

1           **Two-stage formation of pallasites and the evolution of their parent bodies**  
2   **revealed by deformation experiments**

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14  
15          **ABSTRACT**

16          Pallasites, stony-iron meteorites predominantly composed of olivine crystals and Fe-Ni metal,  
17          are samples of the interior of early solar system bodies and can thus provide valuable insights  
18          into the formation of terrestrial planets. However, pallasite origin is controversial, either  
19          sampling the core-mantle boundary or the shallower mantle of planetesimals that suffered an  
20          impact. We present high strain-rate deformation experiments with the model system olivine +  
21          FeS melt ± gold melt to investigate pallasite formation and the evolution of their parent bodies  
22          and compare the resulting microstructures to two samples of Seymchan pallasite. Our  
23          experiments reproduced the major textural features of pallasites including the different olivine  
24          shapes, olivine aggregates, and the distribution of the metal and sulfide phases. These results  
25          indicate that pallasites preserve evidence for a two-stage formation process including inefficient  
26          core-mantle differentiation and an impact causing disruption, metal melt injection, and fast

27 cooling within months to years. Olivine aggregates, important constituents of angular pallasites,  
28 are reinterpreted as samples of a partially differentiated mantle containing primordial metallic  
29 melt not stemming from the impactor. The long-term retention of more than 10 vol% of metal  
30 melt in a silicate mantle sampled by olivine aggregates indicates high effective percolation  
31 thresholds and inefficient metal-silicate differentiation in planetesimals not experiencing a  
32 magma ocean stage.

33

## 34 **1. Introduction**

35

36 Pallasites are stony-iron meteorites that are predominantly composed of large olivine crystals  
37 embedded in a matrix of iron-nickel and minor amounts of troilite (Fig. 1). Since their first  
38 description by Peter Simon Pallas in the late 18<sup>th</sup> century, researchers have aimed to explain the  
39 unusual composition and texture of pallasites, a challenge that can be summarized by three  
40 major questions:

- 41 1. Why is olivine the dominant and often singular silicate phase in pallasites?
- 42 2. Which process led to mixing between the olivine and the metal phase?
- 43 3. Why has the large density contrast between the olivines and the presumably molten  
44 metallo-sulphidic components not led to a subsequent gravitational separation?

45 The last question was termed “the pallasite problem” (Wahl, 1965) and received attention  
46 as a key constraint for the origin of pallasites, but plausible formation models must also address  
47 how the unusual phase mixture formed in the first place. Various models were proposed over  
48 time to explain pallasite formation and to reconcile cosmochemical, isotopic, magnetic, and  
49 textural data gained from pallasites (see Table 4 in Boesenberg et al. (2012) for a compilation).  
50 For the purpose of this study, we group previous models according to the internal or external  
51 origin of the metal phases. Most models belong to the former group that explains pallasites as  
52 part of a continuous differentiation sequence ranging from chondrites, acapulcoites, lodranites,

53 pallasites to iron meteorites/achondrites (Boesenberg et al., 2012; Ringwood, 1961). In these  
54 models both the metal and the silicates stem from a common chondritic precursor that formed  
55 a pallasite parent body (PPB). The pallasite problem is often addressed by positioning the rocks  
56 at the core-mantle boundary where a lack of gravitational segregation appears plausible  
57 (Boesenberg et al., 2012; Wasson and Choi, 2003). These regions have either retained part of  
58 the metal during inefficient differentiation (Boesenberg et al., 2012) or core-melt was re-  
59 intruded during a deformation event (Scott, 1977; Yang et al., 2010). The wide variety of  
60 cooling rates in the temperature interval between  $\approx 700\text{-}400$  °C determined in pallasites was  
61 linked to different burial depths and taken as evidence against a core-mantle origin for pallasites  
62 (Yang et al., 2010). This observation led to recent models of (i) a destructive impact forming a  
63 “pallasite planetesimal” (Yang et al., 2010) or (ii) of upwards diking of core-metal originating  
64 from an inward crystallizing core (Johnson et al., 2019). The dunitic composition of pallasites  
65 is either explained as an olivine cumulate layer (Buseck, 1977) or as restitic after high-grade  
66 fractional melt removal (Boesenberg et al., 2012).

67         The second, more recent, group of models assumes an external source of the metal phases  
68 that are injected into a dunitic middle to upper mantle of a PPB during the collision with an  
69 impactor (Bryson et al., 2015; Tarduno et al., 2012). The external-metal models were devised  
70 to explain remanent magnetization in olivine inclusions suggesting an active dynamo while the  
71 rocks cooled below the Curie temperature of  $\approx 360$  °C. This required a sufficient distance from  
72 a hot convecting core (Tarduno et al., 2012). Furthermore, the external-metal models also  
73 reconcile the low Ir content of the pallasite metal with the remanent magnetization. The former  
74 may suggest a highly evolved melt stemming from a largely crystallized core (e.g. Wasson and  
75 Choi, 2003), while the latter requires a largely molten convecting core. In the external models  
76 both constraints are fulfilled by the impactor core and the PPB core, respectively (Tarduno et  
77 al., 2012).

78            Since the different pallasite models often imply contradictory answers to the fundamental  
79 questions raised above, the use of pallasites as witnesses for the differentiation history of early  
80 solar system bodies is hitherto limited. The current study offers new insights into pallasite  
81 origins by using deformation experiments of olivine-metal at high temperature to explain the  
82 enigmatic textures of pallasites.

83

## 84 **1.2 Pallasite textures**

85

86 Pallasite textures denote the spatial distribution and shape of olivine, Fe-Ni metal and troilite  
87 in natural samples. When reporting the results of laboratory experiments below we use the term  
88 “microstructure” for easy discrimination.

