

Reviewer #1 (Remarks to the Author):

This manuscript by Kimura and others reported the melting point of the MgO, which is the second abundant mineral in the lower mantle, under high pressure condition by using experimental method.

The authors address important issue, but I feel that the current manuscript is too specific to be published in Nature Communications. I can recommend this paper for the publication only if the manuscript is substantially re-written with additional experimental data.

Weak implication

Based on the experimental data, the authors discussed about ULVZ, which is observed at core-mantle boundary region by seismology. Unfortunately, current manuscript does not provide any new insight about ULVZ. The melting temperature of MgO is higher than the previous LHDAC experiments and is consistent with theory. So what? How the expected nature of ULVZ or deep magma will be changed by the new data? Current manuscript made no effort even to extrapolate the melting temperature to CMB condition.

Experimental data

Experimental pressure ( $\sim < 30$  GPa) is far away from CMB condition (135 GPa). I would expect experimental pressure will be extended in the revised manuscript. Otherwise, numerous data are required to reduce the uncertainty of extrapolation.

Did authors obtain 1-D or 2-D mapping of the temperature distribution (e.g., Du and Lee 2014 GRL)?

The authors argue that the melting temperature may have large uncertainty when it was determined by the criteria using plateau in W-T curve. I agree with this point, but I can not be convinced by the scenario provided in the discussion part (line 64-81). I can not understand how the deformation and the plateau are correlated each other. Moreover, the deformation of the sample chamber is often observed at around  $< 30$  GPa. This is another reason why high-P data are necessary.

Reviewer #2 (Remarks to the Author):

The paper presents an experimental study of the melting curve of MgO up to 32 GPa, based on diamond anvil cell (DAC) and laser heating. The main result of the paper is a new estimate of the melting curve, which is much higher than that reported in a previous DAC study (Ref. 6), and agrees well with shock data and results obtained from theoretical calculations.

The authors also offer an explanation as to why the early results reported in Ref. 6 may have underestimated the MgO melting curve, which would be down to the incorrect identification of the onset of melting. The criterium used to locate the melting transition is the plateau in the temperature vs power curve: when the system melts the temperature stops increasing for a while, due to the latent heat of melting.

However, the authors observe two plateaus in their data. When they recover the sample after reaching the first plateau, located at the lower temperature, they find that the sample has expanded in the direction perpendicular to the axial compression, showing plastic flows. A TEM analysis of the recovered sample also showed stripe patterns, characteristic of shear deformation. The resulting thinning of the sample could have affected the efficiency of the heating, causing the plateau. On the other hand, the sample recovered after reaching the second plateau, at the higher temperature, showed the characteristics of a recrystallized sample, with Ar droplet inclusions which have

immiscibly dissolved into the liquid sample.

he data are convincing, and appear to resolve an outstanding issue with the melting behaviour of MgO. The authors also point out that there is a whole class of transition metals for which DAC experiments also appear to measure relatively low melting curves, or more specifically low melting slopes, and

they suggest that also these experiments may have been affected by wrong identification of the onset of melting. In fact, similar suggestions were recently proposed (Anzellini et al, 2013) for the melting curve of Iron, where large discrepancies existed between some early DAC experiments on one side (Boehler) and shock data (Holmes) and ab-initio calculations (Alfe` ) on the other. The paper by Anzellini et al. also based on DAC, reconciled these discrepancies. Iron of course is very important for the deep Earth, being the main component of the Earth's core. It might be more appropriate therefore to mention the iron story rather than the molybdenum one, or at least add a mention to the iron story.

The other comment that I would make is that it is somewhat far fetched to extrapolate the present results, up to 32 GPa, to the core mantle boundary pressure of 135 GPa, and so the authors may want to add some words of caution.

I would recommend publication of the work once the authors have addressed the above comments.

Reviewer #3 (Remarks to the Author):

Review of Article "Melting temperatures of MgO under high pressure by micro- texture analysis" by T. Kimura, H. Ohfuji, M. Nishi, and T. Irifune [Manuscript # NCOMMS-16-11983]

This paper investigates the melting behavior of MgO periclase to pressures of the upper portions of the lower mantle using the laser-heated diamond-anvil cell (LHDAC). The high-pressure melting curve of MgO has been contentious over the past couple of decades yielding disagreement within and between experimental and theoretical studies. When extrapolating to the core-mantle boundary (CMB), the differences between studies can be several thousands of degrees different! This is an important problem, but I'm not entirely convinced of the data even though the logic makes sense and the data appear fairly consistent with recent melting estimates of MgO from melting of ferropericlase (Mg,Fe)O.

The authors are using two methods to infer melting of MgO: laser power-temperature relationship and quenched micro-texture. While these are common ways that some studies have inferred melt, the methods have problems, as they show in their manuscript. Quenched texture may reflect solid-solid structural phase transitions or even "shear-induced plastic flow." Discontinuities in laser power vs. temperature profiles, used frequently to infer melting temperature, may also be caused by recrystallization or grain growth prior to melting. They did not mention, perhaps on purpose, that many think that the plateau in laser power vs. temperature may be due to latent heat, although this has been shown to not likely be the case (e.g., Geballe & Jeanloz, 2012).

Their experiments are similar to those run by (Zerr & Boehler, 1994) in which they use single-crystal MgO in an Ar pressure medium/thermal insulation and heating by CO2 laser. In the earlier study, Zerr & Boehler used the visual observation of a large increase in temperature to mark the onset of melting inferring that the melt could now absorb the laser energy more readily. The current study uses temperature plateaus and quenched texture. The initial test of the system of melting MgO at room pressure yielded a promising temperature in that it was very similar to literature values. More information is needed on this test. Was the melting done in air or under an inert atmosphere? Is there chemical information of the quenched sample (does the sample oxidize

or hydrate)? What does the texture look like?

Now moving on to the experiments at high pressures. In these experiments, the authors claim to observe two plateaus in the laser-power vs. temperature profiles: the first they infer to be when the Re gasket flows due to weakening of the material under high temperatures. A second plateau was observed some 1500-1700 degrees greater than the first plateau. This plateau, they claim, is when the sample melts. But in lines 79-81, they say "no clear change was observed" in the sample chamber or sample morphology. But I thought the premise of this study was to look at the micro-texture upon quench?

This takes me to Figure 3 in which back-scattered electron (BSE) images and TEM images are shown. BSE gives info on atomic number Z: typically brighter regions are represented by higher Z material thus giving a sense of composition. As such I'm confused on what I'm looking at in the BSE images. Why the color change in Figure 3i or between the crystals in HT? Shouldn't the color be the same throughout given it is only MgO? Or is there contamination from Ar? Water? Carbon (diamond)? Or something else? In Figure 3a,b,d, we see very different textures in HT and LT for what should be all be below the melting temperature. In Figure 3f,g,i, we see much different texture. Additionally, what is happening in the middle of Figure 3f, the region between "HT" and "LT"? The "chilled margins" make sense since that is likely self-insulation layers from thinning Ar layers, but there isn't such a feature in the other regions? Why not?

