

1 Revision 1

2 **Precise determination of the effect of temperature on the density of solid and liquid**  
3 **iron, nickel, and tin**

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8

9 **Abstract**

10 Density and thermal expansion coefficient of metals are one of the most  
11 fundamental characteristics to describe the equation of state. Especially for liquid metals,  
12 the reported data for density and thermal expansion coefficient vary in the literature,  
13 even at ambient pressure. To determine density of solid and liquid metals precisely at  
14 high temperatures and ambient pressure, we newly developed a high-temperature  
15 furnace. The densities of solid Sn, Ni, and Fe were determined from sample image with  
16 uncertainty of 0.11%–0.7% in the temperature range of 285–1803 K with increments of  
17 1–10 K. The density of solid Sn decreased linearly with increasing temperature up to  
18 493 K, and the decrease became drastic above that to the melting temperature ( $T_m$ ). By

19 contrast, for solid Ni and Fe, the densities decreased linearly with increasing  
20 temperature up to the  $T_m$  (1728 and 1813 K) without any drastic density drop near  $T_m$ .  
21 This suggests that Ni and Fe do not exhibit the “premelting effect”.

22 For liquid, the density of liquid Fe was determined with uncertainty of 0.4%–  
23 0.7% in the range of 1818–1998 K with temperature increments of 5 K. The obtained  
24 thermal expansion coefficient ( $\alpha$ ) of liquid Fe was well approximated as either a  
25 constant value of  $\alpha = 2.42(1) \times 10^{-4} \text{ K}^{-1}$  or a linear function of temperature ( $T$ );  $\alpha =$   
26  $1.37(10) \times 10^{-3} - [6.0(6) \times 10^{-7}]T [\text{K}^{-1}]$  up to at least 2000 K.

27

28 **Keywords:** Density, Thermal expansion, Metal, Liquid

29

## 30 INTRODUCTION

31 Density is one of the most fundamental physical properties for understanding  
32 planetary interiors. To estimate the structure and composition of a planetary core, the  
33 equation of state (EOS) of Fe alloys is an indispensable information, along with  
34 planetary exploration data. The density of Fe alloys at ambient pressure is one of the  
35 most fundamental characteristics used to describe the EOS, as well as its elastic  
36 properties (bulk modulus,  $K$ , its pressure derivative [ $dK_T/dP$ ], and the thermal expansion

37 coefficient,  $\alpha$ ). Although the density and thermal expansion of the core constituting  
38 metals (Fe and Ni) have been reported, they have not been determined precisely near the  
39 melting temperature ( $T_m$ ). An unusual drastic decrease in the elastic wave velocities  
40 near  $T_m$ , the so-called “premelting effect,” has been reported for some solid metals (Sn  
41 and Fe) based both on experiments and theoretical calculations. It is reported to occur at  
42  $T/T_m > 0.99$  for Sn at ambient pressure (Nadal and Le Poac, 2003) and at  $T/T_m > 0.96$   
43 for Fe at 360 GPa (Martorell et al., 2013a). If the premelting effect occurs in the elastic  
44 wave velocity, it could also occur in density, which is closely related to the elastic wave  
45 velocity, and thermal expansion. This could significantly influence the EOS and elastic  
46 properties of the core. However, the density behaviors of Sn and Fe near  $T_m$  have not  
47 been precisely known.

48 For liquid density, the density ( $\rho$ ) of liquid Fe at ambient pressure has been  
49 reported by many authors (e.g., Luca, 1960; 1972; Kirshenbaum and Cahil, 1962;  
50 Froberg and Weber, 1964; Brillo and Egry, 2004; Assael et al., 2006; Kobatake and  
51 Brillo, 2013; Le Maux et al., 2019). However, the reported  $\rho$  values have some  
52 significant deviation ( $\Delta\rho \sim 0.2 \text{ g/cm}^3 \sim 2.8\%$ ) and the thermal expansion coefficient has  
53 not been precisely constrained yet.

54 In this study, density measurements of solid Sn, Ni, Fe, and liquid Fe were

55 performed using a newly developed high-temperature furnace with observation  
56 windows. The effect of temperature on the density of solid Sn, Ni, Fe, and liquid Fe was  
57 precisely investigated at ambient pressure.

58

## 59 METHODS

### 60 **Experimental methods**

61 The samples were metal rods of Sn (99.9% in purity, Nilaco Corp.), Ni (99.5%  
62 in purity, Nilaco Corp.), and Fe (99.9% in purity, Nilaco Corp.). The diameter and  
63 height of the sample were 2.99–3.00 and 2.09–3.00 mm, respectively. The top and  
64 bottom interfaces of the samples were mirror-polished.

65 The experiments were conducted using a high-temperature gas furnace  
66 (Nagano Co Ltd., Tokyo) installed at Osaka University (Fig. 1a). Heating was  
67 performed using a composite resistance heater composed of carbon  
68 fiber-reinforced carbon composite (C/C). The C/C heater is superior to generate high  
69 temperatures (~2173 K) stably under vacuum or inert gas conditions, and has a small  
70 thermal expansion coefficient, which enables rapid heating (~1500 K/min). The C/C  
71 heater plate is located around the sample stage and has a slit for sample observation.  
72 The sample is set on a sintered alumina plate located at the center of the chamber (Fig.

73 1b). During the experiment, the temperature was monitored using a K-type  
74 thermocouple (TC). The TC junction was located at same height as the sample and 18.6  
75 mm away from the sample center horizontally. The  $T_m$  values of Ni and Fe were  
76 determined to be 1728 and 1812 K (TC temperature), respectively, based on the shape  
77 change of the sample. These results are in good agreement with the literature. Therefore,  
78 the temperature at the TC junction was identical to that at the sample center.  
79 Temperature was controlled using a proportional integral differential controller. The  
80 heating rates were in the range of 3–25 K/min. The measurement was performed under  
81 an argon atmosphere at 0.05 MPa (flow rate: 0.8–1.0 l/min).