89            *Olivine shapes.* The olivines are highly variable among different pallasites and have  
90 therefore received much attention as an important element in deciphering pallasite formation  
91 history (Boesenberg et al., 2012; Buseck, 1977; Ringwood, 1961; Scott, 1977; Solferino et al.,  
92 2015; Yang et al., 2010). In some pallasites, such as Brenham, olivines are well-rounded  
93 crystals, other pallasites such as Imilac, Admire, or Seymchan contain polygonal crystals  
94 (named angular olivine (Scott, 1977)) and smaller olivine fragments that are commonly  
95 surrounded by metal (Fig. 1). Angular pallasites also display cohesive aggregates with  
96 diameters up to 30 cm (Fig. 1) called olivine masses (Scott, 1977), olivine nodules (Ulff-Møller  
97 et al., 1998), or olivine clusters (Boesenberg et al., 2012). Throughout this work, we use the  
98 neutral term “olivine aggregates” for its descriptive connotation and apply it to all aggregates  
99 with two or more cohesive crystals.

100            The commonly accepted paradigm for the textural variation of olivines suggests that  
101 angular and fragmental olivines are produced by inter- and intragranular fracturing of olivine  
102 aggregates, respectively, caused by a deformation event also responsible for the mixing with  
103 metal components (Scott, 1977). Angular olivines have also been termed “euhedral” or

104 “anhedral” (Boesenberg et al., 2012), since their outer shape sometimes resemble crystal facets  
105 formed in silicate melt. However, since such “crystal facets” are commonly artifacts of former  
106 grain boundaries and triple junctions derived from the broken-up aggregates, such generic  
107 terminology is avoided here.

108       During high temperature annealing angular and fragmental olivines are expected to turn  
109 into the rounded shape, thereby approaching textural equilibrium determined by the high  
110 surface energy of the metal melt (Scott, 1977). Hence, round-type pallasites may conceal a  
111 deformation history (Scott, 1977; Solferino and Golabek, 2018). Rounding and grain-growth  
112 processes have been investigated by experimental studies that suggested a time between 100  
113 kyr and several Myr at experimental temperatures of 1100 to 1400 °C to create the large round  
114 olivine grains found in pallasites (Saiki et al., 2003; Solferino and Golabek, 2018; Solferino et  
115 al., 2015). All three olivine shapes—round olivine crystals, angular olivines, and smaller olivine  
116 fragments—are found in close proximity in some pallasites such as Seymchan. This observation  
117 needs to be reconciled with models to account for the olivine morphology since the rounding  
118 and growth process should first affect the smallest grains (e.g. Solferino et al., 2015).

119       *Fe-Ni metal and troilite textures.* In previous studies the textures of the metallo-sulphidic  
120 components have often been treated indirectly as the negative mould of olivine textures. For  
121 example, Boesenberg et al. (2012) considered several geometrical closed packing arrangements  
122 for olivine assigning the metal the role of filling the interstices, and in the Scott (1977) model  
123 metal acts as the passive infill of inter- and intragranular fractures creating the various olivine  
124 shapes. This implies a molten state of the metal phase when forming these textures, which  
125 appears to be commonly accepted by all authors, thus forming the basis for our experiments.

126       Compared to the dominant Fe-Ni phase, troilite (FeS) is of minor volumetric importance  
127 in pallasites (ca. 0.5 vol% in Esquel (Ulff-Møller et al., 1998) and 4-5 vol% in Brenham  
128 (Spinsby et al., 2008)). However, the textures associated with troilite deserve scrutiny, as it is  
129 frequently located in fractures that relate to the pallasite deformation history.

130 The intergrowth of kamacite and taenite lamellae in the form of Widmanstätten pattern or  
131 as plessite has been widely used to infer cooling rates for pallasites (Yang et al., 2010);  
132 however, in this study we do not consider these textural features as they form at temperatures  
133 well below the solidus of all phases and are therefore secondary.

134

## 135 **2. Materials and Methods**

136

137 The deformation experiments utilize the model system olivine with FeS ± gold (Au) melt to  
138 simulate the effect of high strain-rate deformation during an impact. The goal was to  
139 experimentally reproduce the range of pallasite textures summarized in the previous section and  
140 thereby better understand their formation. Experiments were performed using the multianvil  
141 press of the novel neutron instrument SAPHiR (Six Anvil Press for High Pressure Radiography  
142 and Diffraction) based at the FRM II neutron source of the Technical University Munich (TUM)  
143 (Fig. 2a). Additionally, three experiments were performed with the previously described Mavo  
144 press based at Bayerisches Geoinstitut (BGI) (Manthilake et al., 2012). Both the SAPHiR and  
145 the Mavo presses are identical three-axis multianvil apparatuses with a cubic sample geometry  
146 that allow controlled deformation experiments at high pressure and temperature conditions  
147 (Manthilake et al., 2012).

148 The samples were prepared using mechanically ground San Carlos olivine that was mixed  
149 with 20 wt% of synthetic FeS powder (ChemPur 99.9% purity). The powder was filled and  
150 mechanically compacted in un-welded cylindrical rhenium (Re) capsules measuring 3 mm  
151 across and 3 mm in height. Previous experimental studies revealed some reaction of FeS melt  
152 directly in contact with the capsule, however, it was shown that the capsule remained generally  
153 intact and the sample inside was uncontaminated (Cerantola et al., 2015; Walte et al., 2007,  
154 2011). In order to simulate the introduction of additional melt during deformation, several  
155 samples contained a small cavity in the centre filled with either pure FeS powder, Au powder,

156 or  $\approx 1 \times 1$  mm solid Au pieces (Fig. 2). Under the experimental conditions, Au and FeS form two  
157 immiscible melt phases. This allowed for easy discrimination in the recovered samples between  
158 melt that had already been situated in the olivine matrix (FeS) before deformation and the  
159 intrusion of ‘external’ melt (Au). The high surface tension of gold melt in contact with olivine  
160 (Walte et al., 2011) and slow heating ensured that the gold remained largely separated from the  
161 surrounding olivine–FeS aggregate during the initial heating and annealing stage and that it  
162 only intruded into the aggregate during deformation.

163 The cylindrical samples were placed into 12 mm cubic assemblies with pyrophyllite  
164 pressure medium and external gaskets (Fig. 2). A Re-foil resistance furnace was used for  
165 heating; the temperature was controlled by the heating power following an empirical ‘electric  
166 power vs. temperature’ calibration based on previous experiments. The absolute temperature  
167 error is estimated to be  $\pm 30$  K. No solid–melt phase separation was observed along the capsule  
168 axis in any of the experiments despite long static annealing times, indicating that thermal  
169 gradients had little effect on phase distribution and the microstructures. Further details on  
170 experimental technique and calibration have been described previously (Manthilake et al.,  
171 2012).