Why are the "HT" regions so different in extent? It looks like a factor of 2. Is this reasonable? What are the expected temperature gradients between the "HT" and "LT" regions? How long did the heating experiments last? Did they try to just heat to at high laser power, rather than ramping up to see if the texture was the same with melting, rather than an effect of just grain growth with time?

What do the electron diffraction images show? Is it MgO? Or something else?

Now on to thermal pressure... This is tricky. There have been several studies that have claimed that thermal pressures are negligible when using a soft pressure medium such as Ar (e.g., Fischer et al, 2013; Zerr & Boehler, 1994) and others that suggest that  $P_{th} \sim 0.5 \alpha K \Delta T$  (e.g., Goncharov et al., 2010), thus when  $\Delta T$  is large,  $P_{th}$  may also be large. In any case, when comparing to Zerr & Boehler, who didn't add  $P_{th}$ , the agreement becomes even worse since Zerr & Boehler do not include thermal P.

Almost as an aside, the authors make mention of the controversies in melting temperature in refractory metal by LHDAC experiments. The possibility that it is occurring due to "shear-induced anisotropic plastic flow" is an intriguing idea... But shouldn't this happen along the same P/T path for all samples since it would be dictated by the Re gasket? If this method proves to be reliable (I'm not yet convinced), this may be good to state it in the discussion, but not the abstract as it is off topic.

Figure 2b, c, d: How much time was the sample heated for between c and d? The temperature holds steady, but as the gasket got weak, the sample appear to expand. What is causing the browning of the sample? Did the diamonds burn (especially shown in part d)?

Figure 3: The agreement with Du et al. (2014) is rather striking, although the estimates of the melting temperature at the CMB are different by nearly 1000 K (8000 vs 8900 K). Why the discrepancy?

Figure S3: There appears to be a slight shift in the MgO peaks between the "HT" and "LT" regions. What do you attribute this to? Are the MgO (and Ar) volumes consistent for this pressure? What are the uncertainties? Why are the "HT" peaks broader?

Supplementary Table S2: I'm very puzzled at the relevance on the values given in this table. The average over such a large region (20  $\mu\text{m}$  x 20  $\mu\text{m}$ ) is problematic. I'd prefer to see a compositional map. Is the melt region enhanced in Ar or the other way around? How was Ar quantified? What were the "real" totals before normalization?

Minor problems:

Line 9: add "most" after, "Periclase (MgO) is the second..."

Line 10: remove "chemical". This is redundant.

Line 11: "mantle-core boundary" should be "core-mantle boundary" to go with standard convention.

Line 175: missing "nm" after "500 to 800"

I really want to like this paper, but unfortunately I am not convinced by the data shown.

1 **Response to review of the manuscript “Melting temperatures of MgO**  
2 **under high pressure by micro-texture analysis”**

3  
4 The reviewer’s comments are shown in blue and our responses in black color.

5  
6 **Response to Referee #1**

7 >This manuscript by Kimura and others reported the melting point of the MgO, which is the  
8 second abundant mineral in the lower mantle, under high pressure condition by using  
9 experimental method. The authors address important issue, but I feel that the current manuscript  
10 is too specific to be published in Nature Communications. I can recommend this paper for the  
11 publication only if the manuscript is substantially re-written with additional experimental data.

12  
13 We made efforts to expand our temperature and pressure range toward ~6000K and  
14 ~50GPa, respectively, which are certainly the limits in the current state of the art of the LHDAC  
15 technology combined with CO<sub>2</sub> laser heating, and extensively re-written the manuscript  
16 according to the referee’s recommendation (see below).

17  
18 >Based on the experimental data, the authors discussed about ULVZ, which is observed at  
19 core-mantle boundary region by seismology. Unfortunately, current manuscript does not  
20 provide any new insight about ULVZ. The melting temperature of MgO is higher than the  
21 previous LHDAC experiments and is consistent with theory. So what? How the expected nature  
22 of ULVZ or deep magma will be changed by the new data? Current manuscript made no effort  
23 even to extrapolate the melting temperate to CMB condition.

24  
25 As experimental scientists, we believe in experimental data rather than theoretical  
26 predictions; as for the melting temperatures of MgO, at high pressure, there have been only one  
27 report by Zerr and Boehler (1994) up to 30 GPa, for which we found a serious problem in their  
28 criteria for determination of melting temperatures. Determination of the melting temperatures  
29 by careful examination of micro-textures of the run products combined with the conventional  
30 method of using the temperature-laser power relations yielded substantially higher melting  
31 temperatures of MgO by about 1000K at ~30 GPa than those of the earlier study, which is really  
32 new and has significant implications for the determination of melting temperatures of other  
33 refractory materials, as discussed in the text. Moreover, we have just succeeded to expand the

34 pressure region to ~50 GPa, which further confirm our results in the earlier version of our paper,  
35 demonstrating that our experimental results should be used for the melting curve of MgO. It is  
36 fortune that our data are quite consistent with the theoretical predictions and dynamic  
37 compression data, but we believe carefully determined experimental data are most valuable in  
38 scientific research.

39 As for the implications of our new data, we added an important point in Discussion on the  
40 rheological properties of lower mantle materials, which have been constrained based on the  
41 homologous temperatures ( $T/T_m$ ) of the constituting major minerals of MgSiO<sub>3</sub> bridgmanite and  
42 MgO (line 168-182). We have extrapolated our melting temperatures to the bottom of the lower  
43 mantle, and discussed the expected melting relations and the composition of the ultra-deep  
44 magma to be formed near the mantle-core boundary, which are quite different from those based  
45 on the earlier melting experiments by Zerr and Boehler (1994) (see, line 151-167).

46

47 [Experimental data](#)

48 >Experimental pressure (~< 30 GPa) is far away from CMB condition (135 GPa). I would  
49 expect experimental pressure will be extended in the revised manuscript. Otherwise, numerous  
50 data are required to reduce the uncertainty of extrapolation.

