

1 *Revision 1*

2 **Origin of milky optical features in type IaB diamonds: dislocations, nano-inclusions, and**
3 **polycrystalline diamond**

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ABSTRACT

10 The milky appearance shown by certain type IaB diamonds has been subjected to several
11 recent studies but the origin of this feature is not fully understood. Here several type IaB diamonds
12 with milky appearance have been studied by cathodoluminescence (CL), electron backscatter
13 diffraction (EBSD), and transmission electron microscopy (TEM). CL of several hazy type IaB
14 diamonds shows scattered or orientated micro-sized spots or short linear luminescence features.
15 TEM observation revealed that those spots and linear features are caused by dislocation loops that
16 are likely responsible for the hazy appearance of the host diamonds. It also shown that type IaB
17 diamonds with a cloudy appearance contain nano-sized inclusions with negative crystals of
18 octahedral shape. Some of these negative crystals contain a precipitate that can be explained by a
19 compressed disordered cubic δ -N₂ phase observed by high-resolution TEM. In one of the milky
20 IaB diamonds with platelet defects, polycrystalline areas composed of columnar diamond crystals
21 elongated radially in [110], similar to ballas diamond, were revealed by EBSD. Taking into
22 account these observations, it is suggested that the dislocation loops, nano-sized inclusions
23 (negative crystals) and/or characteristic grain boundaries of the radiating fibrous crystals would be

24 the origins for the milky appearance of the type IaB diamonds studied here. Those results add
25 complementary explanation accounting for the milkiness of type IaB diamonds studied before.

26 **Key words:** Type IaB diamonds, milky, dislocations, voidites, polycrystalline diamond

27 INTRODUCTION

28 The origin of milky diamonds is enigmatic. Unlike diamonds with intense fluorescence
29 that creates an “oily” appearance, milky diamonds contain areas with distinct textures, scattering
30 light and resulting in a hazy or cloudy appearance. With dense “clouds” inside, they can be termed
31 “fancy white” diamonds. The exact cause of their opacity is not fully understood. In previous
32 studies, the term “cloudy” has been used to refer to fibrous diamonds or describe diamonds with
33 submicroscopic internal inclusions. Extensive studies have shown that those frosted zones are
34 filled with disk- crack-like graphite inclusions (Rakovan et al. 2014) or high-density fluids (HDFs)
35 from a deep origin, with compositions ranging between carbonatitic and saline end-members
36 (Navon et al., 1988; Izraeli et al. 2001; Klein-BenDavid et al. 2007; Tomlinson et al. 2009;
37 Logvinova et al. 2011). However, cloudy inclusions filled with HDFs usually appear gray or black,
38 while the milky diamonds are generally white in color, suggesting a different filling.

39 It has been noticed that milky white diamonds are most often type IaB (Fritsch 1998), with
40 nitrogen in B-aggregates (four nitrogen atoms around a single vacancy, Loubser and van Wyk,
41 1981). The development of B centers is usually accompanied by the formation of platelets, planar
42 defects in {001} planes that measure a few nanometers to a few micrometers in diameter (Clackson
43 et al., 1990; Speich et al., 2017). In some pure type IaB diamonds, that have been termed “irregular”
44 (Woods 1986), platelets have experienced degradation and may be absent completely. Platelet

45 degradation is often accompanied by the formation of voidites (e.g. Barry et al. 1987). Previous
46 studies have confirmed that voidites and nano-inclusions can be found in type IaB diamonds with
47 milky zones (Rudloff-Grund et al. 2016; Navon et al. 2017). However, the composition of those
48 nano-inclusions is still a subject of debate. Electron energy loss spectroscopy (EELS; Bruley and
49 Brown 1989) and energy- dispersive X-ray spectroscopy studies (EDX; Rudloff-Grund et al. 2016)
50 reveal the presence of nitrogen in the voidites. Based on moiré patterns, the material filling the
51 voidites has been identified as NH₃ (Barry 1986; Hirsch et al. 1986a) or a tetragonal N₂ phase
52 (Luyten et al. 1994; Navon et al. 2017). If such voidites are the products of platelet degradation,
53 elements that constitute the platelets should be present in the voidites or alongside the dislocation
54 loops. However, the structure and composition of the platelets are still unclear. In previous studies,
55 the structural model of platelets with a nitrogen double layer (Lang 1964) was excluded because
56 EELS investigations have shown that the concentration of nitrogen within the platelets is too low
57 to fit with this model (Berger and Pennycook 1982). The generally accepted platelet model is that
58 of a pentagonal interstitial carbon arrangement in the {100} planes as proposed by Humble (1982).
59 Theoretical studies have indicated that the strain associated with small platelets could lead to
60 shallow electronic gap levels that could promote optical transitions from point defects such as
61 nitrogen and vacant sites close to the platelet (Goss et al. 2003). Investigation of the products
62 from platelet degradation would provide more information about their constitution.