172 Heating to the target temperature (1300 or 1350 °C) commenced after reaching the target  
173 pressure of 1 GPa. Slow heating ensured the release of residual stresses from the compression  
174 before crossing the FeS solidus, thus preventing melt extrusion and furnace damage. After  
175 reaching the target temperatures, all samples were statically annealed at constant temperature  
176 before performing the deformation part of the experiments (table 1). A relatively long annealing  
177 time was chosen to attain a coarser grain-size and to ensure textural equilibrium of the olivine-  
178 FeS melt microstructure. Some experiments were heated up to 1350 °C in order to shorten the  
179 annealing time, yet, the sample deformation occurred after reducing the temperature to 1300  
180 °C.

181 After static annealing the samples were deformed either by plane strain (pure shear,  
182 abbreviated PS in table 1), extension followed by compression (E-C), or with an oblate strain  
183 field (OS). Pure shear and extension-compression deformation was achieved by shortening of  
184 one anvil axis and retracting the second anvil axis, while the third axis remained neutral. Oblate  
185 strain deformation was conducted by simultaneously retracting two anvil axes, which was  
186 accompanied by shortening of the third axis causing a reduction of the mean sample pressure  
187 during deformation. The use of different deformation geometries was necessary to simulate the  
188 whole range of pallasite textures as described in the results.

189 The predominantly brittle deformation textures of fragmental and angular pallasites  
190 suggest high strain-rates that may occur for example during an impact on the pallasite parent  
191 body. One experiment deformed at the highest sample shortening rate allowed by the press  
192 control software ( $\dot{\epsilon} \approx 6 \times 10^{-4} \text{ s}^{-1}$ , M 719), however, showed that these strain-rates are too low to  
193 reproduce pallasite-like microstructures (see section 3.1). Hence, in order to achieve strain-rates  
194  $> 1 \times 10^{-3} \text{ s}^{-1}$  the target positions of two anvil axes were manually altered in the control software  
195 by several increments of 20-100  $\mu\text{m}$  (corresponding to a compression or stretch of the samples  
196 by ca. 0.5 – 3.0 %) until attaining the targeted finite strain. Each increment is implemented by  
197 the hydraulic anvil positioning system within  $\approx 2$  seconds, which resulted in a series of short  
198 deformation steps in the sample with an instantaneous strain rate of up to  $1 \times 10^{-2} \text{ s}^{-1}$ . The  
199 resulting average strain-rate for the experiments, reported in table 1, was reconstructed from  
200 the accumulated anvil displacement and the total deformation time from the first to the last  
201 increment, which lasted for 20 to 60 s. The strain and the strain-rate values reported in table 1  
202 assumed that the displacement of the anvil axis is fully accompanied by a length-change of the  
203 samples. After reaching the target strain most samples were immediately quenched by shutting  
204 off the electric current resulting in a temperature drop below 300 °C within a few seconds. In  
205 order to investigate post-deformation annealing, two experiments (SA 178, SA 181) were held  
206 at constant temperatures of 1300 or 1350 °C, respectively, for two hours after deformation.



207 The recovered samples were cut with a diamond wire saw, embedded in epoxy, ground  
208 and polished. The deformed pure shear samples were sectioned in the  $x$ - $z$  plane of the strain  
209 ellipsoid, i.e. the section that contains the compression and the extension axis. The samples  
210 deformed by oblate strain were sectioned in the  $x$ - $y$  plane, thereby containing the two  
211 extensional axes with a normal orientation of the compression axis. Imaging of the samples was  
212 performed using a Zeiss reflected light microscope at TUM, and a LEO Gemini 1530 scanning  
213 electron microscope (SEM) at BGI, with an acceleration voltage of 20 or 30 kV and 4 nA beam  
214 current in secondary electron (SE) or backscattered electron mode (BSE).

215 To compare experimental results with natural pallasite textures, two representative  
216 samples of the Seymchan meteorite were investigated in detail: (i) a high-resolution image of a  
217 large slab of Seymchan (#5168) was provided by the American Museum of Natural History  
218 (New York City, NY, USA); and (ii) a freshly prepared slice of Seymchan (#SEY-18-01) was  
219 purchased from a commercial dealer (KD Meteorites) and investigated at TUM using Zeiss  
220 binoculars, Zeiss reflected light microscopy, and a CCD camera.

221 The metal fraction in natural olivine aggregates was estimated by determining the area  
222 occupied by metal pockets after binarization of optical photographs of polished sections via  
223 digital image analysis using the public domain software ImageJ, developed by the National  
224 Institutes of Health, USA. Binary images were attained by manual tracing of individual melt  
225 pockets; the resulting area fraction is considered to be approximately equal to the volume  
226 fraction. In order to gain an approximate value of the metal fraction of pallasites described in  
227 previous literature, a threshold value was set adequate to distinguish between the light grey of  
228 the metal and the darker colours of the olivine, if high quality images were available (e.g. the  
229 Seymchan slab shown in Fig. 2 of (Yang et al., 2010)).

230

### 231 **3. Results**

232

### 233 3.1 Static annealing and pure shear deformation experiments

234

235 After static annealing only, the FeS melt distribution is characterized by a range of  
236 different-sized melt pockets that display dihedral angles above  $60^\circ$  in accordance with previous  
237 work (Fig. 3a-b) (Minarik et al., 1996; Walte et al., 2007). Melt pockets that are small compared  
238 to the size of adjacent olivine grains are located at grain triple and quadruple junctions; they  
239 have an equant shape and commonly display convex boundaries with the olivines (Fig. 3a).  
240 Larger pools surrounded by a higher number of olivine grains are more irregularly shaped and  
241 the olivine–melt contacts are smoothly curved both concave and convex shaped (Fig. 3b). The  
242 bottom panels of figure 3 show details of the interior of olivine aggregates from Seymchan. The  
243 textures closely resemble the features produced by the static annealing experiments including  
244 the high dihedral angles, the curved olivine metal boundaries and the difference in shape of the  
245 smaller and larger metal pockets (Fig. 3c-d).

