

Dual origin of ferropericlasite inclusions within super-deep diamonds

Sofia Lorenzon^{a*}, Michelle Wenz^b, Paolo Nimis^a, Steven D. Jacobsen^b, Leonardo Pasqualetto^a, Martha G. Pamato^a, Davide Novella^a, Dongzhou Zhang^c, Chiara Anzolini^d, Margo Regier^d, Thomas Stachel^d, D. Graham Pearson^d, Jeffrey W. Harris^e and Fabrizio Nestola^a

^aDepartment of Geosciences, University of Padua, Via G. Gradenigo 6, 35131 Padua, Italy
(sofia.lorenzoni@phd.unipd.it, paolo.nimis@unipd.it, leonardo.pasqualetto@phd.unipd.it,
martha.pamato@unipd.it, davide.novella@unipd.it, fabrizio.nestola@unipd.it)

^bDepartment of Earth and Planetary Sciences, Northwestern University, 2145 Sheridan Rd. 60208, Evanston, Illinois (USA) (michellewenz2020@u.northwestern.edu, s-jacobsen@northwestern.edu)

^cHawaii Institute of Geophysics and Planetology, University of Hawaii, 1680 East-West Road, Honolulu, HI 96822, Hawaii (USA) (dzhang@hawaii.edu)

^dDepartment of Earth and Atmospheric Sciences, University of Alberta, AB T6G2E3 Edmonton, Canada (anzolini@ualberta.ca, mregier@carnegiescience.edu, thomas.stachel@ualberta.ca,
graham.pearson@ualberta.ca)

^eSchool of Geographical and Earth Sciences, University of Glasgow, G12 8QQ Glasgow, United Kingdom (Jeff.Harris@glasgow.ac.uk)

* Corresponding author: Sofia Lorenzon (sofia.lorenzoni@phd.unipd.it)

Department of Geosciences, University of Padua,

Via G. Gradenigo 6, 35131 Padua, Italy.

23 Abstract

24 Ferropericlasite [(Mg,Fe)O] is one of the major constituents of Earth's lower mantle and the most
 25 abundant mineral inclusion in sub-lithospheric diamonds. Although a lower mantle origin for
 26 ferropericlasite inclusions has often been suggested, some studies have proposed that many of these
 27 inclusions may instead form at much shallower depths, in the deep upper mantle or transition zone.
 28 No straightforward method exists to discriminate ferropericlasite of lower-mantle origin without
 29 characteristic mineral associations, such as co-existing former bridgmanite. To explore
 30 ferropericlasite-diamond growth relationships, we have investigated the crystallographic orientation
 31 relationships (CORs), determined by single-crystal X-ray diffraction, between 57 ferropericlasite
 32 inclusions and 37 diamonds from Juina (Brazil) and Kankan (Guinea). We show that ferropericlasite
 33 inclusions can develop specific (16 inclusions in 12 diamonds), rotational statistical (9 inclusions in
 34 7 diamonds) and random (32 inclusions in 25 diamonds) CORs with respect to their diamond hosts.
 35 All measured inclusions showing specific CORs were found to be Fe-rich ($X_{\text{FeO}} > 0.20$). Coexistence
 36 of non-randomly and randomly oriented ferropericlasite inclusions within the same diamond indicates
 37 that their CORs may be variably affected by local growth conditions. However, the occurrence of
 38 specific CORs *only* for Fe-rich inclusions indicates that Fe-rich ferropericlases have a distinct genesis
 39 and are syngenetic with their host diamonds. This result provides strong support for a dual origin for
 40 ferropericlasite in Earth's mantle, with Fe-rich compositions likely indicating redox growth in the
 41 upper mantle, while more Mg-rich compositions with random COR mostly representing ambient
 42 lower mantle trapped as protogenetic inclusions.

43 **Keywords:** Ferropericlasite · diamond · crystallographic orientation relationship · growth relationship
 44 · syngensis · protogenesis.

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1. Introduction

Diamonds are the only natural samples through which we can investigate the mineralogy and geological processes occurring in Earth's mantle at depths down to ~ 800 km depth. Most information is provided by mineral and fluid inclusions entrapped by diamonds during their crystallization (Meyer, 1987; Shirey et al., 2019, 2013; Weiss et al., 2015). Ferropericlasite, an oxide mineral with composition ranging from MgO (periclase) and wüstite (FeO), is the most abundant inclusion in super-deep diamonds, i.e., forming at sub-lithospheric depths. Experiments and theoretical models on pyrolitic compositions indicate that ferropericlasite is stable in the lower mantle, at depths between ~ 660 and 2900 km, and represents ~ 17% of the mantle phase assemblage in a "fertile" mantle bulk composition, the remainder being represented by bridgmanite (76%) and CaSiO₃-perovskite (7%) (Akaogi, 2007; Ishii et al., 2018). The predicted chemical composition of lower-mantle ferropericlasite is Mg-rich, with X_{FeO} (FeO molar fraction) ranging from 0.08 to 0.18 (Hirose, 2002; Irifune, 1994; Ishii et al., 2018, 2011; Kuwahara et al., 2018). Ferropericlasite, however, represents ~ 42% of the inclusions reported within super-deep diamonds, far more abundant and showing much more variable compositions with X_{FeO} up to 0.85 than would be expected for pyrolitic mantle (Walter et al., 2022 and references therein).

Numerous studies (see Walter et al., 2022 for a review) tried to explain these discrepancies and unravel the possible geological processes involved in formation of ferropericlasite-bearing diamonds. Assuming that ferropericlasite-bearing diamonds crystallized in the lower mantle, Liu (2002) proposed a model according to which (Fe-rich) ferropericlasite and diamond can simultaneously precipitate through decarbonation of (Mg,Fe)CO₃. Alternatively, Ryabchikov & Kaminsky (2013) and Kaminsky & Lin (2017) supposed the existence of a non-pyrolitic source in the lower mantle. However, experiments demonstrate that ferropericlasite can be stable in mantle rocks at depths shallower than the lower mantle (Brey et al., 2004). In particular, Thomson et al. (2016) showed that ferropericlasite with variable Fe contents plus diamond can crystallize simultaneously by interaction

between mantle peridotite and slab-derived carbonatite melts in the deep upper mantle or transition zone. Therefore, in the absence of limiting characteristic mineral associations, such as the presence of former bridgmanite, the depth of origin of ferropericlasite-bearing diamonds remains uncertain. Only inclusions associated with low-Ni enstatite, considered to be the back-transformation product of bridgmanite (Stachel et al., 2000), can safely be ascribed to the lower mantle. About 15% of these also co-exist with MgSiO_3 or/and CaSiO_3 phases in diamonds (Walter et al., 2022).