82         The chamber had two optical windows for observation of the sample in the  
83 horizontal direction. For the density measurements, a charge-coupled device (CCD)  
84 (1296(H)x966(V), 12 bit, acA1300–30um; Basler AG) or complementary metal-oxide  
85 semiconductor (CMOS) camera (2048(H)x1536(V), 12 bit, BU302MG; Toshiba Teli  
86 corp.) was set on one side for optical observation, and a backlight (Fiber optic  
87 illuminator) was set on the other (Fig. 1a). The image magnification was optimized  
88 using a telecentric zoom lens (x0.5–1.3) (VS Technology Corp.). The pixel size of the  
89 image was 3.9–4.5  $\mu\text{m}/\text{pixel}$  depending on the magnification. Details of the pixel  
90 calibration and evaluation of image distortion are given in Supplement<sup>1</sup> (I). The

91 exposure time of each image was in the range of 1–80 ms. Images of the sample were  
92 taken with 1–10 K steps and analyzed using the method described in next section.

93

#### 94 **Analytical methods**

95 The density of the sample was obtained from the sample volume at each  
96 temperature and initial mass. The sample volume was measured from the 2-dimensional  
97 (2D) sample image assuming rotational (cylindrical) symmetry. Image analysis was  
98 carried out using ImageJ software (<https://imagej.nih.gov/ij/>). The sample image was  
99 binarized, and brightness profiles of the image were obtained to identify the boundary  
100 between the sample and surrounding part.

101 For solid samples, the diameter and height were measured from the sample  
102 brightness profiles (Figs. 2a and 2b). To obtain a clear image of sample bottom  
103 boundary which is required for precise determination of the sample height, a small  
104 alumina disk is placed underneath the sample (Fig. 2a). In the run AK12 that the small  
105 alumina disk was not used, height error of AK12 is much (one-order) larger than that of  
106 other runs in which the small disk was used (Supplemental<sup>1</sup> Table S4). The validity of  
107 the cylindrical symmetry of the sample was evaluated from images observed from  
108 different directions in separate measurements. The sample was put on the rotation stage

109 at room temperature and the image was observed at rotation angles of 0°, 45°, and 90°.  
110 For 0–90° rotation, the variations were  $(1.0\text{--}2.0)\times 10^{-4}$  cm in diameter and  $(1.0\text{--}3.0)\times 10^{-4}$   
111 cm in height, which corresponds to a volume variation of  $\Delta V=(1.4\text{--}4.6)\times 10^{-5}$  cm<sup>3</sup> and a  
112 density variation of  $\Delta\rho=(8.4\text{--}20.5)\times 10^{-3}$  g/cm<sup>3</sup> (0.11~0.28%). Therefore, these density  
113 variations derived from rotation were small, and the cylindrical symmetry used in this  
114 study could be valid for obtaining the sample volume. We included this density  
115 variation in the density error (density errors are discussed in next paragraph). The  
116 obtained volumes and calculated densities of solid metals (Sn, Ni, and Fe) are  
117 summarized in Supplemental<sup>1</sup> Table S1–S3. In some runs, the density of solid sample  
118 was measured at room temperature from sample image (see Table S1–S3). The  
119 measured density (Sn: 7.278(15), 7.289(13) g/cm<sup>3</sup>, Fe: 7.868(9) g/cm<sup>3</sup>) agree well with  
120 the density database at room temperature (Sn: 7.265 g/cm<sup>3</sup>, Fe: 7.874 g/cm<sup>3</sup>,  
121 Chronological Scientific Tables, 2020). Therefore, it is confirmed that the density  
122 obtained from measurement used in this study is consistent with the value of density  
123 database.

124 Density error of solid samples contains following contributions. (1) *Errors of*  
125 *sample lengths*. The sample diameter and height were obtained from horizontal and  
126 vertical brightness profiles of the sample image, respectively (Fig. 2b). The length

127 errors were taken from difference between maximum (minimum) and average measured  
128 lengths. The volume (density) error was derived from the propagation of these length  
129 errors. The density error derived from length errors are summarized in Supplemental<sup>1</sup>  
130 Table S4. (2) *Rotation error*. The sample volume variation derived from sample rotation  
131 of 0–90°, as stated in previous paragraph, was included in volume (density) error as  
132 summarized in Table S4. (3) *Error of pixel calibration*. As mentioned in supplement (I),  
133 length error derived from pixel calibration are 0.004–0.01 μm. The density error derived  
134 from this length error is calculated to  $3 \times 10^{-4}$ – $8 \times 10^{-4}$ % based on error propagation,  
135 suggesting that the density error from pixel calibration error is negligibly small. Thus,  
136 the density error of solid sample in this study correspond to a sum of contribution of  
137 length errors and that of rotation error. All these errors are summarized in Table S4. In  
138 run AK19, for example, total density errors are 0.137–0.162%, which is obtained from a  
139 sum of density error from length errors (0.004–0.027%) and that from rotation error  
140 (0.132–0.135%).

141 For liquid samples, the volume of a sample droplet was obtained from a 2D  
142 image using Pappus’s centroid theorem, which describes the volume of rotating bodies  
143 as:

$$V = 2\pi RS \quad (1)$$



144 where  $S$  and  $R$  indicate a half area of the sample cross section divided by the rotating  
145 axis and the distance between the rotation axis and center of gravity, respectively. The  
146 value of  $S$  was determined from the 2D image, as shown by the shaded area in Fig. 3a.  
147 The sample boundary was identified from the vertical scan of horizontal brightness  
148 profiles. The initial image was cut at the rotation axis. Next, the image was binarized,  
149 and the area ( $S$ ) and coordinates of the center of gravity were obtained. More detailed  
150 analysis procedure of liquid sample is provided in Supplement<sup>1</sup> (II). The obtained  $V$  and  
151  $\rho$  of liquid Fe are summarized in Supplemental<sup>1</sup> Table S5. The error in volume was  
152 derived from the following errors: (1) error in sample length and rotational symmetry,  
153 and (2) error in boundary threshold. (1) We calculated the sample volume using the  
154 images of the left and of right halves with equation (1) to verify the assumption of  
155 rotational symmetry. The volume (density) difference from left and right half images  
156 corresponds to density error derived from rotation. Sample length errors are included in  
157 the calculated volume. The results are listed in Supplemental<sup>1</sup> Table S6. In run AK20,  
158 for example, the density error from rotation is 0.01–0.038%. This small rotation error  
159 indicates that the liquid sample was symmetric to the rotation axis. (2) The error in the  
160 boundary threshold is derived mainly from gray pixels (6 to 14 pixels) at the edge  
161 boundary of the droplet as shown in the inset of Fig. 3b. We estimated the volume error