246 After shortening by 7 %, small veinlets originating from large melt pockets intrude olivine  
247 grain boundaries by intergranular fracturing (Fig. 4a). At higher strains (11% shortening),  
248 veinlets locally interconnect adjacent melt pockets and form a network surrounding isolated  
249 olivine crystals (Fig. 4b). Similar troilite (FeS) and Fe-Ni metal containing intergranular  
250 veinlets are often found inside Seymchan olivine aggregates either isolated or forming an  
251 interconnected network between metal pockets (Fig. 4d-e); locally, the olivine aggregates are  
252 also cross-cut by transgranular faults (Fig. 4f).

253 Pure shear deformation at higher strain (13–24 % shortening) causes pervasive melt-aided  
254 brittle deformation of the olivine – FeS melt matrix, which created olivine fragments of various  
255 sizes by intragranular fracturing (Fig. 4c). In high strain zones the deformation mechanism can  
256 be described as melt-aided cataclastic flow producing olivine fragments whose size decreases  
257 with increasing strain, while angular grains are produced in lower strain areas and close to larger  
258 melt pools that promote intergranular melt intrusion.

259 In order to investigate the role of strain-rate, one experiment (M 719) was deformed at a  
260 strain-rate of  $6 \times 10^{-4} \text{ s}^{-1}$ , nearly one order of magnitude lower than the other experiments. In this  
261 experiment, olivines display more internal deformation features such as lattice bending and  
262 formation of sub-grain boundaries. Strain is localized into FeS-filled shear zones that are either  
263 anastomosing around large olivines or locally cross-cut them (suppl. Fig. 1). These high strain  
264 zones often contain micron-sized roundish olivine grains that may have been formed by  
265 rounding of small fragments during the longer deformation duration. These brittle-ductile  
266 microstructures are not found in pallasites but resemble results of olivine-FeS melt samples  
267 deformed at similar strain-rates in a previous study (Walte et al., 2011). Hence, the dominantly  
268 brittle structures observed in pallasites suggest high strain rates  $> 1 \times 10^{-3} \text{ s}^{-1}$  supporting olivine  
269 – metal mixing caused by an impact (see Appendix A for further discussion).

270

### 271 **3.2 Extension-compression and oblate strain deformation experiments**

272

273 Although some pure shear compression experiments contained a central cavity filled with  
274 Au or FeS (table 1), the melt remained largely separated from the olivine-FeS aggregate up to  
275 the maximum shortening attained (24 %). In order to promote formation of the characteristic  
276 olivine–metal mush and to better understand the conditions that facilitate the mixing, several  
277 experiments were deformed either by pure shear extension followed by compression or by an  
278 oblate strain geometry as described in the methods section.

279 During extension-compression the central Au reservoir partially collapsed forming melt  
280 filled fractures that intruded and mixed with parts of the adjacent olivine aggregate. This locally  
281 created olivine–melt mushes with isolated olivine crystals and dislodged coherent olivine  
282 aggregates that resemble the olivine aggregates that are observed in pallasites (Fig. 5). In their  
283 interior, the experimental olivine aggregates preserve melt pockets that stem from the pre-  
284 deformation annealing stage and intergranular veinlets similar to the low strain microstructures

285 described in section 3.1. Hence, olivine aggregates preserve the older pre-deformation history  
286 of the experimental samples.

287 Samples deformed by oblate strain responded to the extension by the most efficient  
288 mixing of the Au melt with the adjacent olivine matrix observed in our experiments (suppl. Fig.  
289 2). However, rather than predominantly producing olivine fragments as during high-strain pure  
290 shearing, many of the olivines embedded in the Au matrix display an angular shape (Fig. 6a-  
291 b). For comparison different areas of Seymchan #5168 are shown that are either dominated by  
292 angular olivine or olivine fragments (Fig. 6c-d).

293 In order to investigate the role of pre-existing melt in the olivine matrix, one oblate strain  
294 experiment was performed with a sample containing FeS-melt free olivine surrounding the  
295 central Au-filled cavity (SA 176). In this case, the deformation resulted in the formation of large  
296 melt-filled fractures originating from the central cavity but did not form an olivine–melt mixture  
297 suggesting that pre-existing melt in the olivine matrix is important to facilitate matrix  
298 disintegration (suppl. Fig. 2).

299

### 300 **3.3 FeS–Au microstructures as analogues for troilite textures**

301

302 Areas of our deformed samples in which Au melt mingled with FeS melt revealed  
303 microstructures that closely resemble troilite textures found in our Seymchan samples (Fig. 7)  
304 and described previously (Buseck, 1977): (i) Smoothly curved metal-sulphide contacts and a  
305 preferable location of troilite in fractures and veinlets, and enclosing groups of small olivines.  
306 (ii) The bulging out of troilite from a fracture, forming a drop-like shape at the entrance, or  
307 drawing the surrounding Fe-Ni liquid into narrow fractures (Fig. 7a, d, see also Fig. 12 in  
308 Buseck (1977)). (iii) If fractures contain both liquids, Fe-Ni generally occupies the wider part  
309 and the phase boundary is generally convex towards troilite (Fig. 7b, e). We suggest that these

310 textures can be explained by the lower surface energy of troilite compared to Fe-Ni as discussed  
311 in section 4.1.

312

### 313 **3.4 Post-deformation annealing**

314

315 Indicators for annealing in angular pallasites such as microscopically rounded olivine  
316 edges (Buseck, 1977) and rounding of small olivine fragments (Scott, 1977) can be used to  
317 constrain the post-deformational thermal history of pallasites. Two exploratory experiments  
318 were conducted to investigate annealing processes. The first experiment (SA 178) reproduced  
319 the oblate strain experiment SA 177 but was annealed at 1300 °C for 2 h after deformation,  
320 while the second experiment (SA 181) was deformed and annealed at 1350 °C and only  
321 contained FeS as the melt phase. The first experiment revealed grain boundaries and healed  
322 fractures decorated with small FeS melt droplets and rounding of olivines with a diameter below  
323 10-15 micron (Fig. 8). Pinch-off of veinlets filled with high-dihedral-angle melt is known from  
324 previous experiments (Walte et al., 2011) and represents an additional indicator for annealing.  
325 The microstructure of the second higher temperature experiment displays an even greater  
326 textural equilibration with fully rounded olivines with diameters up to ca. 40 microns (suppl.  
327 Fig. 3). For comparison, veinlet pinch-off is rare in Seymchan and is only observed in some  
328 intragranular fractures with a diameter below ca. 10 µm (Fig. 8c). On the other hand, rounding  
329 is generally observed in small olivines with grain sizes below ca. 300 µm and occurs both in  
330 grains surrounded by troilite and Fe-Ni metal (Fig. 8c bottom). Here, we consider the troilite-  
331 enclosed grains, because the resulting grain sizes can be directly compared to the grain sizes of  
332 rounded olivine within FeS melt pockets of our experiments.