Determining ferropericlasite-diamond growth relationships, for instance whether the inclusion and host crystallized simultaneously or whether the inclusion preceded the host, is crucial for determining the possible genetic processes that formed ferropericlasite-bearing diamonds. Determination of crystallographic orientation relationships (CORs) for inclusion-diamond systems is commonly used to derive information about their growth relationships (Milani et al., 2016; Nestola et al., 2019, 2017, 2014, Nimis et al., 2019, 2018; Pamato et al., 2021; Pasqualetto et al., 2022). In a preliminary study, Nimis et al. (2018) determined CORs for nine Fe-rich ($X_{\text{FeO}} \approx 0.33$ to ≥ 0.64) ferropericlasite inclusions in two diamonds from Juina, Brazil. These inclusions are *specifically* oriented with their diamond hosts, with the principal crystallographic axes of ferropericlasite fixed to those of the diamond host, suggesting an epitaxial relationship. Accordingly, Nimis et al. (2018) proposed that such ferropericlasite nucleated during the growth history of the diamond, probably by the same type of redox reactions investigated by Thomson et al. (2016) at depths of the deep upper mantle or transition zone. In order to increase the statistical significance of the data and to gain further insight into ferropericlasite-diamond growth relationships, we have determined the CORs for 57 ferropericlasite inclusions in 37 diamonds spanning a large compositional range to determine possible associations between ferropericlasite Fe-content and the depth origins of ferropericlasite-bearing diamonds.

2. Samples and Methods

2.1. Samples

97 In this work, we investigated 57 ferropericlasite inclusions within 37 diamonds from two classic super-
 98 deep diamond localities. A representative example of one of these diamonds is shown in Fig. 1. Of
 99 the investigated samples, 34 diamonds with 49 inclusions in total come from Juina, Brazil, and 3
 100 diamonds with 8 inclusions in total come from Kankan, Guinea. All the studied diamonds come from
 101 alluvial deposits. They are colourless to pale yellow-brown and their size ranges from ~ 1.5 to 5 mm.
 102 They show octahedral to irregular shapes and contain from one to four optically visible and
 103 measurable ferropericlasite inclusions. The ferropericlasite inclusions, are sub-rounded to irregular, 50-
 104 200 μm in size, dark in colour and show characteristic iridescence. In some specimens, other mineral
 105 and fluid phases also occur (such as calcite, dolomite, magnesite, nahcolite, olivine, breyite and a
 106 fluid phase similar to that reported in Nimis et al., 2016).

107 *2.2. Single-crystal X-ray diffraction*

108 X-ray diffraction data for 24 ferropericlasite inclusions and 18 diamonds were collected using a Rigaku
 109 Oxford SuperNova diffractometer located at the Department of Geosciences, University of Padua.
 110 This instrument is equipped with a Dectris Pilatus 200K area detector and a *Mova* X-ray micro source,
 111 operating at 50 kV and 0.8 mA. The detector distance is 68 mm and the diffractometer is controlled
 112 by the CrysAlis-PROTM software. Initially, each ferropericlasite inclusion was centred optically and
 113 subsequently more precisely aligned by X-ray diffraction. The diffraction data were collected in 360°
 114 phi-scan mode. Each frame width was 1° and the exposure time was 25-60 s per frame, as a function
 115 of the inclusion size. The CrysAlis-PROTM software was also used to process the collected data. By
 116 indexing the position of the diffracted peaks from the inclusions and the hosts, we determined their
 117 orientation matrices, which represent the inclusion or host orientation relative to the reference system
 118 of the diffractometer. Through the indexing procedure, we could unambiguously distinguish
 119 diffraction peaks from ferropericlasite apart from those of diamond in the same data set.

120 The remaining 33 ferropericlasite inclusions within 19 diamonds were analysed at the single-crystal
 121 X-ray diffraction beamline (13-BM-C) of the GeoSoil Enviro Center for Advanced Radiation Sources

(GSECARS), Advanced Photon Source (APS), Argonne National Laboratory, USA. For the synchrotron X-ray diffraction experiments, centering ferropericlasite inclusions in diamond was facilitated using the 2D radiography attachment on beamline 13-BM-C (Wenz et al. 2019). For diffraction, the X-ray beam was focused to 12 μm horizontal by 18 μm vertical at full-width half-maximum. Final centering and diffraction were carried out on the six-circle goniometer following the methods detailed in Zhang et al. (2017). Step scans were obtained with one-degree steps over 180° with an exposure time of one second per step using a MAR 165 CCD detector. Additional details about the combined 2D radiography and synchrotron X-ray diffraction data collection and software are reported in Wenz et al. (2019).

2.3. COR determination: OrientXplot software and misorientation distribution analysis

The OrientXplot software (Angel et al., 2015) was used to determine and plot the CORs. This program processes each orientation matrix and displays a stereogram of the crystallographic orientations of inclusions relative to their host, avoiding ambiguities arising from crystal symmetry. In this case, both inclusions and hosts are cubic. Consequently, for each ferropericlasite-diamond pair, 576 symmetrically equivalent orientations are possible. Therefore, for each inclusion-host pair, we have chosen to plot the orientation for which $[1\ 1\ 0]_{\text{FPer}}$ is closest to $[1\ 1\ 0]_{\text{Dia}}$ and $[0\ 0\ 1]_{\text{FPer}}$ is closest to $[0\ 0\ 1]_{\text{Dia}}$.

In order to determine the statistical significance of CORs in our inclusion-host systems, we carried out a misorientation distribution analysis. For this purpose, we considered the angles between the crystallographic axes or planes of ferropericlasite and diamond that are the most likely to form non-random CORs (e.g., Nimis et al., 2019; Pasqualetto et al., 2022). The calculated misorientation distributions were compared with a theoretical model of 2 million randomly oriented matrices through the Kolmogorov-Smirnov test for two samples (see Wheeler et al., 2001 for more information). Identification of specific, rotational statistical or random CORs was then based on the presence or not

of a statistically significant similarity between one or more pairs of specific crystallographic directions of inclusions and hosts (Griffiths et al., 2016; Habler and Griffiths, 2017).

To increase the number of data and the statistical significance, the same procedures were extended to include also two ferropericlasite inclusions in diamond *AZI* previously studied by Anzolini et al. (2019).