63 In milky diamonds, some contain clusters and areas of cloudy inclusions that are generally
64 visible as very small whitish pinpoints under microscopic observation (Fig. 1). However, in some
65 milky diamonds, no inclusions are visible even with high magnification up to 200×, and the
66 diamonds generally display a slightly hazy appearance (Fig. 1). The different optical features
67 might indicate diverse causes for their milky appearance. Although platelet degradation is often

68 associated with milky type IaB diamonds, a subgroup of milky diamonds shows sharp platelet
69 peaks, and therefore the cause of their milky appearance is still unclear. To gain a comprehensive
70 understanding of the various origins of the milky features in type IaB diamonds, we examined
71 their optical and structural properties through cathodoluminescence (CL) imaging and
72 transmission electron microscopy (TEM), and investigated their micro-textures by electron
73 backscatter diffraction (EBSD). Based on the results, we discuss origins of the milky appearance
74 in type IaB diamonds and the implications in diamond-forming processes.

75 **METHODS**

76 **Sample characteristics**

77 Samples were collected from diamonds submitted by clients to GIA's laboratory for
78 screening; rough diamonds and faceted diamonds were cut into plates for detailed study. We were
79 able to have a close look of samples listed in Table 1 and six of them were cut into plates. All
80 plates except the one with mineral inclusions on the surface were cleaned in a mixture of HCl and
81 NaNO₃ for one hour at 100°C. The carat weight, clarity grade, and internal characteristics of each
82 stone are listed in Table 1. Features of FTIR and photoluminescence spectra of those
83 diamonds have been detailed in our previous study (Gu and Wang 2018), and FTIR spectra of all
84 samples are provided in supplementary material (Fig. S1). All samples were typical milky type
85 IaB diamonds that contained hazy or cloudy areas. Under high magnification, no inclusions could
86 be resolved with the microscope in the hazy areas; in contrast, whitish spots were spread
87 throughout the cloudy area.

88 **CL images**

89 Experiments were performed at GIA using a Zeiss EVO MA10 scanning electron
90 microscope (SEM) equipped with a high-resolution CL system. The acquisition of digital grayscale
91 panchromatic CL images was achieved using a multichannel analyzer at room temperature,
92 operating in variable pressure mode with the chamber pressure typically at 20 Pa. Images were
93 collected with a variable pressure secondary electron (VPSE) detector at zero bias, using 15–20
94 kV accelerating voltage and specimen probe currents between 1 and 20 nA. A typical probe current
95 of 100 pA and a voltage of 15 kV were used for most samples, while a smaller voltage of 10 kV
96 was used to increase the resolution for fine textures.

97 **TEM analysis**

98 TEM foils were prepared by a focused ion beam (FIB) system (JOEL JEM- 9310FIB). The
99 detailed procedure of FIB milling for TEM foil preparation is described elsewhere (e.g., Ohfuji et
100 al. 2010). Each foil has a dimension of approximately $10 \times 7 \times 0.1 \mu\text{m}$. Transmission electron
101 microscopy was carried out using a JEOL JEM-2010 instrument at Ehime University and using a
102 JEOL 2100F instrument at the Center for Functional Nanomaterials (CFN) at the Brookhaven
103 National Laboratory; both equipped with a field emission gun (FEG), operated at an accelerating
104 voltage of 200 kV. Generally, bright- and dark-field images as well as high-resolution images were
105 acquired as energy-filtered images applying a 20 eV window to the zero loss peak. Diffraction data
106 were calculated from high-resolution images using the fast Fourier transform algorithm (FFT).

107 **EBSD analysis**

108 FE-SEM measurements were carried out on a JEOL JSM-7000F instrument equipped with
109 a Nordlys EBSD detector at Ehime University. Channel 5.0 software was used for data acquisition

110 and analysis. For EBSD using the SEM, an accelerating voltage of 15 kV, a beam current of 4.0
111 nA, and a working distance of 15 mm were applied for the analysis. The sample was coated with
112 carbon of ~10 nm thickness. Prior to the analysis, an orientation contrast (fore-scattered electron)
113 image was taken by photodiode detectors located at the upper edge of the EBSD camera.
114 Orientation maps were obtained with a step size of 2 or 3 μm by indexing a maximum of eight
115 diffraction lines detected from each Kikuchi pattern. After each analysis, “Tango” and “Mambo”
116 components from the Channel 5.0 software were used to draw an orientation map and pole figure(s)
117 respectively.