333 Based on experimental results and theoretical considerations, Saiki et al. (2003) suggested  
334 that the timing of olivine rounding is proportional to the cubed grain size. Hence, if grains with  
335 a diameter of ca. 10-15 µm are rounded after 2 h annealing at 1300 °C, we expect that olivines

336 with a diameter of 300  $\mu\text{m}$  would be rounded within less than ten years if annealing conditions  
337 were similar. At 1350  $^{\circ}\text{C}$  this rounding would only take about three months for grains  
338 surrounded by troilitic melt, which may be more realistic, since the temperature after  
339 deformation was probably higher than 1300  $^{\circ}\text{C}$ . While more systematic experimental studies  
340 employing a more precise temperature control are needed in the future, it is safe to suggest that  
341 cooling of the pallasites below the metal solidus temperature occurred on geologically short  
342 timescales.

343

## 344 **4. Discussion**

345

### 346 **4.1 Formation mechanisms of pallasite textures**

347

348 *Olivine textures.* The mechanisms to form angular and fragmental olivine by inter- and  
349 intragranular fracturing of olivine aggregates suggested by Scott (1977) was confirmed by our  
350 experiments. Our post-deformation annealing experiments also reproduced the subsequent  
351 olivine rounding, which may be fast if high temperatures persisted ( $\geq 1350$   $^{\circ}\text{C}$ ). This implies  
352 geologically very rapid cooling of angular and fragmental pallasites after deformation likely  
353 within months to years. If high temperatures persisted for longer periods of time, rounding may  
354 even be a viable mechanism to form the large rounded olivines in round-type pallasites  
355 suggested previously (Scott, 1977; Solferino and Golabek, 2018; Solferino et al., 2015).  
356 However, our experiments support previous suggestions that the large olivines with round  
357 boundaries found in Seymchan are relicts of the pre-deformation annealing stage rather than a  
358 result of post-deformational annealing (Fig. 5) (Yang et al., 2010; Scott, 2017).

359 *Olivine aggregate fragmentation.* Olivine aggregate disintegration and olivine–metal  
360 mixing is the signature process preserved in angular and fragmental pallasites such as Imilac  
361 and Admire and mixed-type pallasites such as Seymchan. Pallasite textures show that

362 disintegration is coupled with the influx of metal melt, which leads to a loss of cohesion and  
363 inflation of the olivine aggregates. Our experiments simulate the process by oblate strain  
364 deformation, and show that the previous existence of metal melt in the olivine aggregates and  
365 an additional external source of melt are the key elements of this process. The role of the  
366 dispersed metal pockets is likely to weaken the grain boundaries thereby favouring  
367 intergranular fractures over intragranular breaking of olivines. It follows that the metal pockets  
368 must have been liquid *before* aggregate inflation and further metal-melt influx, which supports  
369 the textural evidence for liquid metal pockets presented in section 3.1 and suggests that the  
370 impact was not the primary heat source. While angular olivines were formed by oblate strain  
371 deformation, olivine fragments were dominantly produced by high strain-rate pure shear  
372 deformation. We suggest that the latter simulates the initial deformation caused by an impact,  
373 while the former simulates extensional stress and a widespread disintegration of the host rocks  
374 driven by the subsequent intrusion of external metal melt from the impactor. The appearance  
375 of different pallasites would then be determined by the degree that these processes act at the  
376 particular location with respect to the impact site. For example, Seymchan slab #5168 (Fig. 1a)  
377 exhibits discrete areas that are dominated by angular olivines and areas dominated by smaller  
378 olivine fragments (Fig. 6c-d), which suggests sample-scale deformation localization into the  
379 latter areas that was followed by melt influx inflating both regions. Predominantly fragmental  
380 pallasites such as Admire and predominantly angular pallasites such as Esquel or Imilac may  
381 accordingly sample regional-scale deformation localization within the mantle volume affected  
382 by the impact.

383 *Metal pockets in olivine aggregates.* The olivine–metal pocket textures that are preserved  
384 inside olivine aggregates are characteristic of high-dihedral-angle melt-bearing systems  
385 undergoing static recrystallization and can be explained as follows: the occurrence of metal  
386 pockets of various sizes is energetically favourable for high surface energy melt that promotes  
387 an uneven melt distribution (Walte et al., 2007). The equant shape of small melt pockets is

388 determined by the resulting high dihedral angle (von Bargen and Waff, 1986), while the uneven  
389 shape of the large pockets is due to their lower surface to volume ratio, making them more  
390 susceptible to distortions during ongoing grain growth (Walte et al., 2003). Hence, olivine  
391 aggregates preserve a coarse-grained, equilibrated olivine plus metal melt texture, indicating  
392 long-term static grain growth at high temperature (Solferino and Golabek, 2018). In order to  
393 distinguish these metal pockets from the second generation of metal intruded during the  
394 collision we henceforth call them “primordial”.