162 by changing the threshold value in the range of the gray pixels. The error in boundary  
163 threshold was improved by taking a shadow picture of the sample using a backlight  
164 following the method of Le Maux et al. (2019). The backlight has been used in previous  
165 studies (e.g., Brillo and Egry 2003; Langstaff et al. 2013; Le Maux et al. 2019). The  
166 estimated density error from boundary threshold are listed in Table S6. Thus, the density  
167 errors of liquid sample correspond to a sum of these errors (i.e., length, rotation  
168 symmetry, and boundary threshold) and summarized in Table S6. For example, in AK20,  
169 total density errors are 0.400–0.670%, which is obtained from a sum of density error  
170 from rotation and length errors (0.001–0.038%) and that from boundary threshold error  
171 (0.027–0.666%).

172 The recovered samples were mounted in epoxy and polished for textural  
173 observation and chemical analysis, which were carried out using a scanning electron  
174 microscope (JSM-6010LA; JEOL) equipped with an energy-dispersive spectrometer  
175 (NORAN NSS EDX; Thermo Fischer Scientific) with an accelerating voltage of 20 kV.  
176 Chemical composition of starting and recovered samples are listed in Supplemental<sup>1</sup>  
177 Table S7. Supplemental<sup>1</sup> Fig. S3 shows example of backscattered electron image of the  
178 Fe samples quenched from liquid (AK20). From the sample textures of backscattered  
179 electron images and chemical composition of the recovered samples, no contamination

180 was found in the recovered Sn, Ni, and Fe samples (Supplemental<sup>1</sup> Table S7).

181

## 182 RESULTS AND DISCUSSION

### 183 **Density of tin**

184 The measured density of Sn (body-centered tetragonal [bct] structure) is plotted  
185 as a function of temperature in Fig. 4 (Data is found in Supplemental<sup>1</sup> Table S1). The  
186 density values from two measurements (AK11 and AK14) were reproducible. The  
187 density decreased almost linearly with increasing temperature up to 493 K, which  
188 corresponds to  $T/T_m < 0.98$ . This trend is consistent with a previous study (Touloukian  
189 et al., 1975). However, above 493 K, a drastic decrease in density was observed (see Fig.  
190 4). The decrease in density was approximately 0.7%, which is much larger than the  
191 density error (0.18–0.22%). This phenomenon was possibly derived from the premelting  
192 effect. In a previous study, the premelting effect observed in  $V_P$  and  $V_S$  at  $T/T_m > 0.99$   
193 was considered to occur as a result of decrease in shear modulus (Nadal and Le Poac,  
194 2003). It was found that Sn has a premelting effect near the  $T_m$  for both the density and  
195 elastic wave velocities.

196 The thermal expansion coefficient ( $\alpha$ ) was determined by fitting the density  
197 data up to 493 K with equation (2):

198 
$$\rho = \frac{\rho_0}{\exp(\int_{T_0}^T \alpha dT)} \quad (2),$$

199 where  $\rho_0$  denotes the density at the reference temperature  $T_0$ . The  $\alpha$  of this study and a  
200 previous study are listed in Table 1. In this study,  $\rho_0$  was 7.278(15) at  $T_0 = 300$  K, and  
201 the obtained  $\alpha$  in the temperature range of 285–490 K was  $6.22(3) \times 10^{-5} \text{ K}^{-1}$ . This result  
202 was slightly smaller than that in a previous study ( $6.438 \times 10^{-5} \text{ K}^{-1}$ ; Touloukian et al.  
203 1975).

204

#### 205 **Density of nickel**

206 The measured density of Ni is plotted as a function of temperature in Fig. 5  
207 (Data is found in Supplemental<sup>1</sup> Table S2). The densities of two measurements (AK12  
208 and AK13) are reproducible. Previous results of Ni density are also plotted in Fig. 5.  
209 The density in this study was consistent with that of previous studies (Kohlhaas et al.,  
210 1967; Suh et al., 1988; Abdullaev et al., 2015) with an uncertainty of 0.2–0.5%, except  
211 for a result of Newkirk et al. (1958). All of these results indicate that there is no drastic  
212 decrease in density near  $T_m$ , suggesting that face-centered cubic (fcc) Ni does not have  
213 the premelting effect on density. This result is different from the density behavior of bct  
214 Sn.

215 The density decreases linearly with increasing temperature up to the  $T_m$  (1728

216 K). The  $\alpha$  in this study was determined using eq. (2) to  $6.18(6)\times 10^{-5} \text{ K}^{-1}$  in the  
217 temperature range of 1083–1713 K. The  $\alpha$  of the present and previous studies are listed  
218 in Table 1. The result of this study is consistent with the previous result ( $\alpha =$   
219  $6.07(8)\times 10^{-5} \text{ K}^{-1}$ ; Suh et al., 1988) within the error.