118 RESULTS

119 CL features

120 The descriptions of grayscale CL images collected at room temperature for each sample
121 are summarized in Table 1. All samples with a hazy area or “clouds that are not shown” displayed
122 pinpoints under grayscale CL. Some luminescent pinpoints can be resolved as short lines under
123 high magnification. Those luminescent short lines were either straight or curved, usually ranging
124 in size from a few microns to ten microns in size, and could form sets of lines parallel with each
125 other (Fig. 2b-c). Some were oriented in certain crystallographic directions (Fig. 2a–c). Those
126 short lines were often observed in the black areas of the grayscale CL images (Fig. 2b), and some
127 were distributed between the boundaries of black and white regions (Fig. 2d). Some of the
128 pinpoints could also be observed in growth zone areas. They were confined to a small area or
129 spread throughout the whole diamond. Platelet peaks were not observed in FTIR spectra in those
130 diamonds with luminescent pinpoints throughout the whole stones (i.e., 890000048675,
131 110208618119), while one sample (890000102598) with pinpoints in a small area showed a very

132 small residual peak at 1367.7 cm^{-1} (Table 1, Fig. S1). No typical CL features were observed for
133 diamonds with cloudy inclusions that could be recognized as whitish points under the microscope.

134 **TEM results**

135 FIB foils were prepared from the hazy areas that showed distinct luminescent pinpoints
136 under grayscale CL and from the cloudy areas where tiny whitish pinpoints could be observed
137 under the microscope. TEM images from the hazy areas showed only dislocation loops (Fig. 3a–
138 b). No inclusions or voidites were observed in these FIB foils. Strain contrast was observed at the
139 edge of the dislocations, which showed a typical “coffee-bean” contrast (Hirsch et al. 1965). On
140 the other hand, nano-inclusions of ~ 20 to 200 nm diameter were observed in the FIB foils cut from
141 the cloudy areas (Fig. 3c). They show octahedral (Fig. 3d), elongated (Fig. 3e–f), or rounded (Fig.
142 3c) shapes. Most of the octahedral nano-inclusions were 20 to 30 nm in diameter, whereas some
143 were as large as 100 nm (Fig. 3d). They were usually aligned parallel to $\{111\}$ faces (Fig. 3d). We
144 also observed an inclusion with a rounded outline and a rhombic projection inside (Fig. 3c).
145 Inclusions with elongated shape were as small as 20–30 nm in dimension (Fig. 3d, f), while some
146 of them were up to 200 nm (Fig. 3e).

147 Fig. 4 shows two high-resolution TEM (HRTEM) images taken from exactly the same
148 region as one of the nano-inclusion, with slightly different focus conditions, where faint lattice
149 fringes were recognized in two directions inside the nano-inclusions. The d -spacings of the fringes
150 (2.39 and 2.63 Å) obtained from the images by FFT conversion cannot be indexed by the diamond
151 lattice, and therefore they are derived from an additional crystalline phase precipitated inside the
152 negative crystal.

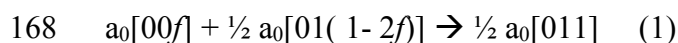
153 **EBSD results**

154 Kikuchi patterns indicated that all samples were single-crystal diamond except for one
155 sample that showed a sharp residual platelet peak in its FTIR spectrum (sample 110207975364).
156 EBSD mapping revealed that this sample had a granular texture with distinct grain boundaries and
157 grain sizes ranging from ~10 to 200 μm (Fig. 5). Approximately 611 grains were detected in the
158 analyzed region and the average grain size is 50 μm according to the EBSD statistical results. Fig.
159 6 shows pole figures projected along the $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$ planes of diamond, where a
160 strong preferred orientation of $[110]$ was observed almost normal to the orientation map. This
161 suggests that the observed granular domains may be cross-sections of fibrous or columnar crystals
162 that are elongated in the $[110]$ direction perpendicular to the sample (section) surface.

163

DISCUSSIONS

164 Previous experiments by other authors have shown that dislocation loops may form at
165 high temperatures close to the graphite-stable region during the platelet degradation process. The
166 conversion of platelets into dislocations may take place by the dislocation reaction (Hirsch et al.
167 1986b)



169 where f is the matrix displacement at the platelet. Under perfect conditions, a dislocation loop of
170 Burgers vector $\frac{1}{2} a_0[011]$ will form and the additional (004) layer will be introduced in the process.
171 Based on the results from previous calculations (Hirsch et al. 1986b) it was proposed that the
172 increase of the dislocation with Burgers vector would be accompanied by the release of vacancies
173 that are subsequently trapped at A centers, resulting in relatively mobile VN_2 defects (H_3 center,