395 *Troilite textures.* While sulphur is highly soluble in Fe-Ni melt, it does not fit into the  
396 crystalline structure of taenite and thus troilite (FeS) is thought to exsolve upon crystallization  
397 of the Fe-Ni-S liquid close to 1000 °C. Hence, troilite would only be present as a separate phase  
398 after the mixing event between the metal and the olivines, which differs from the experiments  
399 where FeS and Au are present as immiscible liquids from the start. However, the resulting  
400 textures are surprisingly similar between the experiments and pallasites (Fig. 7), which suggests  
401 that many troilite textures are melt-pseudomorphs as previously suggested by Buseck (1977).  
402 The textural details can then be understood by considering the lower surface energy of FeS  
403 compared to both Au liquid and crystallizing taenite. Hence, the lower wetting angle of FeS in  
404 contact with the olivine walls causes the observed bulging of the meniscus of Fe-Ni towards  
405 troilite in veinlets and explains that troilite often surrounds groups of small olivine grains and  
406 occupies confined spaces such as fractures and veinlets. These locations are all characterized  
407 by a high grain boundary area to volume ratio and are thus preferred sites for the lower surface  
408 energy liquid. Based on the structural continuity from narrow troilite-bearing fractures inside  
409 olivine aggregates to the breakup of the aggregates at their margins we suggest that most troilite  
410 textures record the final stage of a single deformation event rather than a later troilite  
411 mobilization episode as suggested previously (e.g. Ulf-Møller et al., 1998).

412

## 413 **4.2 Comparison of Seymchan with other main group pallasites**



414

415         Seymchan has an unusual classification history, being first classified as an iron meteorite  
416 (Scott and Wasson, 1976), later as an ungrouped pallasite (Wasson and Wang, 1986), and  
417 finally as a main group pallasite (van Niekerk et al., 2007). Seymchan shares many features  
418 with angular and fragmental pallasites such as the occurrence of olivine aggregates surrounded  
419 by mush-like regions containing olivine fragments and angular olivines in a metal matrix. The  
420 Seymchan olivine aggregates are characterized by a large amount of equilibrated metal-pockets,  
421 which is reflected in the description of Seymchan olivine aggregates as “Brenham-like” by  
422 Yang et al. (2010). The primordial metal fraction of our two Seymchan samples is 9-13 vol%  
423 (Fig. 1), and the olivine aggregates in the large Seymchan slab pictured in Fig. 2 of Yang et al.  
424 (2010) contain ca. 15 vol% primordial metal pockets, which appears to be the upper limit found  
425 in olivine aggregates of Seymchan. For comparison, Brenham and other round-olivine type  
426 pallasites contain a higher metal fraction of ca. 25-30 vol% (Buseck, 1977), setting them apart  
427 from Seymchan olivine aggregates, despite the resemblance of some olivine-metal contacts.  
428 Primordial metal content of olivine aggregates in other angular pallasites has not been  
429 systematically investigated, yet. In fact, to our knowledge a separate source of the metal melt  
430 pockets inside olivine aggregates has only been considered in one study that examined a large  
431 slab of pallasite Esquel (Ulff-Møller et al., 1998). Olivine aggregates in that sample contain up  
432 to 8 vol% primordial metal (Fig. 1 of Ulff-Møller et al. (1998)), which is similar in volume and  
433 appearance to our Seymchan sample #SEY-18-01 (Fig. 1b). A preliminary survey of available  
434 literature, collections, and online images of samples of other main group pallasites has shown  
435 that large olivine aggregates generally contain primordial metal pockets, including Finmarken  
436 (suppl. Fig. 4), Imilac (large slab exhibited in the “vault” on the 1<sup>st</sup> floor of the Natural History  
437 Museum, London), Admire (photographs of commercially available slices), Fukang (Figs. 1  
438 and 4 of Dellagiustina et al. (2019)), and Mount Vernon (e.g. Fig. 2 of Scott (1977)). We  
439 estimate a range of primordial metal of ca. 5-15 vol% that is preserved in olivine aggregates of

440 the various main group pallasites with Admire at the lower end of the spectrum, Esquel and  
441 Imilac in the middle of the range, and Seymchan at the top with the highest percentage of  
442 primordial metal. The general presence of pre-existing metal pockets in olivine aggregates of  
443 both angular and fragmental pallasites is compatible with our observations that a dispersed melt  
444 facilitates matrix breakup.

445

### 446 **4.3 Formation of pallasites and the evolution of a pallasite parent body**

447 Our results indicate that two generations of metal are present in main group pallasites  
448 providing new constraints on pallasite evolution. The first generation is preserved as primordial  
449 metal pockets in olivine aggregates and characterized by long-term static annealing and the  
450 second generation intruded during a short-lived deformation episode that was followed by  
451 freezing within months to years. The timescale of the pre-deformation annealing stage can  
452 currently not be quantified, but geological timescales are likely required for attaining cm-scale  
453 grains, since the high dihedral angle metal pockets slow down grain growth by a process similar  
454 to Zener pinning (Walte et al., 2007).

455 We present a new evolution model for a pallasite parent body (PPB) that is consistent with  
456 these results and previous constraints (Fig. 9): (a) Radiogenic heating of a chondritic PPB  
457 precursor by  $^{26}\text{Al}$  initially producing sulphur-rich melt crossing the Fe-S solidus ( $\approx 980\text{ }^\circ\text{C}$  (Brett  
458 and Bell, 1969)) and triggering the onset of differentiation (Fig. 9a). As the fraction of the newly  
459 formed melt crosses a percolation threshold (5–10 vol% (Bagdassarov et al., 2009; Walte et al.,  
460 2007; Yoshino et al., 2003)), it becomes mobile thus forming a small core. This stage could be  
461 represented by acapulcoite meteorites that show a mobilization of sulphide melt but remained  
462 below the silicate solidus (Floss, 2000; McCoy et al., 1996). (b) As heating continues, partial  
463 melting of silicates begins at ca. 1050–1100  $^\circ\text{C}$  (Mare et al., 2014). This stage in planetesimal  
464 evolution may be represented by lodranites, meteorites that are considered to have lost both

