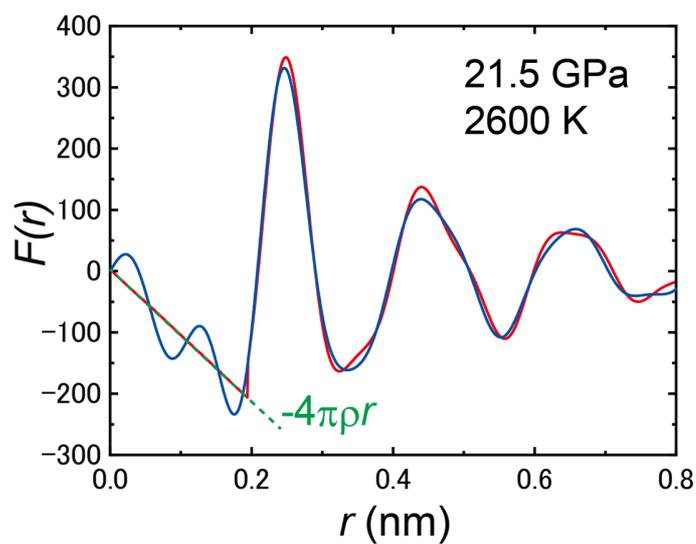
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286 FIG. 1. Structural analyses of liquid iron at high pressures via in-situ XRD measurements.
 287 (a) Structure factor, $S(Q)$, of liquid iron up to 116.1 GPa determined from XRD
 288 measurements in this study, showing peak shifts to larger Q values due to the compression
 289 of liquid iron. (b) Corresponding radial distribution functions, $g(r)$, determined in this study.
 290 Vertical scales are offset for clarity for both $S(Q)$ and $g(r)$ plots.

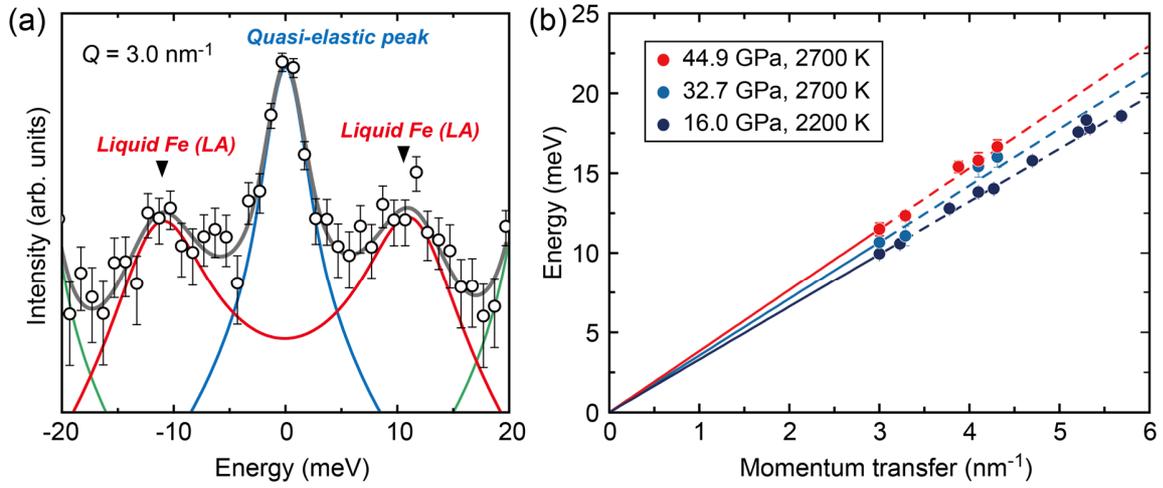
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294 FIG. 2. Distribution function $F(r)$ from run #1 calculated based on the present analytical
295 method (red), in which the extension of $S(Q)$ and parameters of s , α_N , and ρ are determined
296 simultaneously, demonstrating that our new method successfully reduces the oscillations at
297 $r < r_{\min}$ and gives a precise liquid density from the slope. $F(r)$ calculated without extension
298 of $S(Q)$ with assuming $s = 1$ is shown by the blue line for comparison.
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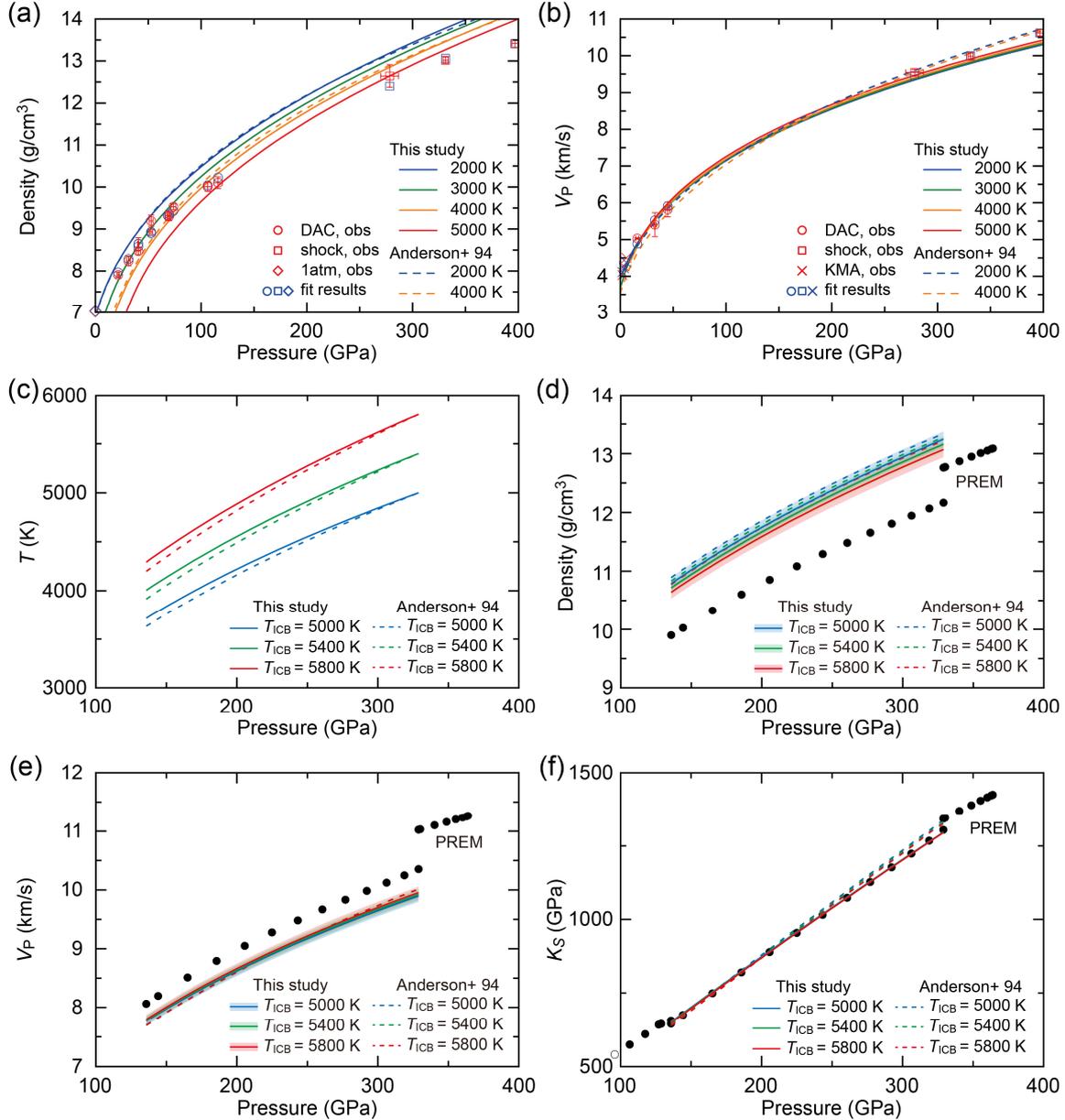