174 Mainwood 1994). In our study, platelet peaks are absent from samples with luminescent pinpoints
175 throughout the diamond under grayscale CL, implying that the platelet peak has been degraded
176 completely. TEM images revealed that these luminescent pinpoints are associated with dislocation
177 loops ranging in tens to hundreds of nanometers. The natural dislocation loops are comparable in
178 shape and size with man-made dislocation loops produced from platelet degradation (Evans et al.
179 1995). Although the reason for their luminescence under CL is still unclear, the vacancy-related
180 defects likely play a role. In our previous study, a zero phonon line (ZPL) at 490.7 nm was
181 observed on those discrete features, a sign that the ZPL at 490.7 nm is associated with dislocation
182 loops. ESR results show that the ZPL at 490.7 nm correlates with the signal from dangling bonds
183 in the dislocation cores (Nadolinny et al. 2009). Photoluminescence mapping results indicate that
184 the peak area ratio of the line at 490.7 nm positively correlates with that at 496 nm (H₄ center,
185 N₄V₂ defect, Mainwood 1994), but no correlation with H₃ defects is seen (Gu and Wang 2018).
186 The presence of the ZPL at 490.7 nm associated with the luminescent pinpoints would imply that
187 the dislocation loops developed from platelet degradation are associated with dangling bonds and
188 vacancy- related defects. The luminescent pinpoints are relatively dense in the CL image,
189 indicating that dislocation loops are well developed throughout the diamond. They may potentially
190 scatter light and cause a hazy appearance in their host diamonds. However, those dislocation loops
191 might not be recognized under the microscope due to their lack of three-dimensional geometry.

192 The presence of nano-inclusions was also found to be responsible for the milky nature of
193 type IaB diamonds studied. Their octahedral or elongated morphology and relatively large size (up
194 to 200 nm) make them easier to be seen as cloudy areas of pinpoints under the microscope.
195 Previous studies reported that man-made negative inclusions can be created by platelet degradation
196 (Evans et al. 1995). Those inclusions, called voidites are mostly less than 10 nm in size and are

197 usually found on the dislocation loops (Evans et al. 1995). Voidites have been considered by-
198 products of platelet degradation. According to equation (1), the formation of an additional (004)
199 layer during the development of dislocation loops will create the same number of vacancies as
200 there are atoms in the plane. Since the atomic density of the voidites is about a half of the diamond
201 lattice, the number of vacancies produced during the conversion of platelets by artificial heat
202 treatment is roughly equal to the number required to form the voidites observed by Evans et al.
203 (1995). However, the nano-inclusions observed in natural samples (Kaminsky et al. 2013, 2015)
204 are much larger than those produced experimentally by platelet degradation. In recent studies of
205 Brazilian diamonds from the Juina area (Rudloff-Grund et al. 2016), a distinctly bimodal size
206 distribution of the nano-inclusions was observed. Whereas the suite of octahedral nano-inclusions
207 is generally around 20–30 nm, the larger nano-inclusions were up to 150–200 nm and mostly
208 elongated. In our studies, the elongate inclusions were found to be mostly small (around 20–30
209 nm) like the case of the octahedral inclusions, but occasionally large (up to 200 nm). Those nano-
210 inclusions can present independently and are usually located at a distance from dislocation loops,
211 implying their syngeneic origin (Kaminsky et al. 2013, 2015), i.e. the fluid was directly trapped
212 during the formation of the host.

213 The materials that precipitated within nano-inclusions (negative crystals) in type IaB
214 diamonds have been identified mainly by HRTEM and Raman spectroscopy in previous studies
215 (Barry 1986; Luyten et al. 1994, Rudloff-Grund et al. 2016; Navon et al. 2017). The *d*-spacings
216 obtained from the lattice fringes on the HRTEM images of precipitates suggest a few candidates
217 for the crystalline phases. One of them is a solid NH₃ phase having a *d*-spacing of about 2.4 Å,
218 which compares well with that of a cubic NH₃ phase compressed to 3.16 GPa ($d_{200} = 2.38$ Å)
219 (Barry 1986). Although a much larger fringe of 1.41 nm was observed in nano-inclusions in Juina

220 diamond, it might be caused by the lattice of the stressed diamond itself (Rudloff-Grund et al.
221 2016). In another recent study on Juina diamond (Navon et al. 2017), the d -spacings (2.4 and 2.5
222 Å) obtained from the lattice fringes were interpreted to be derived from a tetragonal γ -N₂ phase
223 (Schuch and Mills 1970) with unit cell dimensions of $a = 3.66$ Å and $c = 4.98$ Å, whereas the
224 observed intersection angles did not match with the cubic symmetry. Raman spectroscopy showed
225 a sharp peak at ~ 2355 cm⁻¹ and a weaker peak at ~ 2367 cm⁻¹, which can be assigned to ν_2 and ν_1
226 vibrational modes of the tetragonal δ -N₂ phase, respectively. The residual pressure estimated from
227 the Raman shift obtained from the inclusion was about 10.9 GPa (Navon et al. 2017). In our study,
228 the d -spacings obtained from HRTEM images are 2.39 Å and 2.63 Å, which deviate slightly from
229 the results of Navon et al. (2017) but are similar to the results of Luyten et al. (1994) who reported
230 d -spacings of 2.40 and 2.64 Å. Luyten et al., (1994) proposed a tetragonal crystal structure for the
231 non-equilibrium modification of the solid N₂ phase found in their study. However, we found that
232 the d -spacings obtained in our study are very close to the disordered cubic δ -N₂ phase ($d_{(1-20)} =$
233 2.63 Å and $d_{(21-1)} = 2.40$ Å) observed at 9.5 GPa and room temperature by X-ray diffraction in a
234 diamond anvil cell (Hanfland et al. 1998). Therefore, it is likely that the material filling the
235 octahedral nano-inclusions is a cubic δ -N₂ phase under compression to an equivalent high pressure.
236 This residual pressure (~ 9.5 GPa) held in the inclusion is comparable to that (10.9 GPa) reported
237 in the previous study (Navon et al. 2017), suggesting that the inclusions might have been trapped
238 under similar P - T conditions.