2.4. Ferropericlasite chemical composition

The chemical compositions of three ferropericlasite inclusions (inclusions in samples *AZ_08*, *AZ_15* and *AZ_20*) were determined using a Tescan Solaris dual beam FE-SEM, equipped with an Ultim® Max 65 EDS spectrometer. Analytical conditions were 15 keV, 3 nA, and 20 s counting time. Analyses were standardised using pure oxides as standards, excepting Na, which was calibrated on albite. In addition, the chemical data of four ferropericlasite inclusions within *KK207* diamond were collected by electron probe micro-analysis using a JEOL JXA-8900R with 5 wavelength dispersive spectrometers, located at the University of Alberta. The beam energy was 20 keV energy with 30 nA of beam current and 2 µm diameter. The counting time was 20 seconds for Si Kα, Fe Kα, Mn Kα, Ni Kα, Zn Kα, 30 seconds for V Kα, Ti Kα, Cr Kα, 40 seconds for Na Kα, K Kα, Ca Kα, Mg Kα, and 120 seconds for Al Kα.

3. Results

3.1. Crystallographic orientation relationships (CORs)

A COR is defined as a systematic relation between the crystallographic orientations in an inclusion-host system. Four types of CORs can be distinguished based on the degrees of freedom between inclusion and host orientations: specific, rotational statistical, dispersional statistical and random (Griffiths et al., 2016; Habler and Griffiths, 2017). This classification is only descriptive and independent from the mechanisms of their formation. In specific CORs, at least two crystallographic directions of the inclusion are fixed to the host (0 degrees of freedom). In rotational statistical CORs,

170 only one inclusion crystallographic orientation is fixed to that of the host (1 degree of freedom). In
 171 dispersional statistical CORs, an inclusion crystallographic direction is not exactly fixed to the host,
 172 but is dispersed around it within a certain misorientation angle range (2 degrees of freedom, but within
 173 strict limits). In all other cases, the inclusion crystallographic directions are randomly oriented relative
 174 to the host (2 degrees of freedom, with no limit).

175 The CORs for all the 57 analysed ferropericlasel inclusions are shown in Fig. 2. Sixteen inclusions
 176 have the three principal crystallographic axes (a_1, a_2, a_3) within 0-12° of those of their diamond hosts
 177 (Fig. 3a). Despite the angular mismatch being in some cases greater than the measurement
 178 uncertainties of $\pm 4^\circ$ (Nimis et al., 2019), all inclusions have their $[1\ 1\ 2]$ axis within uncertainty of $[1$
 179 $1\ 2]_{\text{Dia}}$ at $< \pm 4^\circ$. These results are similar to those reported by Nimis et al. (2018) on nine ferropericlasel
 180 inclusions in two diamonds. As suggested by Nimis et al. (2018), the small angular misorientation of
 181 the main crystallographic axes may be due to a slight rotation around the $[1\ 1\ 2]$ direction, caused by
 182 post-entrapment plastic deformation, which is well documented in super-deep diamonds (e.g. Agrosi
 183 et al., 2017; Howell et al., 2012). All these inclusions are thus interpreted to have been *specifically*
 184 oriented at the time of their incorporation. Another nine inclusions have their $[1\ 1\ 0]$ direction almost
 185 parallel (within $\pm 4^\circ$) to $[1\ 1\ 0]_{\text{Dia}}$ and the other crystallographic directions randomly rotated around
 186 this axis (Fig. 3b). These relationships indicate a rotational statistical COR. The remaining inclusions
 187 (32 inclusions in 25 diamonds) do not show any particular crystallographic orientation with respect
 188 to their hosts (Fig. 3c). The statistical significance of the observed specific and rotational statistical
 189 CORs was tested by comparing the observed misorientation angle distributions against a theoretical
 190 random distribution (Kolmogorov-Smirnov test for two samples, $p < 0.001$).

191 Diamonds containing more than one ferropericlasel inclusion (13 out of 37 diamonds) showed further
 192 interesting features. In three of these samples (5a08, 5a26, 5a27), inclusions that are specifically
 193 oriented coexist with others that are randomly oriented (Fig. 4a). Diamond 5a06 contains one
 194 specifically oriented inclusion and one that suggests a rotational statistical COR (Fig. 4b). In

diamonds *5a04 and 6b23*, one inclusion with a rotational statistical COR and one randomly oriented inclusion coexist (Fig. 4c). Finally, in four diamonds (*KK34, KK207, 6a05, 6b17*) more than one inclusion share a similar orientation, but they are randomly oriented relative to their diamond hosts (Fig. 5).

3.2. Ferropericlase chemical composition

The chemical compositions of ferropericlase inclusions in diamond *AZ_08* (1 inclusion), *AZ_15* (1 inclusion), *AZ_20* (1 inclusion) and *KK207* (4 inclusions) are reported in Table 1. The X_{FeO} fraction ranges from 0.14 to 0.32. Previous data for other crystallographically analysed ferropericlase inclusions in diamonds studied by (Anzolini et al., 2019) and Nimis et al. (2018) are reported in the same Table.

Discussion

Our analysis of 57 ferropericlase inclusions within 37 diamonds shows that ferropericlase can develop specific (16 inclusions in 12 diamonds), rotational statistical (9 inclusions in 7 diamonds) and random (32 inclusions in 25 diamonds) CORs with respect to their diamond hosts. Non-random (i.e., specific and rotational statistical) CORs indicate that mechanical or surface interaction occurred between ferropericlase and diamond during formation of the inclusion-host system (Habler and Griffiths, 2017; Wheeler et al., 2001). Mechanical juxtaposition of two well-shaped crystals is most likely to generate rotational statistical CORs, in which the two crystals share the axes normal to the juxtaposed faces (Nimis et al., 2019; Wheeler et al., 2001). In our samples characterised by rotational statistical COR, ferropericlase and diamond share a common $[1\ 1\ 0]$ axis. If the driving force for this COR was mechanical, this would imply juxtaposition of the $\{1\ 1\ 0\}$ faces of both minerals. Although ferropericlase and diamond can rarely develop $\{1\ 1\ 0\}$ faces during their growth (Koretsky et al., 1998; Sunagawa, 1990), their crystals commonly have octahedral habits with well-formed $\{1\ 1\ 1\}$ faces. Consequently, one would expect to observe frequent rotational statistical CORs around $[1\ 1\ 1]$

219 and not around $[1\ 1\ 0]$. Therefore, we do not favour a role of mechanical interaction in the
220 development of rotational statistical CORs in our samples.

221 Surface interaction may also cause the development of non-random CORs (Wheeler et al., 2001). In
222 fact, under favourable conditions, two mineral grains may align their crystal lattices or one of their
223 lattice directions to minimize their interface energy. Nimis et al. (2018) discussed the possible
224 scenarios that could lead to crystallographic alignment between inclusion and host by surface
225 interaction in super-deep diamonds. These scenarios include (1) grain rotation during static
226 recrystallization, or (2) mutual growth or epitaxial nucleation during crystallization from a fluid or
227 melt. Scenario 1 was considered to be highly unlikely, given the high-stress environment in which
228 super-deep diamonds form. Scenario 2 implies precipitation of the included minerals during the
229 growth history of diamond and we suggest may apply to all investigated ferropericlasite-diamond pairs
230 showing non-random CORs.