220

### 221 **Density of solid iron**

222 Densities of solid Fe are plotted as a function of temperature in Fig. 6a together  
223 with the result of liquid Fe. The present data is found in Supplemental<sup>1</sup> Table S3. The  
224 reproducibility of the measured density was confirmed from four measurements (AK15,  
225 AK16, AK18, and AK19) (see Fig. 6a). The two discontinuous density jumps observed  
226 at 1162(1) and 1666.0(5) K were derived from phase transitions of the  $\alpha$  phase  
227 (body-centered cubic (bcc))/ $\gamma$  phase (fcc) and  $\gamma$  phase (fcc)/ $\delta$  phase (bcc), respectively.  
228 Previously reported densities of solid Fe are also plotted in Fig. 6a. Our results are in  
229 good agreement with previous studies of  $\alpha$  and  $\gamma$  phases ( $\alpha$  phase: Straumanis and Kim,  
230 1969; Kohlhaas et al., 1967;  $\gamma$  phase: Onink et al., 1993; Basinski et al., 1955; Kohlhaas  
231 et al., 1967) within the error. In the stability field of the  $\delta$ -phase (1668–1803 K), the  
232 density decreases almost linearly with increasing temperature up to near  $T_m$  (1813 K)  
233 (see Fig. 6b). Thus, the density of solid Fe does not show a drastic decrease near  $T_m$ , i.e.,

234 the premelting effect. This suggests that Fe (bcc) shows a similar behavior with Ni (fcc),  
235 while it differs from the behavior of Sn (bct).

236 The thermal expansion coefficient ( $\alpha$ ) was determined in each phase of Fe by  
237 fitting with equation (2) (Table 1). The  $\alpha$  of the  $\alpha$ -phase was  $4.49(3) \times 10^{-5} \text{ K}^{-1}$  in the  
238 range of 823–1163 K, which is consistent with previous result of  $4.54(2) \times 10^{-5} \text{ K}^{-1}$   
239 (Straumanis and Kim, 1969). The  $\alpha$  of the  $\gamma$ -phase was  $6.37(5) \times 10^{-5} \text{ K}^{-1}$  in the range of  
240 1173–1673 K, which is consistent with  $\alpha = 6.40 \times 10^{-5} \text{ K}^{-1}$  (Komabayashi and Fei, 2010).  
241 The  $\alpha$  of the  $\delta$ -phase was  $7.02(9) \times 10^{-5} \text{ K}^{-1}$  in the range of 1675–1793 K, which is 35%  
242 larger than the result of Kohlhaas et al. (1967) ( $5.20(21) \times 10^{-5} \text{ K}^{-1}$ ). Because the stability  
243 field of the  $\delta$ -phase is a narrow temperature region at high temperature, the number of  
244 measured data points in previous studies was limited. By contrast, results of this study  
245 supplied more data points and a precise temperature dependence of the density because  
246 it was measured with much smaller temperature increments (1–10 K) (Fig. 6b).

247

## 248 **Density of liquid iron**

249 The density of liquid Fe is plotted as a function of temperature in Fig. 7  
250 together with the results of previous studies. The data of this study is found in  
251 Supplemental<sup>1</sup> Table S5. The reproducibility of the measured density was confirmed

252 from two measurements (AK18 and AK20). The present results are in good agreement  
253 with the results of Kirshenbaum and Cahill (1962), Lucas (1960), Brillo and Egry  
254 (2004), and Le Maux et al. (2019) within the error. Compared with the previous results,  
255 the present results give the density with a smaller error and were measured in the  
256 smallest temperature increments of 5 K (Fig. 7).

257 As shown in Fig. 7, the temperature dependence of the density of liquid Fe  
258 shows a concave shape. Thus, the  $\alpha$  can be well approximated as a linear function of  
259 temperature ( $T$ );  $\alpha = 1.37(10) \times 10^{-3} - (6.0(6) \times 10^{-7})T$  [ $\text{K}^{-1}$ ]. If the  $\alpha$  is assumed to be a  
260 constant, the average  $\alpha$  of AK18 and AK20 is  $2.42(1) \times 10^{-4} \text{ K}^{-1}$ . The  $\alpha$  of the present  
261 and previous studies are listed in Table 1. The temperature dependence of the  $\alpha$  of liquid  
262 metals was reported in previous studies (e.g., Kumai and Dass, 1993). The linear  
263 temperature dependence of  $\alpha$  shows a better fit to the measured density variation  
264 (coefficient of determination,  $R^2$ , is 0.996) than does the constant value of  $\alpha$  ( $R^2$  is  
265 0.988). The regression equation of  $\alpha$  as a function of temperature is valid, at least, in the  
266 temperature range of this study (1818–1998 K).

267

268

## IMPLICATIONS

269 The amount of light elements in the Earth's outer core has been estimated

270 based on a comparison between the density of the outer core from the seismic model  
271 and the EOS of liquid Fe alloys. A constant value of the  $\alpha$  of liquid Fe has been widely  
272 used in the EOS. However, in this study, the linear temperature dependence of  $\alpha$  could  
273 be more appropriate for the density variation of liquid Fe. The temperature dependence  
274 of  $\alpha$  is an important issue to describe the EOS of liquid Fe, and it also needs to be  
275 studied at high pressures.

276 In terms of the premelting effect, it is reported that the premelting effect is  
277 connected with rapid increase of defectively coordinated atoms or lattice defects based  
278 on numerical calculations (Delogu, 2006a; 2006b; Manai and Delogu, 2007; Martorell  
279 et al. 2013a). To evaluate the deviation from solid density,  $\rho/\rho_{solid}(T)$  is plotted as a  
280 function of temperature in Supplemental<sup>1</sup> Fig. S4, where  $\rho$  and  $\rho_{solid}(T)$  are measured  
281 density and solid density, respectively. The extrapolated value from the linear trend  
282 before the premelting is used for the value for  $\rho_{solid}(T)$ . The  $\rho/\rho_{solid}(T)$  decreases  
283 suddenly at 485–493 K before melting temperature of 505 K. This may indicate that the  
284 fraction of defect (melt) in solid Sn increases by the premelting effect. Alternate  
285 expression of melt fraction in solid Sn is given in Supplement<sup>1</sup> (III). Occurrence of the  
286 premelting effect in this study is found to be different depending on crystal structures.  
287 This is possibly due that the fraction of defect (melt) is characteristic of lattice geometry



288 (Delogu, 2006b). Further study is necessary to investigate occurrence of the premelting  
289 effect for various crystal structures of Fe-alloys at high pressures.