239 In addition to the irregular diamonds, milkiness is also observed in type IaB diamonds with
240 sharp platelet peaks, that have not undergone complete platelet degradations. Those diamonds
241 show a poly-crystalline texture composed of elongated crystals of 50–200 μm , in which the light
242 passing through is scattered at the grain boundaries resulting in the milky appearance. This

243 microstructure is comparable to that shown by a spherulitic polycrystalline diamond called
244 “ballas”, which consists of columnar to fibrous crystals radiating from a core (DeVries and
245 Robertson 1985; Lux et al. 1997). The elongation direction of the crystals in ballas was observed
246 by X-ray diffraction to be [110] (Trueb and Barrett 1972), which is indeed identical to the case of
247 the milky type IaB diamonds under investigation. The formation of ballas has been explained by
248 the “hailstone model”, which requires a high under-cooling condition and/or a low diffusion rate.
249 According to this model, the extremely fast quenching rate due to impurity enrichment will lead
250 to a passive boundary layer that will reduce the diffusion rate to stimulate a radial (spherulitic)
251 growth of the crystals (Lux et al. 1997). Besides, the occurrence of such spherulitic polycrystalline
252 diamonds is likely restricted to the oldest parts of the continental crust (cratons, generally >2.0 Ga)
253 (Haggerty 2014), and the long resident time would allow nitrogen to be fully aggregated. Based
254 on the hailstone (rapid growth) model, their formation might take place as a result of the interaction
255 of hot fluids with cooled materials which enables rapid diamond growth due to significantly
256 undercooling environments. If they stay in a relatively stable mantle condition for a long geological
257 time after the initial formation, it could lead to the aggregation of nitrogen forming the B center.

258

IMPLICATIONS

259 Our comprehensive observations by CL, TEM and EBSD suggest that the dominant
260 dislocation loops, nano-inclusions and/or the characteristic grain boundaries formed by radiating
261 fibrous crystals are essential origins for milky features shown by the type IaB diamonds studied
262 here. Detailed studies of the relationship of their inner structures with optical scatterings would
263 provide more insights into the mechanism of their coloration and even find more factors that could
264 contribute to their milkiness. Although our study is limited to type IaB diamonds, we expect that

265 other diamonds with similar textures would also have a milky appearance. The milky type IaB
266 diamonds studies here are one of the major sources of our knowledge on volatiles in diamond and
267 have significant geological implications. The precipitate found in octahedral nano-inclusions
268 (negative crystals) in this study can be explained by a disordered cubic δ -N₂ phase under
269 compression to ~10 GPa. Similar high residual pressures were also reported in nitrogen inclusions
270 in “super-deep” diamonds from Juina, Brazil (Navon et al, 2017). The authors estimated the *P-T*
271 conditions of equilibrium between the inclusions and the host diamond using equations of state at
272 mantle geotherm temperatures and concluded that the inclusions are derived from the deepest part
273 of the mantle transition zone at pressures of ~22 GPa. Our results suggest that the present type IaB
274 milky diamonds containing abundant nano-inclusions may also have a very deep origin. Besides,
275 our photoluminescence (PL) results (Gu et al., 2018) revealed that the dominant defects in many
276 milky type IaB diamonds are mainly associated with plastic deformation, which is not inconsistent
277 with a deep origin. In addition, milky features are also found in poly-crystalline type IaB diamonds
278 composed of radiating fibrous crystals similar to ballas. Since the formation of such polycrystalline
279 diamonds likely requires a large driving force such as under undercooling or supersaturated
280 conditions, the milky polycrystalline diamonds may have formed in deep geological settings where
281 the close interaction between C-H-O fluid and crustal/mantle rocks or between two different fluids
282 occurred.