231 Coexistence of non-random and random CORs in some of the studied diamonds (Fig. 4) is not in
232 conflict with the above interpretation, since local physical-chemical and stress conditions may affect
233 the efficiency of surface interactions (Mutaftschiev, 2001; Wheeler et al., 2001). Therefore, the
234 absence of a non-random COR should not be considered as evidence against contemporaneous
235 growth. Also, a rotational statistical CORs could reflect a “starting preferred crystallographic
236 orientation” between ferropericlasite and diamond. This would explain the coexistence in some of our
237 samples of rotational statistical and either specific or random CORs within the same diamond.

238 Four diamonds each contain pairs of ferropericlasite inclusions, which are iso-oriented with respect to
239 each other, but are randomly oriented with respect to their diamond hosts (Fig. 5). In one of these,
240 diamond *6b17*, $[1\ 1\ 0]_{\text{FPer}}$ is 4° from $[1\ 1\ 0]_{\text{Dia}}$, but this relatively small misalignment may well
241 represent just one of an infinite number of possible random orientations. Inclusion iso-orientation
242 without a specific COR with the diamond host is considered to be evidence of a protogenetic origin

243 of the inclusions (Milani et al., 2016; Nestola et al., 2014; Nimis et al., 2019; Pamato et al., 2021;
244 Pasqualetto et al., 2022).

245 Our compilation of ferropericlasel inclusions for which both CORs and chemical data are available
246 (Anzolini et al., 2019; Nimis et al., 2018; and present study) (Table 1) indicates a strong relationship
247 between ferropericlasel Fe content and ferropericlasel-diamond growth relationships. Almost all (12
248 out of 13) Fe-rich ferropericlasel inclusions ($X_{\text{FeO}} > 0.2$) present specific CORs. Evaluating the
249 relationship between the Fe-rich composition of ferropericlasel and the development of specific COR
250 through a not-parametric statistical test (Fisher's exact test), we have obtained very low probabilities
251 ($p < 0.001$) that the presence of this specific COR is independent from the Fe-rich composition of
252 ferropericlasel within the studied population. This indicates that the association between these two
253 parameters is highly statistically significant. On the other hand, 4 out of 5 Mg-rich ferropericlasel
254 inclusions with $X_{\text{FeO}} < 0.2$ present random CORs, while the remaining one is compatible with both a
255 random and a rotational statistical COR. In diamond *KK207*, multiple Mg-rich inclusions show
256 evidence of a protogenetic origin (Fig. 5). Moreover, the reported Mg-rich ferropericlasel inclusions
257 have chemical compositions similar to those of ferropericlases in association with former bridgmanite
258 within diamonds ($n=33$, X_{FeO} ranging ~ 0.10 to 0.31 and one sample having $X_{\text{FeO}} \sim 0.35$, Davies et
259 al., 2004; Harte and Harris, 1994; Hayman et al., 2005; Stachel et al., 2000; Tappert et al., 2009).
260 Note that ferropericlases with $X_{\text{FeO}} > 0.2$ are not in chemical equilibrium with co-existing former
261 bridgmanite and these were probably entrapped during different diamond growth events, reflecting
262 different chemical environments in Earth's mantle (Harte and Harris, 1994; Hayman et al., 2005).
263 These results strongly suggest that Fe-rich and Fe-poor ferropericlases generally form by distinct
264 processes under distinct conditions.

265 We suggest that Fe-rich ferropericlasel inclusions, which frequently present specific CORs, are
266 syngenetic with their diamond hosts and were formed in the deep upper mantle or transition zone by
267 redox processes similar to those reproduced in Thomson et al.'s (2016) experiments (Fig. 6a).

268 Conversely, Mg-rich ferropericlasel inclusions, which have chemical compositions similar to those of
 269 ferropericlases associated with low-Ni enstatite (evidence of a lower mantle origin; Davies et al.,
 270 2004; Harte and Harris, 1994; Hayman et al., 2005; Stachel et al., 2000; Tappert et al., 2009) and
 271 those predicted for lower-mantle ferropericlasel (Akaogi, 2007; Ishii et al., 2018), present random
 272 CORs, and in some cases show clear evidence of protogenesis. Consequently, we propose that these
 273 Mg-rich ferropericlases represent parts of pre-existing mineral assemblages, which were partially
 274 dissolved and passively entrapped by diamond during its precipitation in the lower mantle (Fig. 6b).
 275 These results thus allow future geochemical studies of ferropericlasel to confidently distinguish those
 276 formed at relatively shallow mantle levels by slab mantle interaction from those likely present in the
 277 upper mantle before diamond crystallisation and entrapment. The observed relationships indicate that
 278 Fe-rich ferropericlasel is unlikely to reflect a typical upper or lower mantle composition.

279 **Conclusions**

280 The results of this study can be summarised as follows.

- 281 1) The determination of the relative crystallographic orientations of 57 ferropericlasel inclusions
 282 in 37 diamonds revealed the occurrence of specific, rotational statistical and random CORs.
- 283 2) A non-random COR is typical of Fe-rich ($X_{\text{FeO}} > 0.2$) ferropericlasel inclusions, whereas Fe-
 284 poor ($X_{\text{FeO}} < 0.2$) ferropericlasel inclusions show random CORs and sometimes exhibit clear
 285 evidence of protogenesis.
- 286 3) Fe-rich ferropericlasel inclusions presenting non-random CORs are interpreted to have been
 287 formed together with their host diamonds in the deep upper mantle or transition zone, probably
 288 by interaction of mantle peridotite with slab-derived carbonatite melts.
- 289 4) Mg-rich ferropericlasel inclusions presenting random CORs could be remnants of pre-existing
 290 mineral assemblages, which were entrapped by the growing diamonds in the lower mantle.

291 The dual origin of ferropericlasel inclusions in diamonds (Fe-poor protogenetic vs. Fe-rich syngenetic)
 292 provides a simple explanation for the observed discrepancies between theoretical mineralogical

293 models for the lower mantle and the relative abundance and composition of ferropericlasite inclusions
294 in diamonds.