290

291

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375

## 376 Endnote:

377 Deposit item ---, Supplemental Material. Deposit items are free to all readers and found

378 on the MSA website, via the specific issue's Table of Contents.

379

380 **Figure captions**

381 Fig. 1 (a) High-temperature furnace and measurement set-up. (b) Heater and sample  
382 view from the top inside the chamber. The C/C heater has a slit for sample  
383 observation. The sample rod is 3 mm in diameter. Cross denotes the position  
384 of thermocouple junction.

385 Fig. 2 (a) Binarized image of solid Fe at 1473 K (AK16). Small alumina disk was  
386 inserted between the sample and large alumina plate. The sample bottom  
387 interface without alumina support was kept in flat even high temperatures  
388 (Fig. 2a). (b) Horizontal brightness profile of the area shown by dashed  
389 square in (a). Sample boundary was determined from the brightness profile.  
390 Diameter error corresponds difference between max. and min. diameter  
391 values.

392 Fig. 3 (a) Schematic image of liquid sample droplet for application to Pappus's  
393 centroid theorem. Shaded area (S) denotes a half area of the sample cross  
394 section divided by the rotating axis (y). R denotes the distance between the  
395 rotation axis (y) and center of gravity (G). (b) Raw image of liquid Fe at 1818  
396 K (AK20). (Inset) Gray pixels (1–6 pixels) at the top boundary of the sample.

397 Fig. 4 The effect of temperature on the density of solid Sn. Open and solid circles

398                    respectively denote the results of AK11 and AK14 of this study. The results of  
399                    two measurements are reproducible. Dashed line represents the trend reported  
400                    by Touloukian et al. (1975). Vertical dotted lines denote  $T/T_m=0.98$  (493 K)  
401                    and 1.0 (505 K).

402 Fig. 5            The effect of temperature on the density of solid Ni. Bold dotted and thin  
403                    dashed lines denote linear fit to the density data of this study and that of  
404                    Abdullaev et al. (2015), respectively. Vertical dotted line represents the  $T_m$   
405                    (1728 K). Since small alumina disk was not put beneath the sample in AK12,  
406                    height error of this run was relatively large, causing the larger density error  
407                    (see Supplemental<sup>1</sup> Table S4).

408 Fig. 6            (a) The effect of temperature on the density of solid and liquid Fe. Vertical  
409                    lines at 1162, 1666, and 1813 K denote the phase transitions of  $\alpha$  phase  
410                    (bcc)/ $\gamma$  phase (fcc),  $\gamma$  phase (fcc)/ $\delta$  phase (bcc), and  $\delta$  phase (bcc)/liquid,  
411                    respectively. (b) A zoom-in plot of high temperature part of  $\gamma$  and  $\delta$  phases  
412                    (1600–1820 K).

413 Fig. 7            The effect of temperature on the density of liquid Fe. Bold dotted curve  
414                    represents fit to the density data using a linear temperature dependence of  $\alpha$   
415                    ( $\alpha = 1.37(10) \times 10^{-3} - (6.0(6) \times 10^{-7})T$  [K<sup>-1</sup>]).



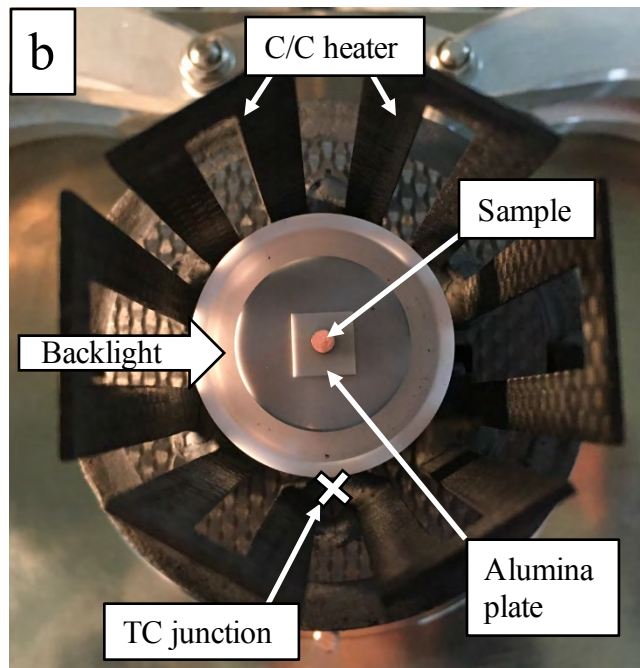
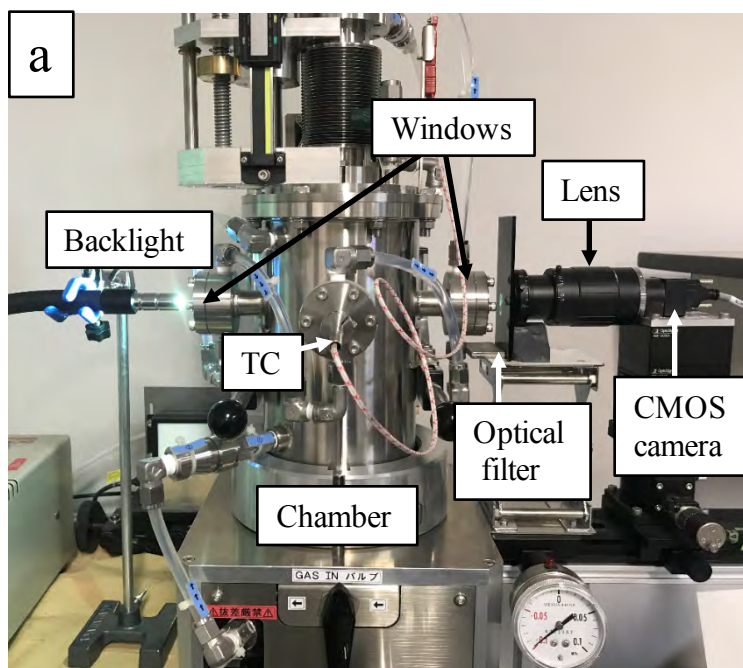