51

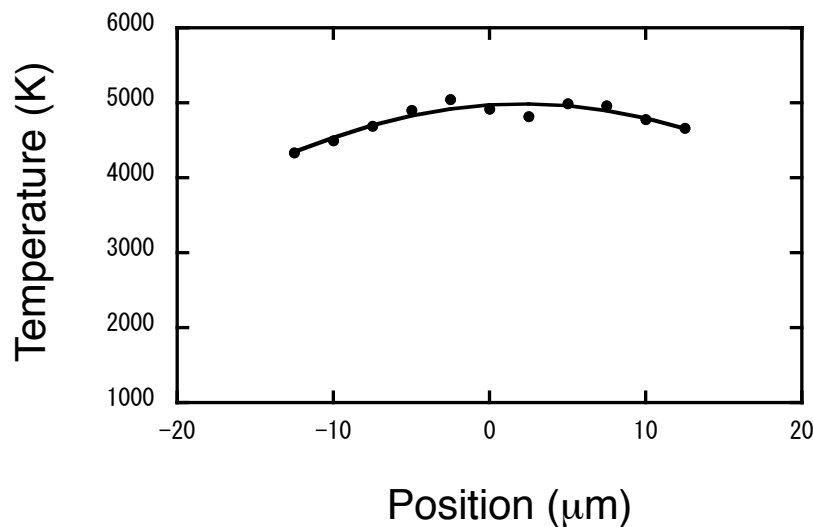
52 It is extremely challenging to determine the melting temperatures of such highly refractory  
53 materials as MgO under high pressure, which has a melting temperature as high as 3098 K even  
54 at the ambient pressure (Ref. 27). In addition to our new technique of the both-sided CO<sub>2</sub> laser  
55 heating system, which allows us to generate such high temperatures at high pressure, as shown  
56 in Supplementary Fig. 3 (Ref. 26), we adopted the triplet lens specially designed for correction  
57 of chromatic aberration (Edmund, 64838-L) as the objective lens. This technique allows us to  
58 accurately measure the temperature as shown in Fig. 1a and Supplementary Fig. 1, and we have  
59 eventually succeeded in determining the  $T_m$  up to ~50 GPa, which is substantially higher than  
60 those of earlier LHDAC experiments and more tightly constrains the melting curve of MgO as  
61 shown in Fig. 4 of the revised manuscript. We should point out that our new data that added in  
62 this figure are very consistent with our results for the experiments to ~30GPa, confirming our  
63 earlier conclusion on the steep gradient of the melting curve of MgO.

64

65 >Did authors obtain 1-D or 2-D mapping of the temperature distribution (e.g., Du and Lee 2014  
66 GRL)?

67

68 We added a typical example of the 1-D temperature profile as Supplementary Fig. 2 to the  
69 revised manuscript. The full width at half maximum (FWHM) of the heated region was  
70 approximately 60  $\mu\text{m}$ , which was estimated by fitting to the Gaussian function, as described in  
71 line 205-207. The temperature difference in the center of the heated area of  $2.5 \times 2.5 \mu\text{m}^2$  was  
72  $\sim 100$  K, obtained from the results of the 1-D profile, which is significantly less than the  
73 averaged-measurement error of  $\sim 200$  K.



74

75

76 **Supplementary Figure 2. Typical example of the radial temperature distribution in**  
77 **the MgO sample under heating at 36 GPa.** The plots and curve represent the  
78 measured temperature and the fitting by the Gaussian function.

79

80 >The authors argue that the melting temperature may have large uncertainty when it was  
81 determined by the criteria using plateau in W-T curve. I agree with this point, but I can not be  
82 convinced by the scenario provided in the discussion part (line 64-81). I can not understand how  
83 the deformation and the plateau are correlated each other. Moreover, the deformation of the  
84 sample chamber is often observed at around  $<30$  GPa. This is another reason why high-P data  
85 are necessary.

86

87 As for the question about the relation between the sample/gasket deformation and the  
88 temperature plateau, the expansion of the sample in the lateral direction (Fig. 2d), which was  
89 induced by the plastic flow of the gasket and gradually proceeded during heating at the first

90 temperature plateau (3500-3800 K), resulted in a considerable thinning of the sample and Ar  
91 pressure medium. The reduction in thickness of the sample chamber (along the compression  
92 axis of the DAC) must have had a significantly negative effect on the heating efficiency, as  
93 described in line 113-120.

94 We are not sure what the reviewer means by “the deformation of the sample chamber is  
95 often observed at around <30 GPa”, but what we are sure is that such an extensive deformation  
96 of the sample chamber is not observed commonly in LHDAC experiments, where the sample is  
97 heated to “moderate” temperatures (2000-2500 K), and so far, has not been reported anywhere  
98 as far as we know. In the additional melting experiments at higher pressures which we newly  
99 conducted, we also observed the deformation of the sample together with the Re gasket at the  
100 first plateau, as shown in Supplementary Fig. 3, suggesting that it is an essential phenomenon in  
101 the present high temperature heating.

102

### 103 **Response to Referee #2**

104 >The paper presents an experimental study of the melting curve of MgO up to 32 GPa, based on  
105 diamond anvil cell (DAC) and laser heating. The main result of the paper is a new estimate of  
106 the melting curve, which is much higher than that reported in a previous DAC study (Ref. 6),  
107 and agrees well with shock data and results obtained from theoretical calculations.

108 The authors also offer an explanation as to why the early results reported in Ref. 6 may have  
109 underestimated the MgO melting curve, which would be down to the incorrect identification of  
110 the onset of melting. The criterion used to locate the melting transition is the plateau in the  
111 temperature vs power curve: when the system melts the temperature stops increasing for a while,  
112 due to the latent heat of melting. However, the authors observe two plateaus in their data. When  
113 they recover the sample after reaching the first plateau, located at the lower temperature, they  
114 find that the sample has expanded in the direction perpendicular to the axial compression,  
115 showing plastic flows. A TEM analysis of the recovered sample also showed stripe patterns,  
116 characteristic of shear deformation. The resulting thinning of the sample could have affected  
117 the efficiency of the heating, causing the plateau. On the other hand, the sample recovered after  
118 reaching the second plateau, at the higher temperature, showed the characteristics of a  
119 recrystallized sample, with Ar droplet inclusions which have immiscibly dissolved into the  
120 liquid sample. The data are convincing, and appear to resolve an outstanding issue with the  
121 melting behaviour of MgO.

122



123 We thank the referee, who adequately addresses the important points in our paper.

124  
125 The authors also point out that there is a whole class of transition metals for which DAC  
126 experiments also appear to measure relatively low melting curves, or more specifically low  
127 melting slopes, and they suggest that also these experiments may have been affected by wrong  
128 identification of the onset of melting. In fact, similar suggestions were recently proposed  
129 (Anzellini et al, 2013) for the melting curve of Iron, where large discrepancies existed between  
130 some early DAC experiments on one side (Boehler) and shock data (Holmes) and ab-initio  
131 calculations (Alfe') on the other. The paper by Anzellini et al. also based on DAC, reconciled  
132 these discrepancies. Iron of course is very important for the deep Earth, being the main  
133 component of the Earth's core. It might be more appropriate therefore to mention the iron story  
134 rather than the molybdenum one, or at least add a mention to the iron story.