295 **CRedit author statement**

296 **Sofia Lorenzon:** Conceptualization, Investigation, Formal analysis, Writing – Original Draft
297 **Michelle Wenz:** Investigation, Formal analysis, Writing – Review & Editing **Paolo Nimis:**
298 Conceptualization, Investigation, Writing – Review & Editing **Steven D. Jacobsen:**
299 Conceptualization, Funding acquisition, Writing – Review & Editing **Leonardo Pasqualetto:** Formal
300 analysis, Writing – Review & Editing **Martha G. Pamato:** Funding acquisition, Writing – Review
301 & Editing **Davide Novella:** Funding acquisition, Writing & Editing **Dongzhou Zhang:** Investigation
302 **Chiara Anzolini:** Writing – Review & Editing **Margo Regier:** Investigation **Thomas Stachel:**
303 Resources, Writing – Review & Editing **D. Graham Pearson:** Resources, Writing – Review &
304 Editing **Jeffrey W. Harris:** Resources, Writing – Review & Editing **Fabrizio Nestola:**
305 Conceptualization, Resources, Supervision, Funding acquisition, Writing – Review & Editing.

306 **Declaration of Competing Interest**

307 The authors declare that they have no known competing financial interests or personal relationships
308 that could have appeared to influence the work reported in this paper.

309 **Acknowledgements**

310 FN thanks the European Research Council (grant agreement n. 307322). DN thanks the Rita Levi
311 Montalcini programme (Italian Ministry of University and Research) for support. This study was also
312 supported in part through the U.S. National Science Foundation (NSF) grant EAR-1853521 to SDJ.
313 Portions of this work were performed at GeoSoilEnviroCARS (The University of Chicago, Sector
314 13), Advanced Photon Source (APS), Argonne National Laboratory. GeoSoilEnviroCARS is
315 supported by NSF grant EAR-1634415. This research used resources of the APS, a U.S. Department
316 of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne

317 National Laboratory under Contract No. DE-AC02-06CH11357. Experiments at beamline 13-BM-C
318 used the PX² facility, supported by COMPRES under NSF Cooperative Agreement EAR-1661511.

319 **Appendix A. Supplementary Material**

320 Supplementary material related to this article – files containing the UB-matrix data of both
321 ferropericlases and diamonds and the chemical data expressed in oxide abundances of ferropericlase
322 inclusions).

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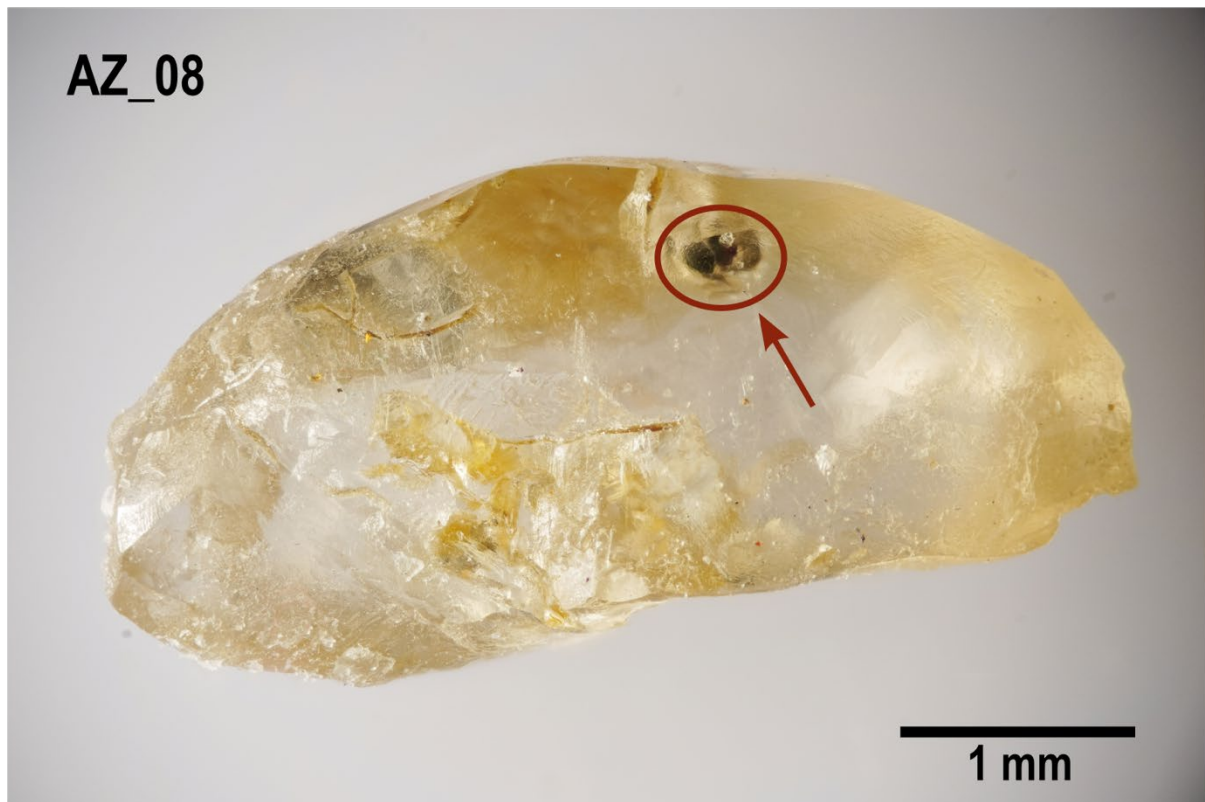
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453 **Figures**

454 Figure 1. One of the studied ferropericlasite-bearing diamonds (AZ_08) under incident light. This
455 specific sample comes from Juina (Brazil), is pale-yellow and has an elongated irregular shape. The
456 ferropericlasite inclusion (within the red circle and indicated by the red arrow) is dark in colour and
457 ~250 μm sized.