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284

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391 **TABLE 1.** Summary of milky type IaB diamond samples analyzed in this study

Sample #	Carat weight	Clarity grade	Platelet*	General observations	Clarity description	CL features
110208637198 ^a	2.02	SI ₂	-	Hazy	Additional clouds are not shown	Pinpoints
890000048675 ^a	2.93	SI ₂	-	Hazy	Clouds are not shown ^d	Oriented pinpoints
880000140049 ^a	1.35	I ₁	-	Cloudy/hazy zones	Clouds are not shown	Pinpoints in growth zone
110208742760 ^a	3.34	I ₁	-	Cloudy/hazy zones	Additional clouds are not shown	Pinpoints in black area
110208618119 ^a	100.87	-	-	Hazy	Clouds are not shown	Pinpoints
890000102598 ^a	5.55	I ₁	1367.7	Hazy	Clouds, pinpoints are not shown	Pinpoints in black area
110208773017 ^b	0.28	SI ₂	-	Cloudy/hazy zones	Clouds and hazy area	Pinpoints in black area
110207975362 ^b	0.19	-	-	Hazy	Clouds and hazy area	Pinpoints in black area
110207975359 ^b	0.25	-	-	Cloudy	Clouds	-
110207975361 ^b	0.30	-	1367.7	Cloudy	Clouds	-
110207975363 ^b	0.33	-	1367.7	Cloudy	Clouds	-
110207975364 ^b	0.44	-	1361.2	Cloudy	Clouds	Granular texture
0927 ^c	0.63	-	-	Cloudy/hazy zones	Clouds and hazy area	Pinpoints in hazy area

392 * Values are given as the FTIR peak position of the platelet defect in wavenumbers (cm⁻¹).

393 ^a Faceted diamonds; ^b those faceted diamonds were cut into plates; ^c diamond rough.

394 ^d “Clouds are not shown” means the definite outlines of the clouds cannot be detected under 10×
 395 loupe, while the area looks hazy/not fully transparent.

396

397 **Figure captions**

398 **FIGURE 1.** Images of all milky type IaB diamonds studied in this paper. Caret weights of each
399 faceted diamonds are given, while images of these samples are not to scale. Microscope images
400 on the right side show typical hazy area in sample 110208618119 under 200× magnification, and
401 cloudy area (indicated by red arrow) in sample 0927 under 100× magnification.

402 **FIGURE 2.** Typical grayscale CL images of hazy diamonds, which show luminous pinpoints or
403 short linear defects. (a) CL image of sample 110208618119. (b) CL image of sample
404 890000102598, size of pinpoints and short lines ranging from a few micros to ~15 micros. (c) CL
405 image of sample 890000048675. Sets of pinpoints form parallel lines. (d) CL image of sample
406 880000140049. Pinpoints distribute in growth zones. (e) CL image of sample 110208773017. (f)
407 CL image of sample 110207975362. Pinpoints can be observed in growth zones.

408 **FIGURE 3.** Typical TEM images of dislocation loops in hazy diamonds (a, b) and nano-inclusions
409 in cloudy diamonds with both octahedral and elongated shapes (c–f). (a) dislocation loop in
410 diamond sample 110208773017. (b) “coffee bean” contrast at the edge of dislocation loop in
411 diamond sample 110207975362. (c) a nano-inclusion with a rounded outline and a rhombic
412 projection inside in sample 110207975359. (d) large octahedral (~100 nm) and small elongated
413 (~30 nm) nano-inclusions in sample 110207975361. (e) large elongated inclusion ~200 nm with
414 octahedral inclusions in sample 110207975363. (f) a group of elongated nano-inclusions with
415 relatively small size below 50 nm.

416 **FIGURE 4.** HRTEM image of an octahedral inclusion measuring ~50 nm. Faint fringes can be
417 observed in two directions (6 to 12 o’clock and 3 to 9 o’clock) in (a) and (c). Corresponding FFT
418 images with *d*-spacings calculated from each pattern are given in (b) and (d).