135  
136 We agree that the earlier work by Boehler on the melting temperatures of Fe could have  
137 been also underestimated because of a similar reason as discussed here. However, we note the  
138 first plateau occurs at temperatures between 3500-4000K, in the pressure range studied here.  
139 The melting temperatures of Fe determined by this author is significantly lower than these  
140 temperatures, and may not have directly relevance to the present phenomenon of plastic flow of  
141 Re gasket. Actually, a recent study using X-ray absorption spectroscopy coupled with DAC  
142 proposed lower melting temperatures, which is consistent with those of Boehler (G. Aquilanti et  
143 al., Proc. Natl. Acad. Sci. 2015). This is the reason why we limit our discussion on the  
144 refractory metals whose melting temperatures higher than ~3000K at the ambient condition.

145  
146 The other comment that I would make is that it is somewhat far fetched to extrapolate the  
147 present results, up to 32 GPa, to the core mantle boundary pressure of 135 GPa, and so the  
148 authors may want to add some words of caution.

149  
150 In response to the reviewer's comment, we have eventually determined the  $T_m$  up to ~50  
151 GPa, which is the highest pressure in the LHDAC experiments relevant to melting temperatures  
152 of MgO, as shown in the revised Fig. 4. The results well agree with the extrapolation based on  
153 our earlier runs up to ~30 GPa, and we are confident our melting temperature can be  
154 extrapolated to further higher pressures. Nevertheless, it is true that there is uncertainties of

155 ~1000 K in our estimated melting temperature (7900K) at the mantle-core boundary pressure,  
156 which is clearly stated in line 269-270.

157

158 I would recommend publication of the work once the authors have addressed the above  
159 comments.

160

161 We thank the referee for the great efforts to carefully check all the materials involved.

162

163 **Response to Referee #3**

164 >Review of Article "Melting temperatures of MgO under high pressure by micro- texture  
165 analysis" by T. Kimura, H. Ohfuji, M. Nishi, and T. Irifune [Manuscript #  
166 NCOMMS-16-11983]

167 >This paper investigates the melting behavior of MgO periclase to pressures of the upper  
168 portions of the lower mantle using the laser-heated diamond-anvil cell (LHDAC). The  
169 high-pressure melting curve of MgO has been contentious over the past couple of decades  
170 yielding disagreement within and between experimental and theoretical studies. When  
171 extrapolating to the core-mantle boundary (CMB), the differences between studies can be  
172 several thousands of degrees different! This is an important problem, but I'm not entirely  
173 convinced of the data even though the logic makes sense and the data appear fairly consistent  
174 with recent melting estimates of MgO from melting of ferropericlase (Mg,Fe)O.

175

176 >The authors are using two methods to infer melting of MgO: laser power-temperature  
177 relationship and quenched micro-texture. While these are common ways that some studies have  
178 inferred melt, the methods have problems, as they show in their manuscript. Quenched texture  
179 may reflect solid-solid structural phase transitions or even "shear-induced plastic flow."  
180 Discontinuities in laser power vs. temperature profiles, used frequently to infer melting  
181 temperature, may also be caused by recrystallization or grain growth prior to melting. They did  
182 not mention, perhaps on purpose, that many think that the plateau in laser power vs. temperature  
183 may be due to latent heat, although this has been shown to not likely be the case  
184 (e.g., Geballe & Jeanloz, 2012).

185

186 First, we should point out that the micro-texture analysis of the recovered sample,  
187 especially the one including the cross section, is not a common but the state of the art technique

188 for melting experiments using LHDAC, in spite of the referee's comment. In fact, this method  
189 has been adopted in only very few recent works (Refs. 9 and 21) for the determination of  
190 melting relations of multi-component systems, where the melting temperatures of the samples  
191 were estimated from a combination of micro-texture and chemical composition analyses.

192 The point in this study is that for accurate determination of the melting temperature of  
193 refractory, single-component material under high pressures, careful evaluation of melting  
194 criteria is essential and important, such as by combining the observation of plateau(s) in  
195 temperature-power relation with the micro-texture analysis of the quenched product. In the  
196 present study, we found two plateaus, the first plateau attributed to the sample deformation  
197 (flattening) initiated by the plastic flow of the Re gasket, and second plateau caused by melting  
198 of the sample. These are also supported by microtexture features shown by each quenched  
199 product: the former shows a mosaic texture formed by significantly deformed (elongated)  
200 crystals involving many dislocations and subgrain boundaries (Fig. 3a, d, and e), while the latter  
201 shows a typical quenching texture composed of chilled margins and columnar crystals grown  
202 inward from the rim (Fig. 3f and g). The presence of small Ar inclusions within the MgO  
203 crystals quenched from the second plateau (Fig. 3h and Supplementary Fig. 4) is also evidence  
204 for melting, where a part of Ar pressure medium was entrained as immiscible droplets in MgO  
205 rapidly crystallized from the melt, as described in the text (line 128-132). A drastic change of the  
206 heat transfer rate due to the convection of the MgO melt may be the cause for the second  
207 plateau, but further study is needed for detail.

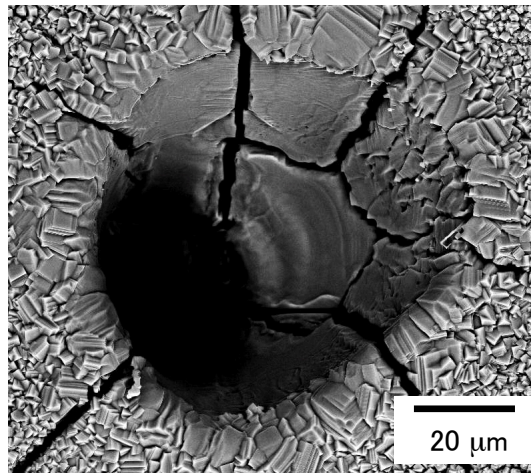
208

209 >Their experiments are similar to those run by (Zerr & Boehler, 1994) in which they use  
210 single-crystal MgO in an Ar pressure medium/thermal insulation and heating by CO2 laser. In  
211 the earlier study, Zerr & Boehler used the visual observation of a large increase in temperature  
212 to mark the onset of melting inferring that the melt could now absorb the laser energy more  
213 readily. The current study uses temperature plateaus and quenched texture. The initial test of the  
214 system of melting MgO at room pressure yielded a promising temperature in that it was very  
215 similar to literature values. More information is needed on this test. Was the melting done in air  
216 or under an inert atmosphere? Is there chemical information of the quenched sample (does the  
217 sample oxidize or hydrate)? What does the texture look like?

218

219 The melting test of MgO at ambient pressure was performed in the air using the same  
220 optical setting. SEM observation of the laser-heated spot of the sample quenched from ~3100 K,

221 where we clearly observed a plateau in the laser-power vs temperature profile, showed a crater  
222 and many cubic MgO crystals grown in the surroundings. Such euhedral MgO crystals most  
223 likely precipitated from the melt upon quenching. According to the reviewer's request, we  
224 performed quantitative analysis of the laser-heated and also surrounding areas by SEM-EDS  
225 and confirmed that they consist exclusively of pure MgO (Mg 38.85 wt%, O 60.59 wt%, Total  
226 99.44 wt%) and neither secondary phase(s) nor impurities were observed. We also checked with  
227 micro-Raman spectroscopy and detected no signs of secondary phase(s) such as brucite  
228  $\text{Mg}(\text{OH})_2$  detected in the areas, as described in our revised Supplementary Notes in line 66-69.