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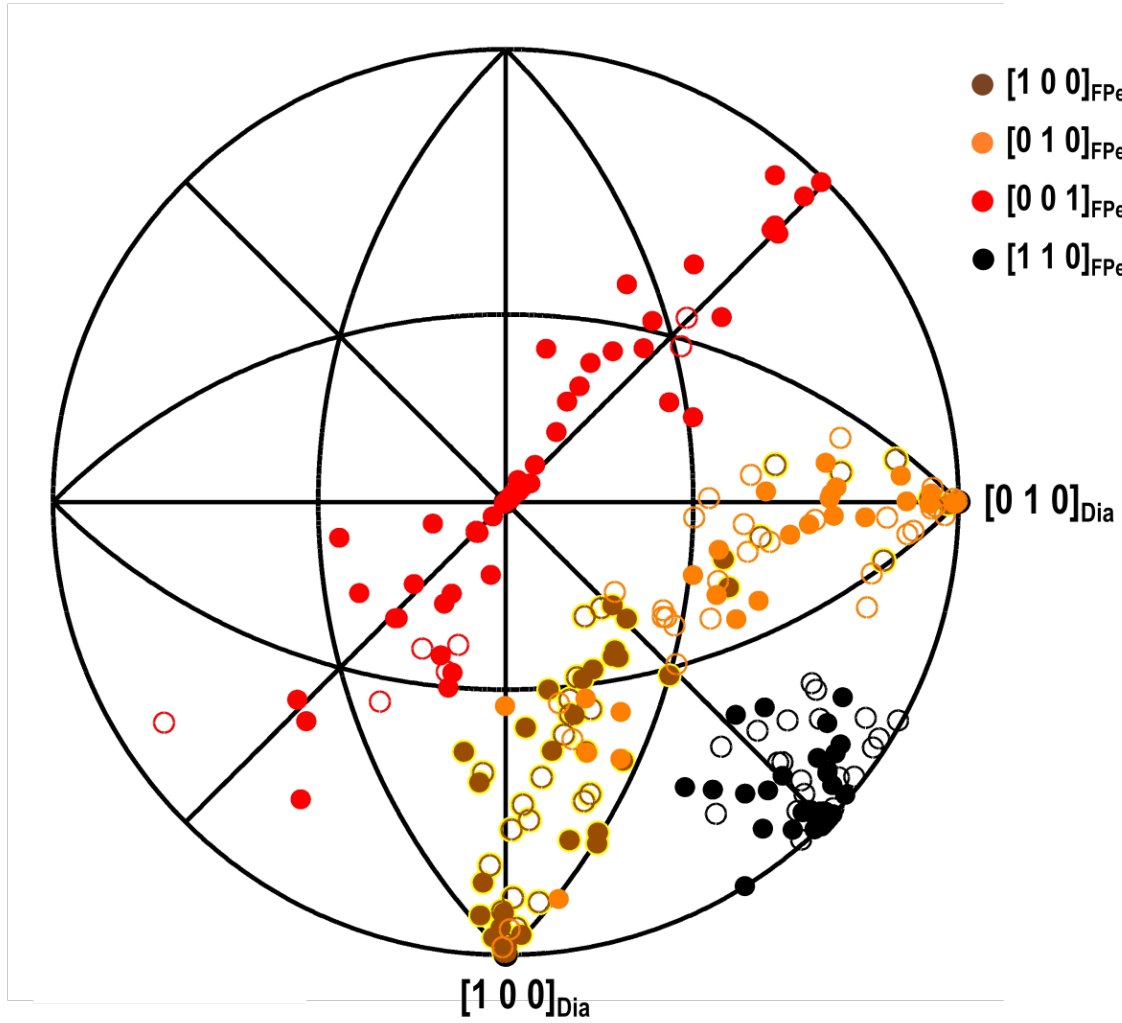
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464 Figure 2. Crystallographic orientation relationships (CORs) between all analysed 57 ferropericlas
465 inclusions and their 37 diamond hosts, plotted using OrientXplot software (Angel et al., 2015). Open
466 symbols plot in the lower hemisphere.



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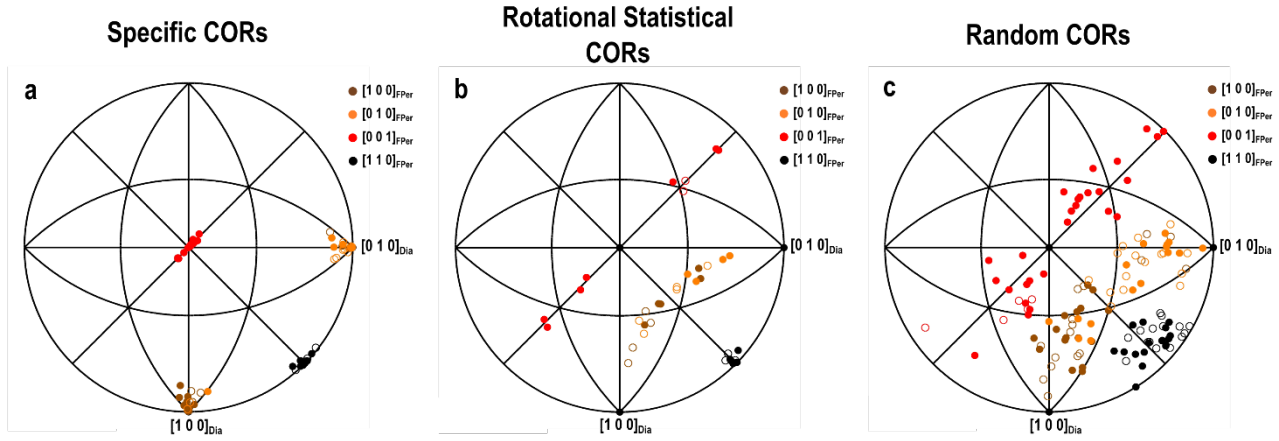
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473 Figure 3. Stereographic projections showing ferropericlasite inclusions presenting a) specific CORs
 474 (16 inclusions in 12 diamonds), b) rotational statistical CORs ($[1\ 1\ 0]_{\text{FPer}} // [1\ 1\ 0]_{\text{Dia}}$, 9 inclusions in
 475 7 diamonds) and c) random CORs (32 inclusions in 25 diamonds) relative to their diamond hosts.



488 Figure 4. Stereographic projections of diamonds containing ferropericlase inclusions which present
 489 a) both specific and random CORs (5a08, 5a26, 5a27), b) both specific and rotational statistical CORs
 490 (5a06) and c) both rotational statistical and random CORs (5a04, 6b23) relative to their diamond
 491 hosts.