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302 FIG. 3. High-pressure inelastic x-ray scattering (IXS) measurements of liquid iron. (a)
303 Typical IXS spectrum of liquid iron collected at 44.9 GPa and 2700 K at momentum transfer
304 $Q = 3.0 \text{ nm}^{-1}$. The spectra include three components: a quasi-elastic peak near zero energy
305 transfer (blue), longitudinal acoustic (LA) phonon mode of liquid Fe (red), and transverse
306 acoustic (TA) phonon mode of diamond (green). The vertical axis is plotted on a logarithmic
307 scale. (b) Longitudinal acoustic phonon dispersion of liquid iron at pressures from 16.0 to
308 44.9 GPa.
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313 FIG. 4. Density (ρ), P-wave velocity (V_P), and adiabatic bulk modulus (K_S) of liquid iron. (a),
 314 (b) Isothermal P - ρ and P - V_P relations calculated from our EoS for 2000 K (blue), 3000 K
 315 (green), 4000 K (yellow), and 5000 K (red) (Table SII). Dashed lines, 2000 K (red) and 4000
 316 K (yellow), are from shock-compression study [8]. Red symbols represent experimental data
 317 (circles, this study; squares, shock experiments [8]; crosses, multi-anvil experiments [5];
 318 diamonds, 1 bar data at 1811 K [25]). Consistency between the red and blue (fit results)
 319 symbols indicates that our EoS well reproduces all experimental data points. (c) Calculated
 320 isentropic temperature profiles with $T_{ICB} = 5800$ K (red), 5400 K (green), and 5000 K (blue)

321 (Table III) (see Supplemental Material [10], Sec. 4). Dashed lines are those proposed by a
322 previous study with a different Grüneisen parameter [8]. (d), (e), (f) Comparison of seismic
323 observations (black circles, PREM [26]) with the ρ , V_P , and K_S of liquid Fe under core
324 pressures along the isentropic temperature profiles in (c). Uncertainties in the present
325 estimates of ρ and V_P are $\sim 1\%$ (see the uncertainty band around each solid curve and
326 Supplemental Material [10], Sec. 4 for details). Dashed lines represent those proposed on the
327 basis of earlier shock-wave data [8].
328

329 Table I. Experimental results on liquid iron.

<i>P-T-ρ</i> relation determined from XRD measurements			
run no.	<i>P</i> (GPa) [†]	<i>T</i> (K)*	<i>ρ</i> (g/cm ³) [†]
#1	21.5(12)	2600	7.91(7)
#2	31.3(17)	2870	8.24(11)
#3	40.6(5)	2880	8.64(15)
#4	40.7(21)	3060	8.48(9)
#5	52.7(16)	3250	8.93(7)
#6	52.8(18)	3340	9.19(13)
#7	68.5(22)	3530	9.32(10)
#8	69.8(19)	3540	9.30(11)
#9	73.8(24)	3630	9.53(7)
#10	106.3(35)	4250	10.01(11)
#11	116.1(39)	4350	10.10(14)
<i>P-T-V_P</i> relation determined from IXS measurements			
run no.	<i>P</i> (GPa) [†]	<i>T</i> (K)*	<i>V_P</i> (km/s) [†]
#12	16.0(16)	2200	5.03(12)
#13	32.7(11)	2700	5.40(32)
#14	44.9(20)	2700	5.82(20)

[†]The numbers in parentheses represent one standard deviation in the last digits.

*±10% uncertainty [10].

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