465 sulphide and silicate melt components (Floss, 2000; McCoy et al., 1997). Laboratory  
466 experiments have shown that the formation of silicate melt causes breakdown of metallo-  
467 sulphidic melt-networks, thereby halting further core-mantle differentiation (Cerantola et al.,  
468 2015), while allowing for gravity-driven percolation of buoyant silicate liquids (Connolly et al.,  
469 2009; Lichtenberg et al., 2019) thereby suggesting that sulphide loss generally predates silicate  
470 melt loss (Fig. 9b). Further heating causes temperatures to rise above or close to the Fe-Ni  
471 liquidus, which is accompanied by continuous fractional melting removing most non-olivine  
472 silicate components from the mantle (Boesenberg et al., 2012; Lichtenberg et al., 2019), while  
473 the metal melts remain trapped within the olivine matrix (Cerantola et al., 2015). (c) The high  
474 temperatures also facilitate static olivine grain growth (Solferino and Golabek, 2018; Solferino  
475 et al., 2015) and textural equilibration of the olivine-metal melt restites, thus creating the  
476 features preserved in olivine aggregates (Fig. 9c). (d) A PPB is disturbed by an impact of a  
477 differentiated body (Tarduno et al., 2012) that causes intense deformation closely followed by  
478 the injection of the impactor's residual core metal into the partially disintegrated mantle (Fig.  
479 9d). (e) Rapid cooling until reaching the Fe-Ni-S solidus preserves the veinlets and prevents  
480 rounding of large olivines, likely caused by exhumation in the aftermath of the impact (Fig. 9e).  
481 A viable way to achieve this is impact rebound, as demonstrated for impact events on  
482 planetesimals (Ciesla et al., 2013; Jutzi et al., 2013). Once the process stalls, pallasite material  
483 remains immobile and slower conductive cooling resumes, possibly slowed down by an  
484 insulating post-collision regolith layer (Tarduno et al., 2012) allowing for the formation of  
485 Widmanstätten patterns (Yang et al., 2010).

486 Previous pallasite formation models either focussed on fractional melting of chondritic  
487 material near the core-mantle boundary (Boesenberg et al., 2012), or considered an already  
488 completely differentiated upper mantle that was modified by an impactor (Bryson et al., 2015;  
489 Tarduno et al., 2012). While the former model also acknowledged texture alteration by impacts,  
490 it does not account for the initially rapid cooling indicated by the preserved veinlets and the

491 position in the shallower mantle during further cooling. On the other hand, the two-body  
492 scenario does not explain the olivine–metal melt equilibrium recorded in olivine aggregates.  
493 Our model presents a synthesis, including both a differentiation and an impact stage, with two  
494 separate generations of metal melt. Thus, it provides a simple explanation for the origin of the  
495 primordial melt pockets, since a uniform dispersion of high dihedral angle metal melt is difficult  
496 to achieve by other mechanisms (as our melt-free matrix deformation experiment illustrates –  
497 section 3.2).

498         A test of our two-stage model would be to compare the composition of small isolated  
499 primordial metal pockets with the outside metal in pallasites. If their origin is distinct, their  
500 composition may also differ (Ulff-Møller et al., 1998).

501         The planned NASA orbiter mission to the asteroid 16 Psyche may present a chance to  
502 directly investigate a PPB. The most recent density estimations for the asteroid suggested a  
503 metal fraction of 30 – 60 vol%, which is compatible with a pallasitic composition of Psyche  
504 rather than an exposed core as previously thought (Elkins-Tanton et al., 2020). If a PPB, the  
505 mission might be able to discriminate between the different models: The presence of a core  
506 overlain by a pallasitic mantle connected to impact structures or evidence for ferrovulcanism  
507 would support external-metal models including our hybrid model or the diking model of  
508 Johnson et al. (2019), respectively. The validation of pyroxene at the surface of Psyche  
509 (Drummond et al., 2018) would be also compatible with our mechanism of dunite –  
510 pyroxenite/basaltic differentiation of the PPB mantle. On the other hand, a gravitational  
511 homogenous pallasitic body would confirm the destructive impact model of Yang et al. (2010).  
512 Finally, internal-metal formation models that predict a layer of pallasite material at the core  
513 mantle boundary could be verified if the rocky mantle of Psyche has been removed.

514

#### 515 **4.4 Olivine aggregates as natural laboratories**

516

517 Olivine aggregates are natural samples of texturally equilibrated silicate–metal melt  
518 aggregates. As such, they present natural laboratories that can be applied to questions of  
519 planetary core-mantle differentiation and the distribution of metallic melt in silicate systems.  
520 Laboratory experiments have suggested metal melt percolation thresholds of ca. 5 vol%  
521 (Yoshino et al., 2003), that may be even further depressed to levels below 1-2 vol% by melt  
522 network hysteresis (Ghanbarzadeh et al., 2017) or by deformation (Bruhn et al., 2000), which  
523 would allow for efficient core-mantle differentiation without a magma ocean stage. The much  
524 higher primordial melt fraction of Seymchan olivine aggregates of ca. 9-15 vol% indicates the  
525 contrary, at least for the case of PPBs. We have demonstrated that these primordial melt pockets  
526 must have been molten for extended periods of time to promote grain growth and to allow for  
527 an efficient fragmentation to form the olivine–metal mixture preserved in pallasites. If these  
528 pockets were interconnected during that period of time, the metal melt would have  
529 gravitationally segregated due to the large grain sizes (Lichtenberg et al., 2019). However, such  
530 high effective percolation thresholds are compatible with our PPB formation model. Here,  
531 efficient metal melt percolation is initially hindered by production of silicate melt long before  
532 the Fe-Ni-S components are fully molten (Cerantola et al., 2015). Subsequently, the aggregates  
533 have sufficient time to undergo textural annealing as the temperature slowly rises towards the  
534 metal liquidus. Annealing causes pinch-off of the narrowest tubules in high-dihedral-angle  
535 networks increasing the percolation threshold and creating immobile melt pockets  
536 (Bagdassarov et al., 2009; Walte et al., 2007).

537

## 538 **5. Conclusions**

539

540 In the introduction we posed three questions on pallasite textures and argued that any  
541 model for their formation must provide plausible answers. We conclude with our answers for  
542 the reader to scrutinize:

543

544 *1. Why is olivine the dominant and often singular silicate phase in pallasites?*

545 Our experiments and observations do not deal directly with the petrological evolution of  
546 pallasites. However, our textural interpretations suggest high temperatures >1450 °C for an  
547 extended period likely induced by the decay of radiogenic isotopes such as <sup>26</sup>Al. This supports  
548 the model of Boesenberg et al. (2012) suggesting that pallasite host rocks are restitic dunites  
549 after high-grade fractional melting and silicate melt removal. We suggest that the mantle of the  
550 PPB was dominantly of dunitic composition with only minor pyroxenes, while possibly  
551 pyroxenitic and basaltic upper mantle and lithosphere layers remained un-sampled.