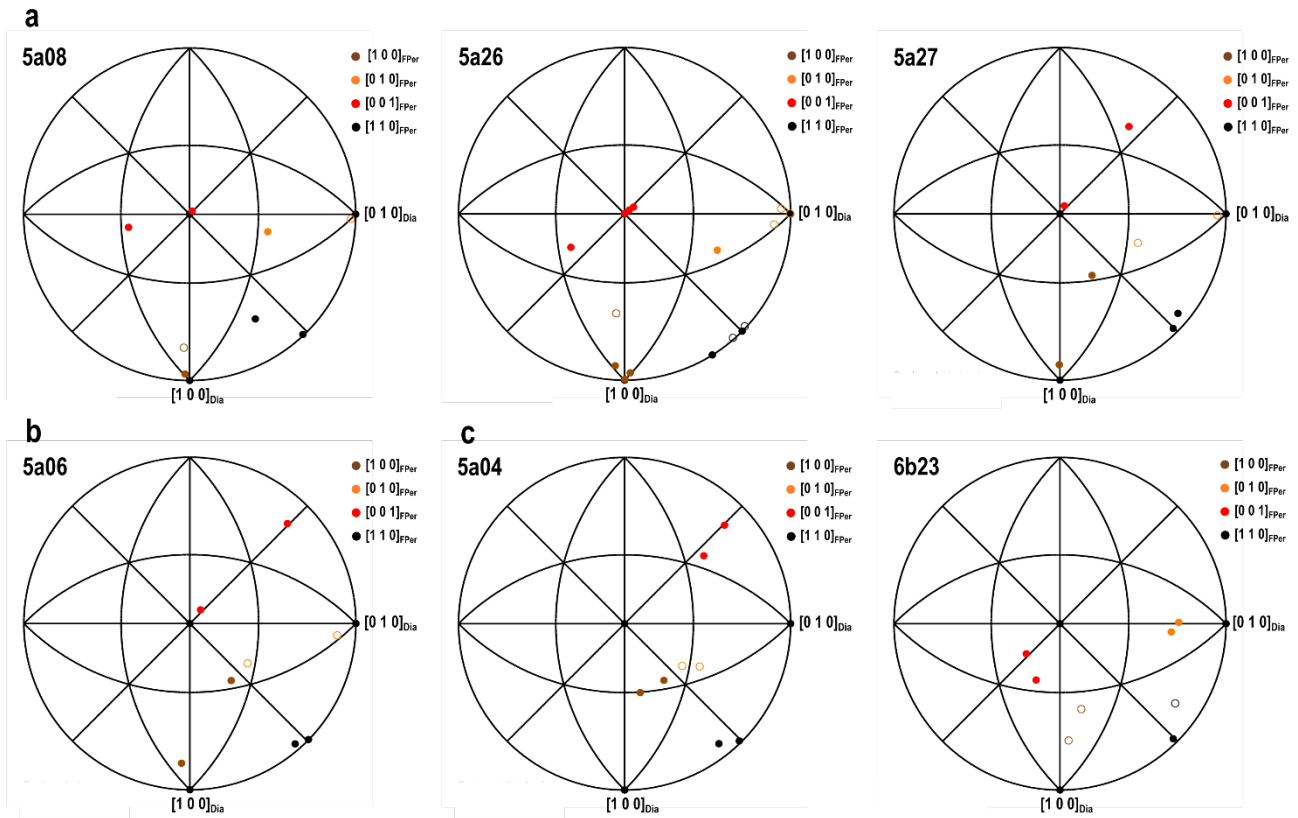


Figure 5. Crystallographic orientation relationships (CORs) of ferropericlasite inclusions within *KK34*, *KK207*, *6a05* and *6b17* diamonds. Multiple ferropericlasite inclusions are defined by the numbers close to dots (i.e. 1, 2,...). Blue circles indicate the ferropericlasite inclusions presenting similar CORs, but different and random CORs to their diamond host. These inclusions are protogenetic, representing remnant parts of pre-existing mono-crystals that were dissolved and entrapped during diamond precipitation.