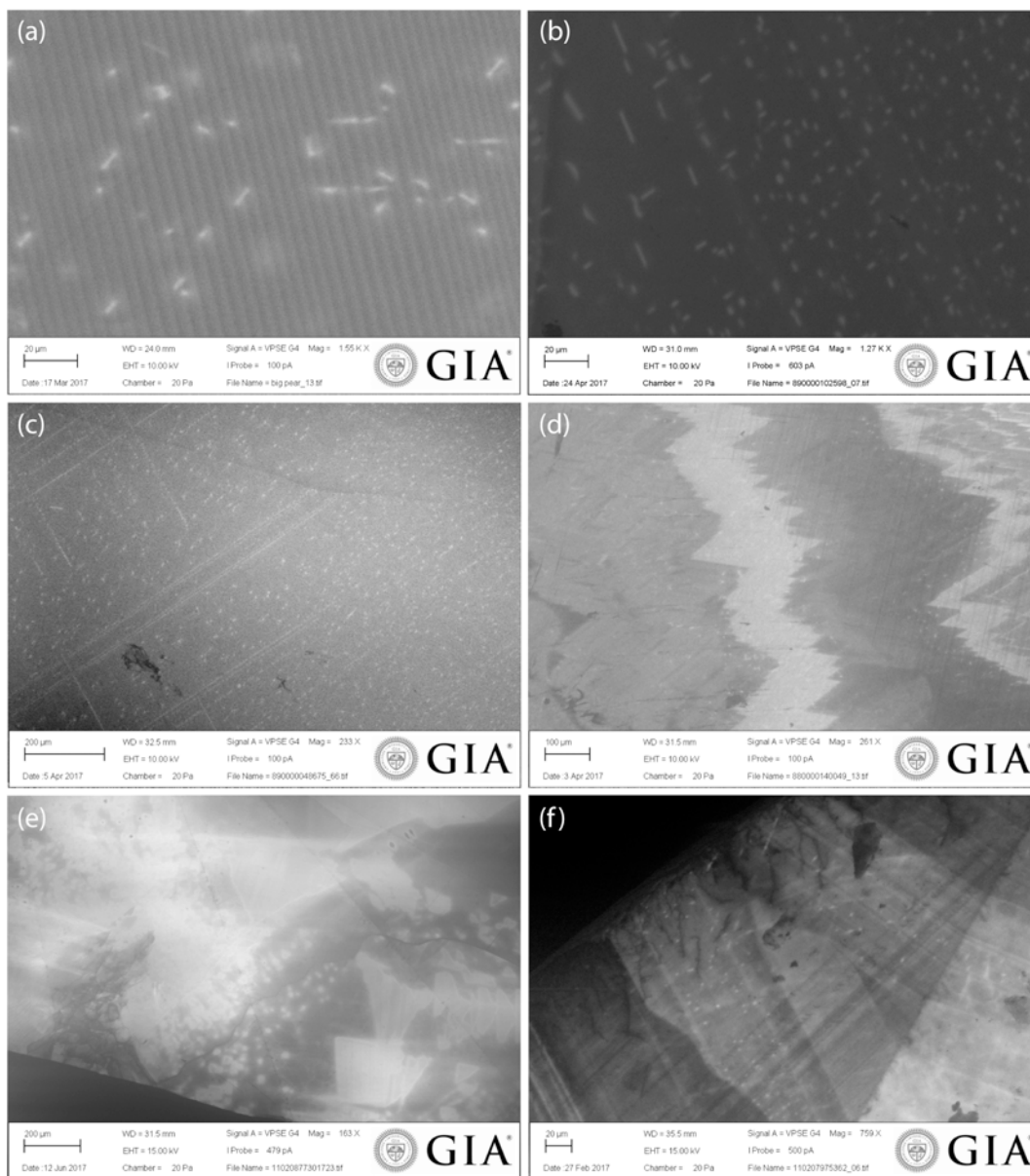
419 **FIGURE 5.** EBSD characteristics of polycrystalline diamond: (a) grain boundary map generated
420 by indexing diffraction lines detected from each Kikuchi pattern (b) artificial colors based on
421 different orientations.

422 **FIGURE 6.** Pole figures from the mapped area of polycrystalline diamond, based on 114,642 data
423 points, showing that the preferred orientation of the poly-crystals is along the $\langle 110 \rangle$ direction.



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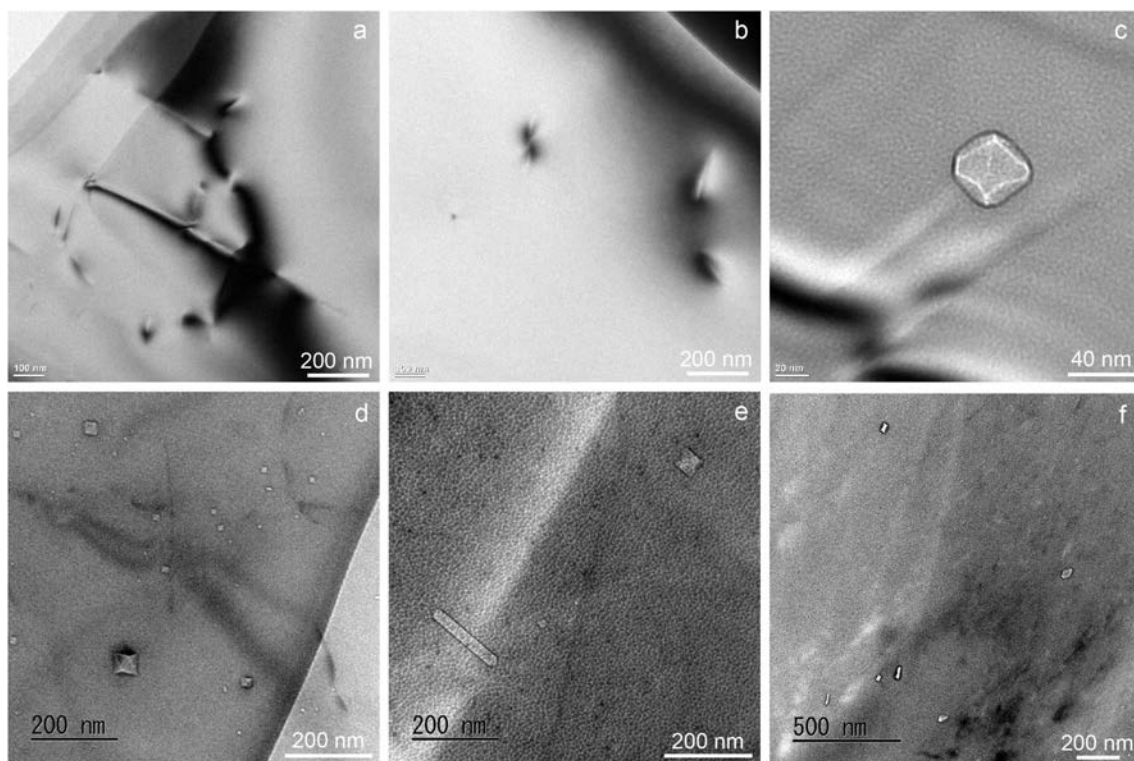
FIGURE 1.