229  
230 **Figure I. Back-scattered electron image of the MgO sample quenched from ~3100 K at**  
231 **ambient pressure.**

232  
233 >Now moving on to the experiments at high pressures. In these experiments, the authors claim  
234 to observe two plateaus in the laser-power vs. temperature profiles: the first they infer to be  
235 when the Re gasket flows due to weakening of the material under high temperatures. A second  
236 plateau was observed some 1500-1700 degrees greater than the first plateau. This plateau, they  
237 claim, is when the sample melts. But in lines 79-81, they say "no clear change was observed" in  
238 the sample chamber or sample morphology. But I thought the premise of this study was to look  
239 at the micro-texture upon quench?

240  
241 There are indeed clear differences in microtexture between the samples quenched from the  
242 first and second plateaus, as shown in Fig. 3 (line 101-134). However, they can only be seen in  
243 the cross-sections of the recovered samples but are not clearly recognized for the sample after  
244 laser-heating in the DAC. This is a reason why our direct observation of the quenched samples  
245 on their cross-sections is important to judge melting in addition to the (second) temperature

246 plateau. However, to avoid any confusion, we revised the sentence to “Unlike the case of the  
247 first plateau, neither deformation of the sample itself nor the sample chamber was observed.”  
248 (line 99-100).

249

250 >This takes me to Figure 3 in which back-scattered electron (BSE) images and TEM images are  
251 shown. BSE gives info on atomic number Z: typically brighter regions are represented by higher  
252 Z material thus giving a sense of composition. As such I'm confused on what I'm looking at in  
253 the BSE images. Why the color change in Figure 3i or between the crystals in HT? Shouldn't the  
254 color be the same throughout given it is only MgO? Or is there contamination from Ar? Water?  
255 Carbon (diamond)? Or something else?

256

257       There seems to be a confusion here. The SEM images in Figure 3 show electron  
258 channeling contrast which is produced as a result of the interaction of energetic electrons with  
259 crystalline materials and is very sensitive to its crystallographic orientations (Newbury et al.,  
260 1986), as we mention in the method section. Thus, the variation in contrast in the images  
261 reflects the variety of crystallographic orientations of the MgO crystals in the product, which is  
262 helpful for us to recognize the individual grain size and shape. Images taken by using  
263 back-scattered electron (BSE) detector also have information on atomic number Z as the  
264 reviewer pointed out and are indeed widely used to discuss the chemical features of materials.  
265 In the present study, we used a relatively low accelerating voltage (5 kV) and high beam current  
266 (3 nA) to enhance the contribution of crystallographic orientation contrast. We also checked by  
267 SEM-EDS mapping analysis that there is neither chemical contamination (such as by Ar or C)  
268 nor impurities in the samples, although small Ar inclusions were found to be present within  
269 individual crystals and their boundaries by STEM-EDS.

270       To avoid such confusion, we modified the caption of Fig. 3 by replacing the phrase  
271 “Back-scattered electron images” with “Orientation contrast images”.

272

273 >In Figure 3a,b,d, we see very different textures in HT and LT for what should be all be below  
274 the melting temperature. In Figure 3f,g,i, we see much different texture. Additionally, what is  
275 happening in the middle of Figure 3f, the region between "HT" and "LT"? The "chilled  
276 margins" make sense since that is likely self-insulation layers from thinning Ar layers, but there  
277 isn't such a feature in the other regions? Why not?

278

279 >Why are the "HT" regions so different in extent? It looks like a factor of 2. Is this reasonable?  
280 What are the expected temperature gradients between the "HT" and "LT" regions? How long  
281 did the heating experiments last? Did they try to just heat to at high laser power, rather than  
282 ramping up to see if the texture was the same with melting, rather than an effect of just grain  
283 growth with time?

284

285 According to the temperature distribution measured across the hot spot (Supplementary Fig.  
286 2), the temperature in the LT region is estimated to be ~3000 K when the temperature in the HT  
287 region reached at ~5000 K. Since laser-heating at such high temperatures (which is needed for  
288 the melting of MgO) may lead to failure of the diamond anvil(s), the heating duration at each  
289 laser power had to be short (only for several seconds), then the total heating duration over the  
290 first and second plateaus is ~1 min each. However, it is clear from the microtexture that such a  
291 short-time heating is adequate to judge melting.

292 In the both samples quenched from the first and second plateaus, the microtexture of  
293 MgO is indeed quite different between the HT and LT regions. This is simply due to the large  
294 temperature gradient horizontally across the whole sample chamber, since the hot spot is much  
295 smaller than that of the sample hole (~ 120  $\mu\text{m}$ ), as shown in the 1-D temperature profile  
296 (Supplementary Fig. 2). The LT regions of the both samples (quenched from the first and  
297 second plateaus) consist of significantly deformed crystals due to the large shear stress induced  
298 by the gasket flow during heating, in which the temperature was, however, not high enough to  
299 promote the recrystallization and grain growth. More details are described in line 108-116.

300 On the other hand, the microtexture of the HT regions is quite different between the two  
301 samples. The sample quenched from the first plateau shows recrystallization and grain growth  
302 features of MgO, while that from the second plateau shows a typical quenching texture from the  
303 MgO melt. The latter is characterized by chilled margins and elongated crystals grown inward  
304 from the chilled margins (Fig. 3f and g). Originally, the chilled margins were continuous along  
305 the periphery of the lens-shaped melted region, but are now seen only at the lower and upper  
306 edges (Fig. 3f and g). The porous area in the middle of Fig. 3f is also a chilled margin composed  
307 of extremely fine grains of MgO, but it has not been well-polished due to drop-out of many  
308 grains, since this region was not sufficiently supported by the epoxy resin compared with the  
309 rim parts (that is directly in contact with the resin).

310

311 >What do the electron diffraction images show? Is it MgO? Or something else?

312

313 The electron diffraction patterns (shown as insets of Figs. 3c, e, h and j) were obtained  
314 from the center of the corresponding bright-field images using a selected-area aperture of a 1.4  
315  $\mu\text{m}$  diameter. All the observed diffraction spots (and discontinuous rings) are assigned to MgO,  
316 as indexed in the revised Fig. 3.

317

318 >Now on to thermal pressure... This is tricky. There have been several studies that have claimed  
319 that thermal pressures are negligible when using a soft pressure medium such as Ar (e.g.,  
320 Fischer et al, 2013; Zerr & Boehler, 1994) and others that suggest that  $P_{th} \sim 0.5 \alpha K \Delta T$   
321 (e.g., Goncharov et al., 2010), thus when  $\Delta T$  is large,  $P_{th}$  may also be large. In any case, when  
322 comparing to Zerr & Boehler, who didn't add  $P_{th}$ , the agreement becomes even worse  
323 since Zerr & Boehler do not include thermal P.