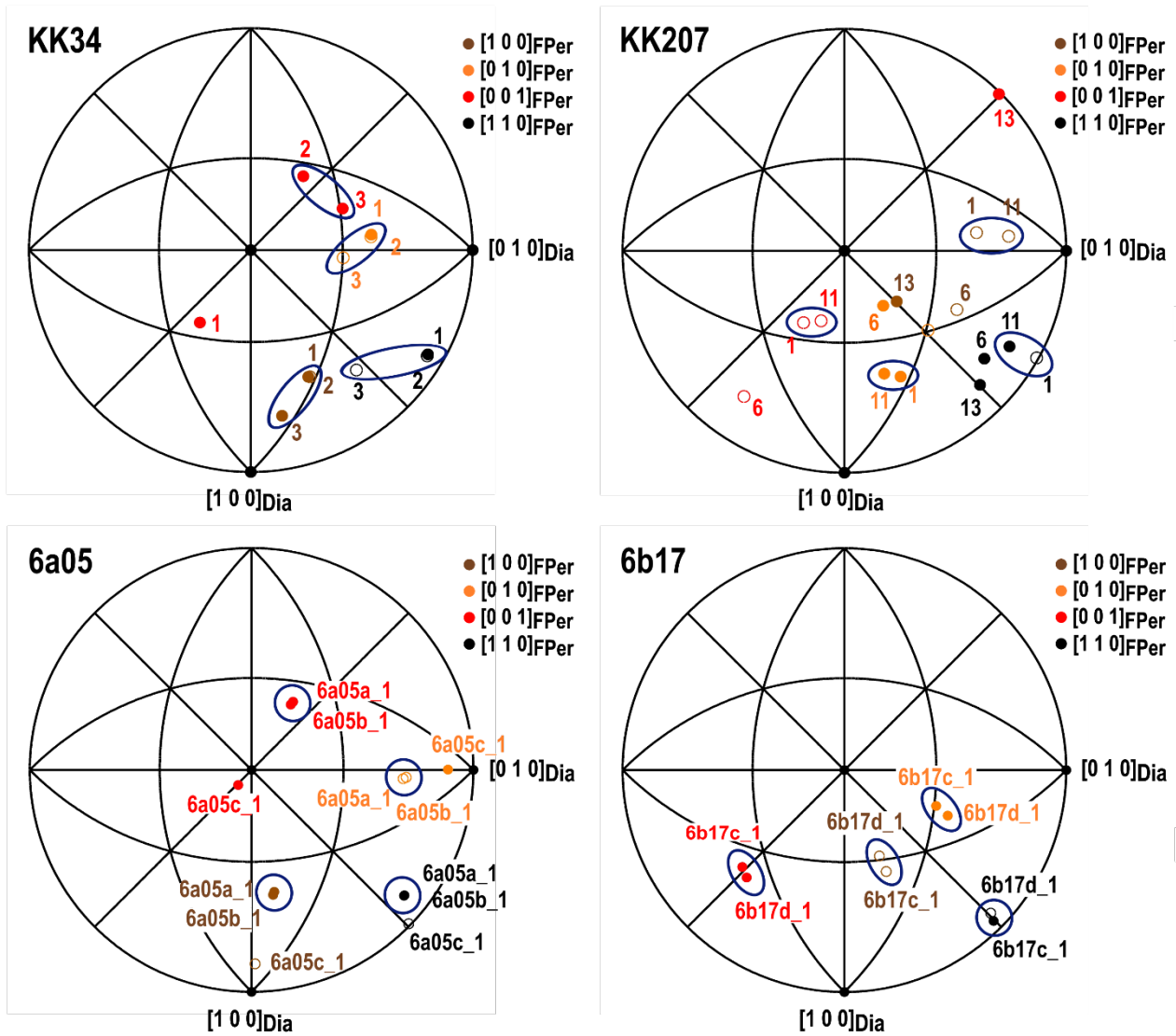
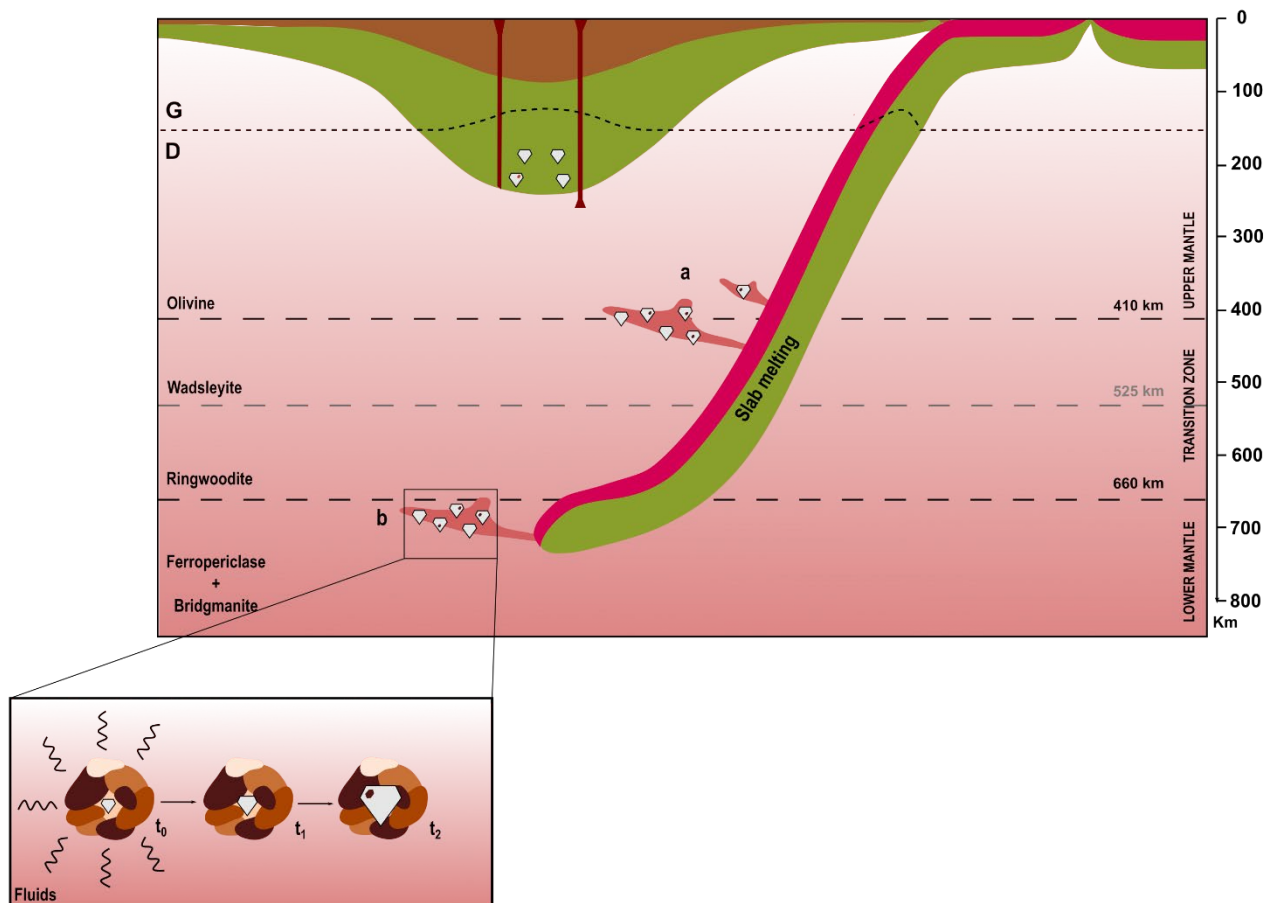


Figure 6. Possible scenarios for the formation of Fe-rich and Fe-poor ferropericlas-bearing diamonds in Earth's mantle. a) Precipitation of diamonds and Fe-rich ferropericlas due to reactions between slab-derived carbonatite melts and peridotitic rocks, at depths of the deep upper mantle or of the transition zone (Thomson et al., 2016). In this case, the ferropericlas inclusions are syngenetic and generally develop specific CORs with their diamond hosts. b) Formation of diamonds in the uppermost lower mantle. In this case, pre-existing Mg-rich ferropericlas inclusions are partially dissolved and passively incorporated into the growing diamonds, without development of particular CORs. Multiple inclusions in individual diamonds may be iso-oriented if they are derived from the same original ferropericlas grain. Only in this case do the inclusions have chemical compositions similar to those experimentally predicted for ferropericlas in the lower mantle.



523 **Tables**

524 Table 1. Chemical composition and interpreted COR of ferropericlasel inclusions.

	Diamond	Inclusion	Chemical composition	Type of COR
Nimis et al. 2018	BZ270	1	(Mg _{0.66} Fe _{0.34})O	Specific
		2	(Mg _{0.65} Fe _{0.35})O	Specific
		3	(Mg _{0.65} Fe _{0.35})O	Specific
		4	(Mg _{0.65} Fe _{0.35})O	Specific
		5	(Mg _{0.66} Fe _{0.34})O	Specific
	JUc4	1	(Mg _{0.43} Fe _{0.57})O	Specific
		2	(Mg _{0.56} Fe _{0.44})O	Specific
		3	(Mg _{0.57} Fe _{0.43})O	Specific
		4	(Mg _{0.36} Fe _{0.64})O	Specific
Anzolini et al. 2019	AZ1	AZ1_1	(Mg _{0.61} Fe _{0.39})O	Specific
		AZ1_2	(Mg _{0.59} Fe _{0.41})O	Specific
This study	AZ_08	AZ_08_01	(Mg _{0.68} Fe _{0.32})O	Random
	AZ_15	AZ_15_01	(Mg _{0.80} Fe _{0.20})O	Random
	AZ_20	AZ_20_01	(Mg _{0.69} Fe _{0.31})O	Specific
	KK207	1	(Mg _{0.86} Fe _{0.14})O	Random
		6	(Mg _{0.86} Fe _{0.14})O	Random
		11	(Mg _{0.86} Fe _{0.14})O	Random
		13	(Mg _{0.86} Fe _{0.14})O	Rotational statistical

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