Figure 1

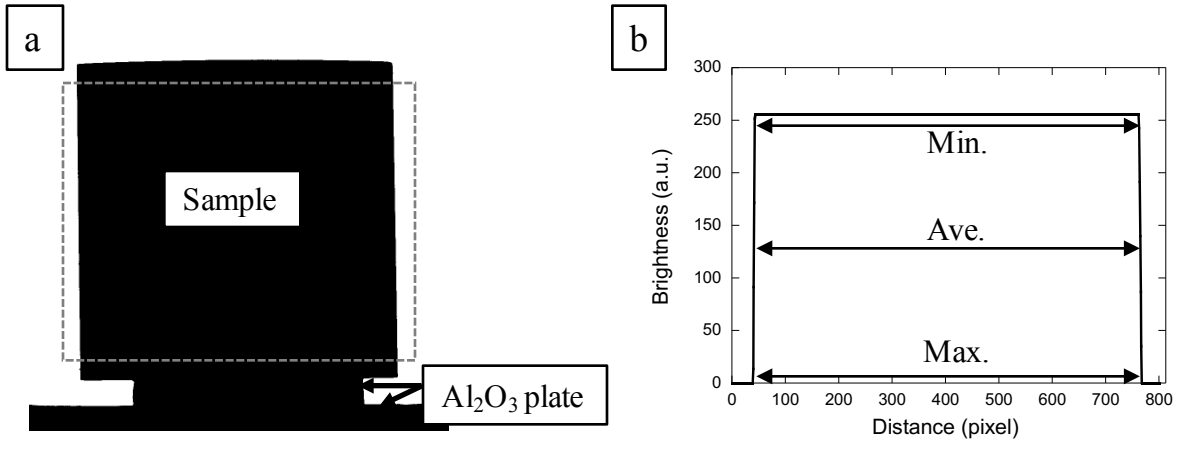


Figure 2

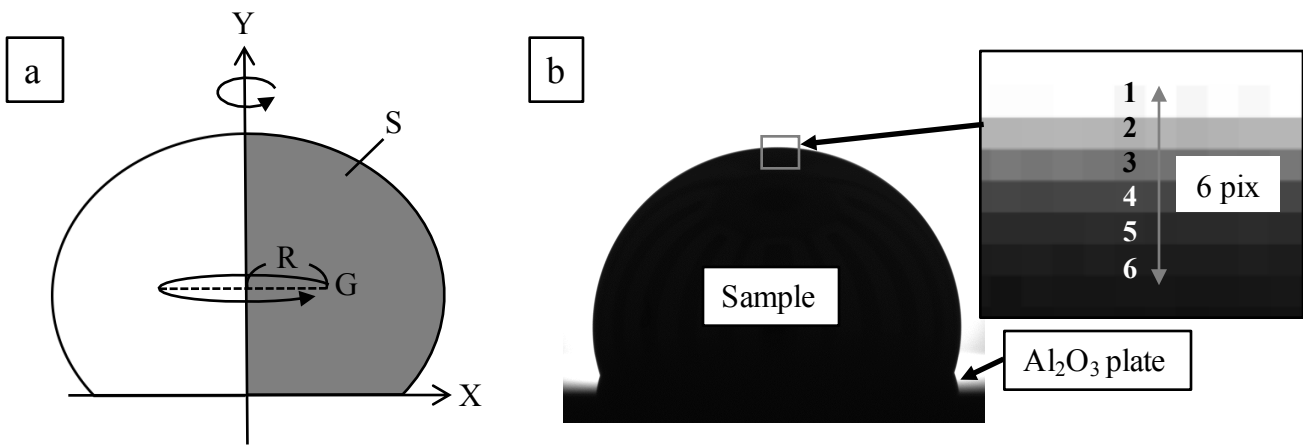


Figure 3

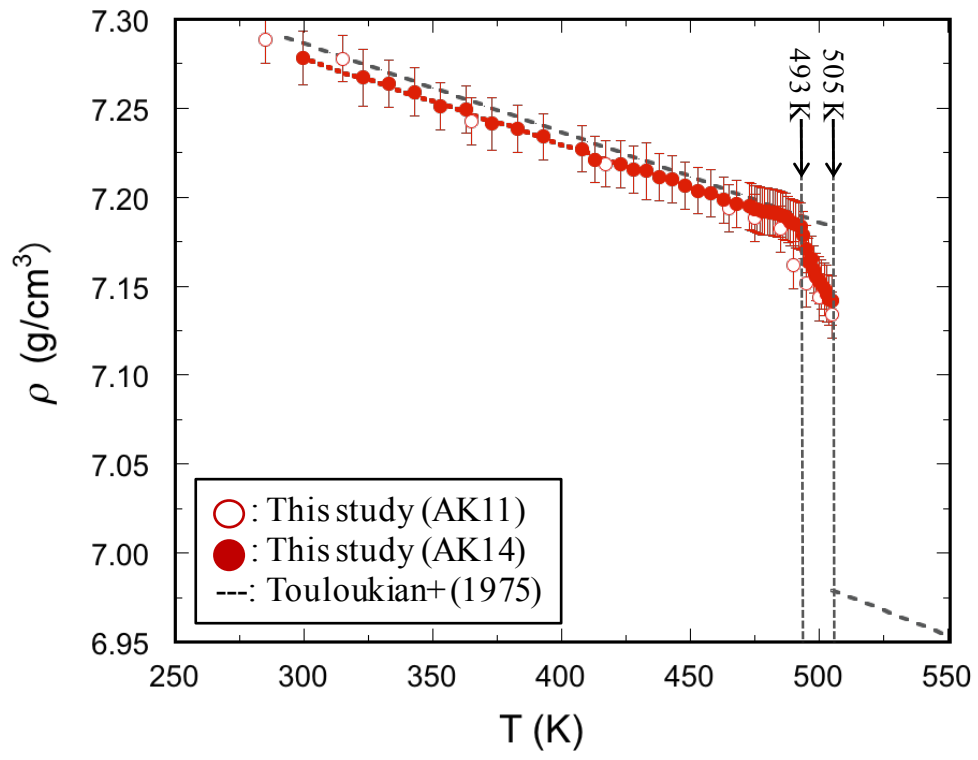