324

325 We believe the thermal pressure correction is necessary as discussed in line 240-242. The  
326 pressure increase by 6.6 ~ 9.0 GPa (20 ~ 70 %), depending on temperature, by this correction as  
327 shown in Supplementary Table 1. As is suggested by the referee, if the same correction is  
328 applied to the results of Zerr & Boehler, their melting curve becomes even shallower.

329

330 >Almost as an aside, the authors make mention of the controversies in melting temperature in  
331 refractory metal by LHDAC experiments. The possibility that it is occurring due to  
332 "shear-induced anisotropic plastic flow" is an intriguing idea... But shouldn't this happen along  
333 the same P/T path for all samples since it would be dictated by the Re gasket? If this method  
334 proves to be reliable (I'm not yet convinced), this may be good to state it in the discussion, but  
335 not the abstract as it is off topic.

336

337 It is clear from our observation that the deformation of the sample was caused by a large  
338 shear stress that is likely induced by the plastic flow of the Re gasket. Whether the deformation  
339 of the gasket occurs during laser-heating or not simply depends on whether the stress increases  
340 beyond the yield strength of Re. Thus, such an extensive gasket/sample deformation is not  
341 commonly observed in other LHDAC experiments heating at moderate temperature (below  
342 ~3000 K), but was observed in the present very-high-temperature heating experiments.  
343 However, the gasket deformation may not always result in sample deformation, since the plastic  
344 deformation of the sample also depends on its yield strength. In this sense, temperature plateau

345 (like the first plateau in this study) caused by sample deformation may be observed at  
346 lower/higher temperature or even not observed depending on materials. Choice of laser (e.g.  
347 CO<sub>2</sub> or YAG) as well as optic system which is directly linked to beam quality such as focused  
348 beam diameter may also influence the behavior.

349

350 >Figure 2b, c, d: How much time was the sample heated for between c and d? The temperature  
351 holds steady, but as the gasket got weak, the sample appear to expand. What is causing the  
352 browning of the sample? Did the diamonds burn (especially shown in part d)?

353

354 As described above, the heating duration at each laser power was several seconds and the  
355 total heating duration between c and d of the Fig. 2a was less than one minute. Therefore, the  
356 deformation of the sample together with Re gasket occurred gradually over the repeated heating,  
357 where the temperature stayed almost constant with increasing laser power (1<sup>st</sup> plateau). The  
358 origin of the brown color seen in the laser-heated center is unclear, but might be associated with  
359 the incorporation of Ar pressure medium into the surface of the MgO (particularly, at the grain  
360 boundaries, as shown in the TEM images (Fig. 3c)) during the recrystallization. The presence of  
361 Ar thin layers/droplets at grain boundaries of MgO crystals might cause a scattering of optical  
362 light particularly in a specific wavelength range. A further examination is necessary to clearly  
363 answer to the reviewer's point, but this is definitely not an easy task.

364

365 >Figure 3: The agreement with Du et al. (2014) is rather striking, although the estimates of the  
366 melting temperature at the CMB are different by nearly 1000 K (8000 vs 8900 K). Why the  
367 discrepancy?

368

369 Extrapolation from our revised melting curve gives a melting temperature of ~7900 K at  
370 the CMB condition, which is in very good agreement with that estimated by Du et al. (2014),  
371 although in general, some uncertainty is always associated with such a large extrapolation.

372

373 >Figure S3: There appears to be a slight shift in the MgO peaks between the "HT" and "LT"  
374 regions. What do you attribute this to? Are the MgO (and Ar) volumes consistent for this  
375 pressure? What are the uncertainties? Why are the "HT" peaks broader?

376



377 We calculated the pressures at the HT and LT regions from the diffraction patterns using  
 378 the equation of state (EoS) for MgO (Tange et al., 2009) and Ar (Jephcoat, 1998), as shown in  
 379 Table I. The pressure values calculated from the two EoS (for MgO and Ar) are almost  
 380 consistent with each other. The pressure difference between the HT and LT regions is as small  
 381 as ~1 GPa, which can be interpreted to reflect the subtle difference in the stress condition, as  
 382 can be guessed from the very contrasting microtextures between the two regions (line 121-134),  
 383 in addition to the pressure gradient in the sample chamber.

384

385 **Table I. Lattice constant of MgO ( $a_{MgO}$ ) and pressures at the HT and LT regions,**  
 386 **calculated from the equation of state for MgO ( $P_{MgO}$ ) and Ar ( $P_{Ar}$ ).  $\Delta P$  represents the**  
 387 **pressure difference between  $P_{MgO}$  and  $P_{Ar}$**

Region	$a_{MgO}$ (Å)	$P_{Ar}$ (GPa)	$P_{MgO}$ (GPa)	$\Delta P$ (GPa)
HT	4.05	22.5	23.8	1.3
LT	4.05	23.3	24.7	1.4