552

553 *2. Which process led to mixing between the olivine and the metal phase?*

554 Pallasite formation models can be grouped as internal and external according to the source for  
555 the metal. Our model is an internal-external hybrid; we suggest that 5-15 vol% of metal in  
556 angular pallasites have an internal origin remaining in the mantle after partial differentiation in  
557 the form of primordial melt pockets, while the rest of the metal (20-30 vol%) was externally  
558 derived from an impactor. The impact resulted in deformation of the mantle followed by  
559 expansion caused by the influx of core-metal from the impactor. The primordial melt pockets  
560 were instrumental for allowing pervasive intrusion of the external melt into the dunites  
561 producing the characteristic olivine–metal mixture.

562

563 *3. Why has the large density contrast between the olivines and the presumably molten metallo-*  
564 *sulphide components not led to a subsequent gravitational separation?*

565 The answer to the “pallasite problem” is twofold: (i) Primordial melt pockets in olivine  
566 aggregates prove that a moderate metal melt-fraction of up to 15 vol% can be retained in the  
567 mantle for long periods of time. (ii) After further metal melt injection from the impactor, rapid

568 cooling until reaching the Fe-Ni-S solidus prevented gravity-driven separation of metallic  
569 phases from the olivine.

570

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572

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582

## 583 **Appendix A Supplementary Materials**

584 Supplementary materials can be found online.

585

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694  
695

696 **Figure captions**

697

698 **Fig. 1.** Photographs of (a) Seymchan slab #5168 of the American Museum of Natural History,  
699 New York and of (b) Seymchan slice #SEY-18-01 with textural interpretations. Both samples  
700 display zones containing angular and fragmental olivines and olivine aggregates of various  
701 sizes. The metal pockets in the aggregates are interpreted to predate the surrounding metal–  
702 olivine mush zones, hence termed ‘primordial’ (see text for discussion).

703

704 **Fig. 2.** (a) Inside of the SAPHiR press. Six primary anvils transmit the force on a central stack  
705 of secondary anvils that contain the sample assembly (6-6 anvil geometry). (b) Sketch of the  
706 assembly used for the experiments. (c) Optical micrograph of a deformed sample with a central  
707 cavity filled with gold.

708

709 **Fig. 3.** (a, b) Reflected light micrographs of microstructures after 89.5 h static annealing. (c, d)  
710 Binocular images of equilibrated Fe-Ni metal pockets in Seymchan #SEY-18-01 indicating  
711 textural equilibrium.

712

713 **Fig. 4.** Pure shear deformation experiments (top, SEM backscatter images) compared to details  
714 of Seymchan olivine aggregates (bottom, optical micrographs). (a-c) Intergranular veinlets  
715 originating from melt-pockets after 7% and 11% vertical shortening, respectively, and  
716 pervasive cataclastic microstructure after 20 % shortening dominantly creating olivine  
717 fragments. (d, e) Veinlets interconnecting equilibrated metal pockets. (f) A fault (dashed orange  
718 line) cross-cuts an olivine aggregate. Fault-related olivine fragments ‘float’ in equilibrated  
719 metal pockets (black arrows) indicating that the metal was predominantly molten during the  
720 deformation.

721

722 **Fig. 5.** Olivine – metal mush in (a) experiments and (c) Seymchan showing adjacent round  
723 olivines (red arrows) and smaller olivine fragments. The former olivines are relicts of pre-  
724 deformation annealing. (b) Olivine aggregates in experiment (6.5 % extension followed by 13  
725 % shortening) and (d) nature. Note ‘primordial’ melt pockets (white arrows) that stem from  
726 pre-deformation annealing and veinlets produced during deformation (yellow arrow). All  
727 images are optical micrographs.

728

729 **Fig. 6.** Formation of fragmental and angular olivines by different strain geometries. (a) Pure  
730 shear deformation creates olivine fragments by predominantly intracrystalline fracturing. (b)  
731 Oblate strain (extension in the image plane) forms angular olivines by intercrystalline  
732 fracturing. (c-d) Olivine – metal mush zones in Seymchan are locally dominated by fragmental  
733 or angular olivines, respectively, suggesting deformation localization. The high metal fraction  
734 suggests that both regions subsequently underwent extension by metal influx.

735

736 **Fig. 7.** Optical micrographs of Au – FeS melt microstructures in deformed samples (top)  
737 compared to Fe-Ni – troilite textures in Seymchan pallasite (bottom). These textures are best  
738 explained by liquid immiscibility (see text for discussion).

739

740 **Fig. 8.** Optical micrographs of (a-b) experimental microstructures and (c) Seymchan textures.  
741 (a) Oblate strain inter- and intragranular fracturing in the olivine – FeS melt matrix (top) and  
742 angular olivine – Au mixture (bottom). (b) 2 h annealing after deformation causes pinch-off of  
743 most FeS melt veinlets and creates round olivines in the mush zone. (c) Seymchan textures  
744 indicating annealing. While intergranular veinlets are generally continuous in Seymchan, some  
745 narrow intragranular fractures have pinched-off (top). The smallest olivine grains in Seymchan  
746 generally display a rounded shape (bottom).

747

748 **Fig. 9.** Schematic diagram for the evolution of a PPB and the concurrent texture evolution  
749 (magnifications). Colours of magnifications are chosen in order to be comparable with the  
750 textural mapping in Fig. 1. (a) Formation of sulphur-rich melts in a chondritic precursor body  
751 and partial core differentiation via percolation (wavy arrows). (b) Crossing of the silicate  
752 solidus traps remaining metal melt in the matrix while partial melting of the silicates is  
753 accompanied by silicate melt ascent (wavy arrows). (c) Completion of partial differentiation  
754 without a magma-ocean stage leaves a mantle largely consisting of olivine (+/- orthopyroxene)  
755 plus remaining primordial metal melt. (d) An impact causes deformation of the mantle (top  
756 magnification) closely followed by intrusion of the impactor's core melt (bottom  
757 magnification). (e) The impact rebound causes freezing of the metal melt followed by slow  
758 conductive cooling. Credit: Reiner Müller.