Figure 4

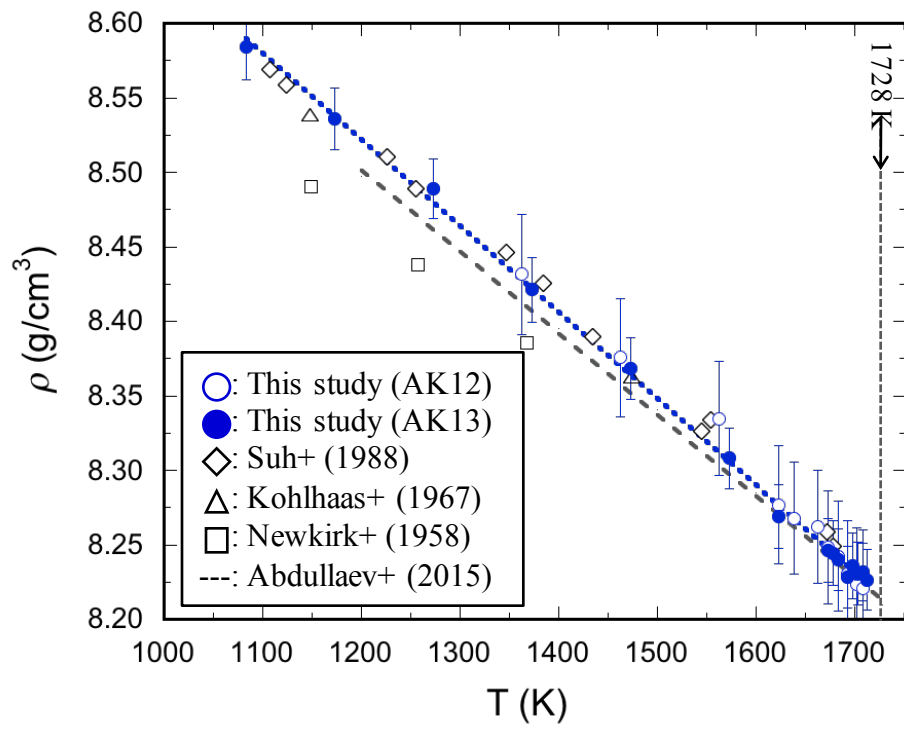


Figure 5

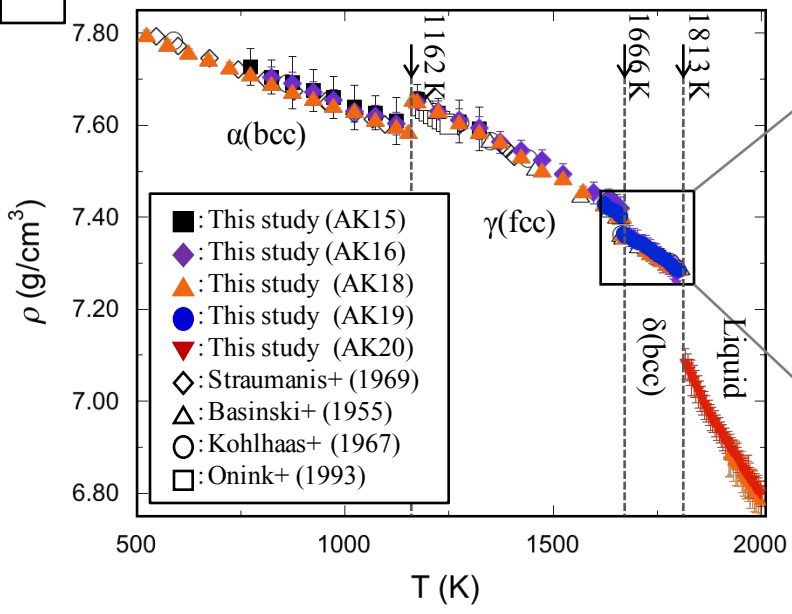
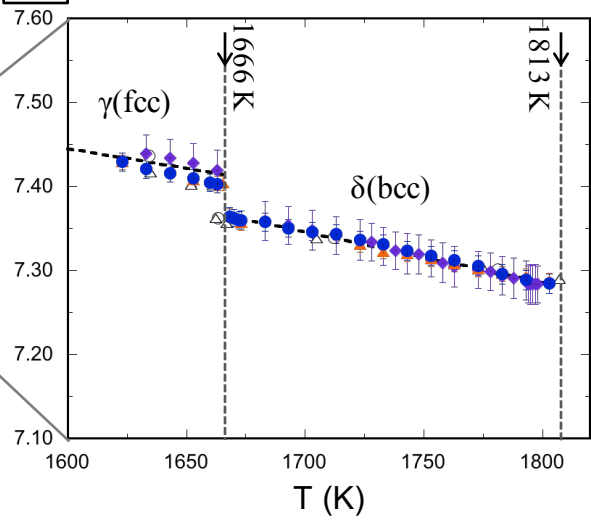
**a****b**