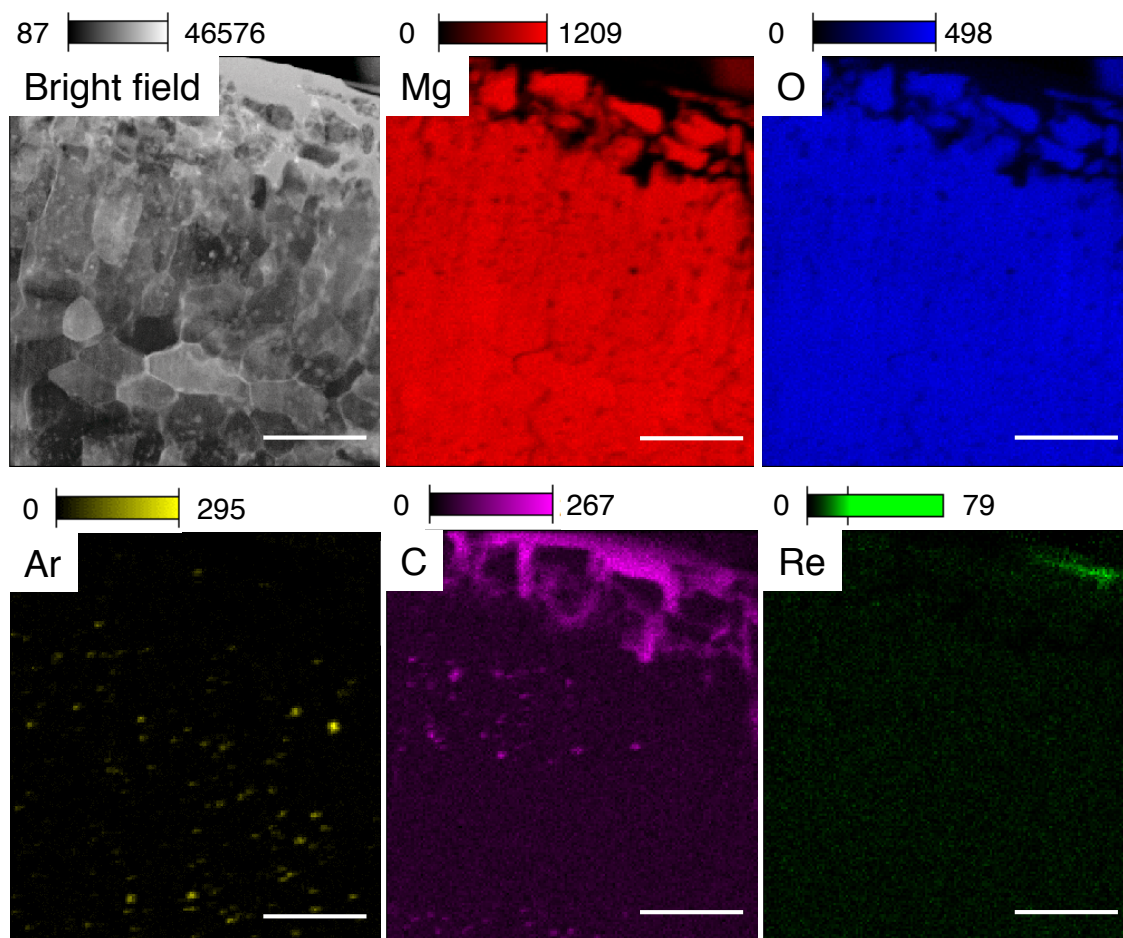
388

389 >Supplementary Table S2: I'm very puzzled at the relevance on the values given in this table.  
 390 The average over such a large region (20 um x 20 um) is problematic. I'd prefer to see a  
 391 compositional map. Is the melt region enhanced in Ar or the other way around? How  
 392 was Ar quantified? What were the "real" totals before normalization?

393

394 Unfortunately, the sample recovered from 32 GPa, 5200K (from which we obtained the  
 395 quantitative data) is not available any more for an additional SEM-EDS analysis, because it was  
 396 used to prepare cross-section foils by FIB for further examination by TEM (STEM-EDS).  
 397 Alternatively, we performed an elemental mapping on one of the cross-section foil by  
 398 STEM-EDS and found many small Ar inclusions within MgO crystals and at the grain  
 399 boundaries, as shown in Supplementary Fig. 4. The origin of the Ar droplets is described above.

400



401

402 **Supplementary Figure 4. Bright-field scanning TEM image and X-ray maps of Mg,**  
 403 **O, Ar, C, and Re collected from the laser-heated area of the sample recovered**  
 404 **from 32 GPa and 5200 K.** Scale bar represents 2  $\mu\text{m}$ . The X-ray maps identified that  
 405 the quenched sample consists of elongated and granular MgO crystals with many small  
 406 Ar inclusions. The concentrations of C and Re are artifacts, derived from the epoxy resin  
 407 and re-deposition from the sputtered Re gasket during Ar ion milling, respectively.

408

409

410 **Minor problems:**

411 **Line 9: add "most" after, "Periclase (MgO) is the second..."**

412 We added it in the sentence (line 9).

413

414 **Line 10: remove "chemical". This is redundant.**

415 We removed the word from the sentence (line 10).

416

417 Line 11: "mantle-core boundary" should be "core-mantle boundary" to go with standard  
418 convention.

419 We changed the words in the sentence (line 19).

420

421 Line 175: missing "nm" after "500 to 800"

422 We added it in the sentence (line 214).

Reviewer #1 (Remarks to the Author):

According to the comments raised by reviewers, authors substantially revised the manuscript with additional experimental data and geophysical implications. I'm sure that the manuscript is now more robust and attractive than previous.

However, I'm still not fully convinced by the explanation for lower melting T proposed by Boehler's group. In addition, newly provided T-profile raises one important question, which is not mentioned in the manuscript.

Authors argue that the "first plateau" is due to plastic flow of the sample, Ar medium and gasket, and such deformation can be recognized only at higher T. However, I can hardly see the significant deformation of the (MgFe)SiO<sub>3</sub> sample or gasket in Figure 2 by Zerr and Boehler (1993 Science), although it was heated at around 5000 K with Ar medium and (probably) Re gasket. Is this because of the (MgFe)SiO<sub>3</sub> sample? If so, only MgO is problematic?

Authors newly provided the T-profile during heating (Fig S2). T value are distributed  $\pm$  around 500 K as usual in the laser-heated DAC experiments. Which points were used for "temperature" in W-T curve (e.g., Fig 2)? This must be described with reasonable explanation, otherwise the temperature has systematic uncertainty of  $\pm$  around 500 K.

Reviewer #2 (Remarks to the Author):

The authors have addressed my comments, and I feel I can recommend the manuscript for publication.

Reviewer #3 (Remarks to the Author):

This is a re-review of this paper and I will first state that this manuscript is much better than the previous version I read. The additional data (e.g., at room P and higher pressures) as well as inclusion of the viscosity was much needed.

The techniques they use, the laser power vs. temperature plateaus and sample texture, are by themselves relatively common (the former more so than the latter), although the way that the authors implement them are novel. Determining that the first laser plateau occurs because of sample/gasket deformation, whereas the following plateau occurs because of melting.

I am conflicted though to recommend publishing in Nature Communications. How about a more specific Earth science journal (Nature Geoscience, GRL, EPSL)?

1 **Response to the review on our manuscript entitled “Melting**  
2 **temperatures of MgO under high pressure by micro-texture analysis”**

3  
4 Here are our responses (shown in black) to the reviewer’s comments (in blue).

5  
6 **Response to Referee #1**

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16 (1993 Science), although it was heated at around 5000 K with Ar medium and (probably) Re  
17 gasket. Is this because of the (MgFe)SiO<sub>3</sub> sample? If so, only MgO is problematic?

18  
19 In the melting experiments by Zerr and Boehler (1993) the details of the CO<sub>2</sub> laser optics  
20 (such as the beam diameter on the target and the temperature distribution in the sample under  
21 heating) as well as the detailed information of the gasket material used (Re?) and the sample  
22 chamber (e.g. thicknesses of the sample and Ar pressure transmitting media below and above)  
23 were not mentioned, despite that such information is essential for understanding what actually  
24 occurred during heating (at the temperature plateaus). Therefore, it may not be proper to directly  
25 compare their result on (Mg,Fe)SiO<sub>3</sub> melting with our result on MgO, since the result could  
26 potentially be influenced by such factors. Nevertheless, as the reviewer pointed out, in their  
27 experiment neither a significant deformation of (Mg,Fe)SiO<sub>3</sub> sample nor a plastic flow of the  
28 gasket likely have occurred as far as seeing the images of the sample chamber taken before and  
29 after the laser-heating (Fig. 2A and B of their paper). This may be due to a stress release  
30 associated with the phase transition from enstatite (initial starting material) to perovskite  
31 (bridgmanite) that must have occurred during heating. According to the equations of state of  
32 each phase (Stixrude and Lithgow-Bertelloni, 2005, Geophys. J. Int.) a significant volume  
33 reduction (~10% at 25 GPa and 300K) is expected through the phase transition, and this could

34 contribute to the relaxation of the thermal stress by laser-heating. Indeed, a signature of the  
35 abrupt volume change can be found in their figure (Fig. 2B of Zerr and Boehler, 1993), where  
36 large cracks (fracturings) were created in the laser-heated spots. Therefore, we suppose the  
37 significant volume change of the sample through the phase transition is a potential reason why  
38 the deformation of the sample/gasket was not observed in Zerr and Boehler's melting  
39 experiment on (Mg,Fe)SiO<sub>3</sub>.

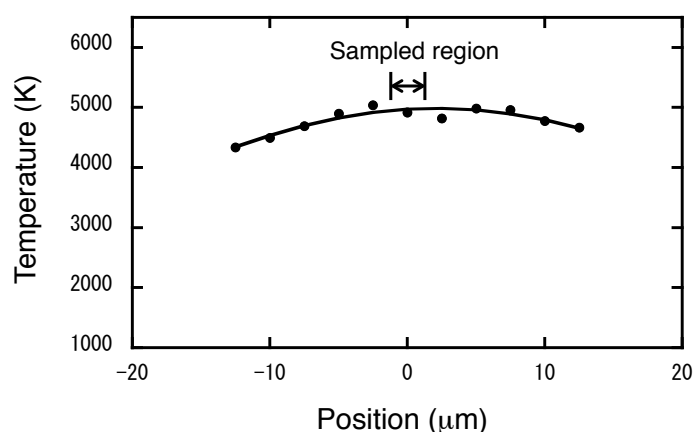
40

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42 500 K as usual in the laser-heated DAC experiments. Which points were used for “temperature”  
43 in W-T curve (e.g., Fig 2)? This must be described with reasonable explanation, otherwise the  
44 temperature has systematic uncertainty of ± around 500 K.

45

46 In response to the reviewer's comment, we have added a statement to explain how we  
47 determined the temperatures in the revised text (Line 216-219). The temperature at each laser  
48 power was determined by reading the peak value of the lateral temperature distribution profile,  
49 as shown in the revised figure (Suppl. Fig. 2). The temperature difference from the neighboring  
50 points is as small as ~100 K, which is substantially smaller than the averaged measurement  
51 error of ~200 K.

52



53

54 **Supplementary Figure 2. Typical example of the lateral temperature distribution in the**  
55 **MgO sample under heating at 36 GPa.** The plots and curve represent the temperatures  
56 measured near the hottest spot and the fitting by the Gaussian function, respectively.

57

58 **Response to Referee #2**

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60 publication.

61

62 **Response to Referee #3**

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65 pressures) as well as inclusion of the viscosity was much needed.

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71 because of melting.

72

73 >I am conflicted though to recommend publishing in Nature Communications. How  
74 about a more specific Earth science journal (Nature Geoscience, GRL, EPSL)?

75

76 As discussed in the last paragraph of the main text (Line 183-192), determination of  
77 the melting temperature of refractory metals such as Mo, Ta and W at high pressure by  
78 LHDAC experiments remains controversial, as is the case for MgO. The present study  
79 raises the possibility that shear-induced plastic deformation of the sample (together with  
80 the gasket) during high temperature heating is also responsible for the anomalously  
81 low-temperature melting curves of these metals determined by the earlier LHDAC  
82 experiments. Indeed, a significant anisotropic plastic flow of Ta (and potentially other  
83 refractory metals) before melting is theoretically predicted to cause the underestimation  
84 of the melting temperature determined from the plateau in the power vs temperature  
85 profile by LHDAC (Wu et al., 2009, Nature Material). The present report is probably  
86 the first experimental demonstration of such a novel idea and has profound implications  
87 to understand the real melting behavior of such refractory materials. Therefore, we  
88 believe the present work attracts the interest of a wide range of readers and deserves to  
89 be published in Nature communications.

Reviewer #1 (Remarks to the Author):

I'm satisfied with discussion about (Mg,Fe)SiO<sub>3</sub>. I hope it will be incorporated into manuscript. On the contrary, newly explained criterion of temperature looks problematic for me. I can recommend this manuscript for publication in the Nature Communications once authors answer this concern.

Authors newly explained that the "temperature" is taken at the peak of the temperature distribution. I'm strongly concerned that this overestimates the melting temperature. Their microscopic image showed that the melting pool is around 40 μm in diameter (Fig. 3). On the other hand, the temperature is about -500 K / 10 μm lower than peak temperature according to the T-distribution provided in the revised manuscript (Supplementary Fig. 2). At melting temperature, liquid and solid must be coexisting each other, and thus the liquid-solid surface should be at melting temperature. However, the liquid-solid surface is ~20 μm away from the center, where the "melting temperature" was obtained. If I assume T-gradient of -500 K / 10 μm and the diameter of melt pocket = 40 μm, actual melting temperature of MgO can be ~1000 K lower than their result at ~30 GPa.

I believe that the paper is more robust if authors demonstrate the comparison between T-distribution and microscopic image of the recovered sample, which is recently demonstrated by Japanese group using same apparatus (Fig. 4 of Ozawa et al., 2016 EPSL).



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10 answer this concern.

11  
12 We added the discussion on the melting of (Mg,Fe)SiO<sub>3</sub> perovskite by a previous study (Zerr  
13 and Bohler, 1993) as a comparison with the present case for MgO melting in the  
14 Supplementary Notes (line 65-77), according to the reviewer’s request.

15  
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27 T-distribution and microscopic image of the recovered sample, which is recently demonstrated  
28 by Japanese group using same apparatus (Fig. 4 of Ozawa et al., 2016 EPSL).

29  
30 We checked the radial temperature distribution profiles collected during heating at the second  
31 plateau and found that it became flat, while the profiles obtained at lower temperatures showed  
32 a standard Gaussian distribution, as shown in the revised Fig. 2b of our manuscript. Please note  
33 that the temperature-flattened region reached to ~40 μm, which is indeed equivalent to the

34 lateral dimension of the melting pool observed on the cross-section by SEM (Fig. 3f). Therefore,  
35 we don't think our melting temperatures were overestimated due to the lateral temperature  
36 gradient. The flattening in the profile is most likely due to a rapid increase in heat transfer  
37 caused by the convection of the MgO melt. We added the explanation in the revised manuscript  
38 (line 86-88 and 100-104).