Figure 6

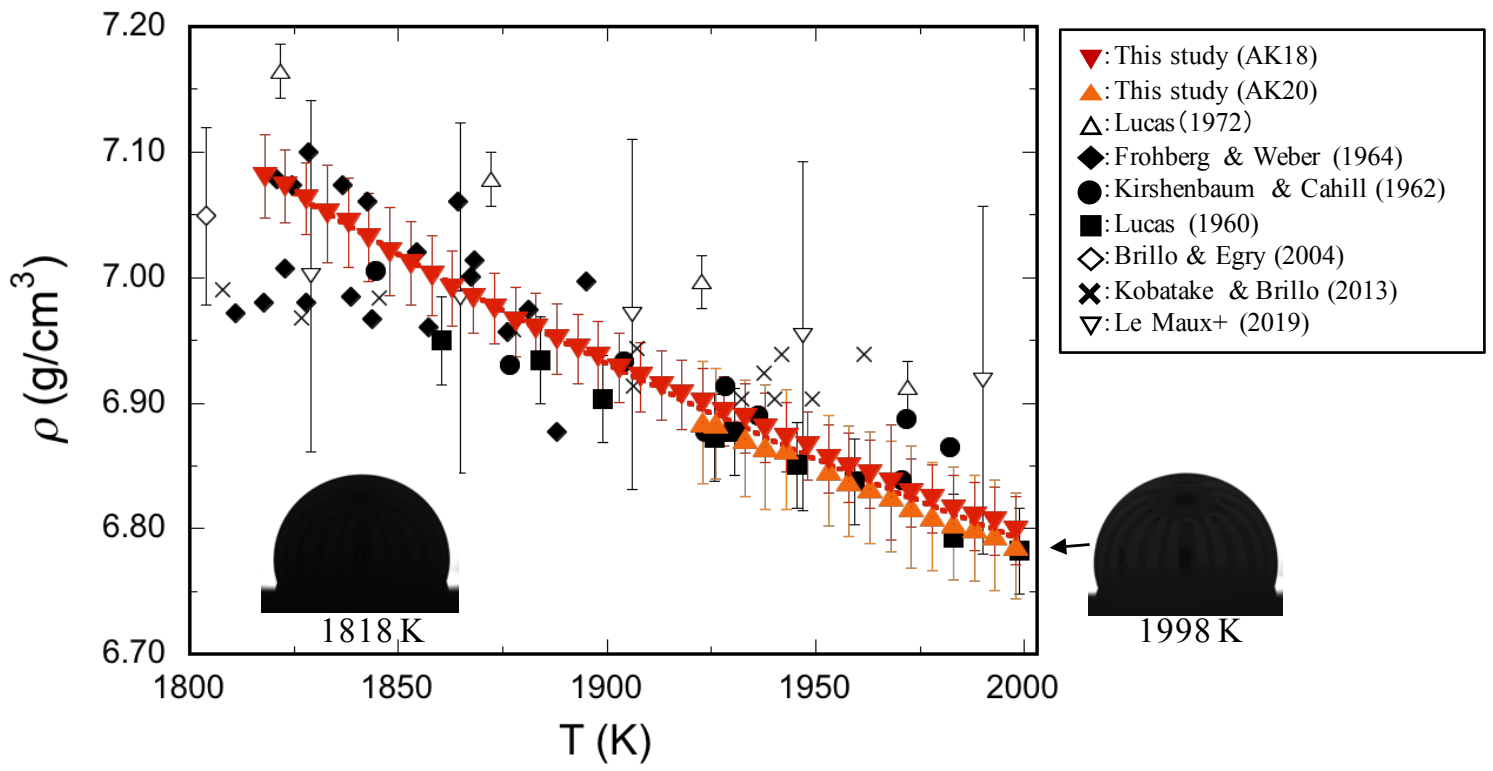


Figure 7

**Table 1. Thermal expansion coefficient ( $\alpha$ ) of solid and liquid metals in this study and previous studies.\***

Sample	phase	$\rho_0$ [g/cm <sup>3</sup> ]	$T_0$ [K]	T range [K]	a** [10 <sup>-5</sup> /K]	b** [10 <sup>-7</sup> /K]	Method	Reference
Sn	bct	7.278	300	285-490	6.22 (3)		Image	This study (AK11,14)
Ni	fcc	8.584	1083	1083-1713	6.18 (6)		Image	This study (AK12,13)
Fe	$\alpha$ -bcc	7.868	295	295-1153	4.49 (3)		Image	This study (AK15-18)
Fe	$\gamma$ -fcc	7.653	1173	1163-1665.5	6.37 (5)		Image	This study (AK15-18)
Fe	$\delta$ -bcc	7.365	1668	1668-1803	7.02 (9)		Image	This study (AK16-19)
Fe	liquid	7.081	1818	1818-1998	24.2 (1)		Image	This study (AK18,20)
Fe	liquid	7.081	1818	1818-1998	137 (10)	-6.0(6)	Image	This study (AK18,20)
Sn	bct	7.290	293		6.44			Touloukian et al. (1975)
Ni	fcc	8.626	976	976-1678	6.07 (8)		X-ray diff.	Suh et al. (1988)
Ni	fcc	8.497	1200	1200-1728	6.40 (6)		Gamma method	Abdullaev et al. (2015)
Fe	$\alpha$ -bcc	7.840	427	427-1023	4.54 (2)		X-ray diff.	Straumanis and Kim (1969)
Fe	$\gamma$ -fcc	7.533	1400	1400-1600	6.4(1)		X-ray diff.	Komabayashi and Fei (2010)
Fe	$\delta$ -bcc	7.362	1663	1663-1781	5.20 (21)		X-ray diff.	Kohlhaas et al. (1967)
Fe	liquid	7.164	1822	1822-1972	23.7 (9)		Archimedean	Lucas (1972)
Fe	liquid	6.95	1860	1860-2477	14.3 (5)		Archimedean	Lucas (1960)
Fe	liquid	7.005	1845	1845-2448	16.0 (2)		Archimedean	Kirshenbaum and Cahill (1962)
Fe	liquid	6.99	1808	1808-1961	7.9 (7)		Image	Kobatake and Brillo (2013)

\*Numbers in parenthesis indicate an error in last digit(s).

\*\* $\alpha$  is expressed using a linear function of T as  $\alpha=a+bT$