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FIGURE 2

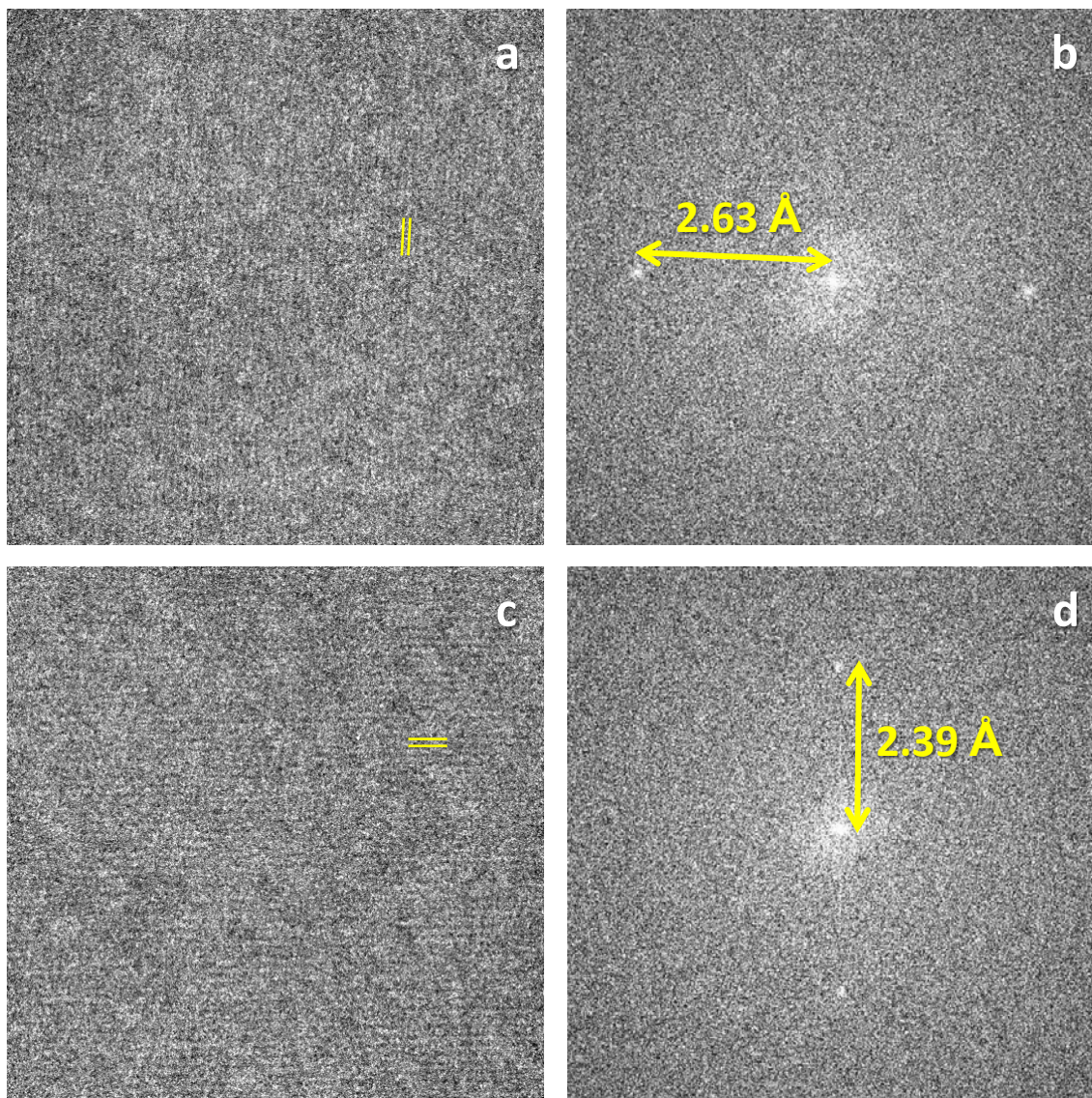
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431 **FIGURE 3**

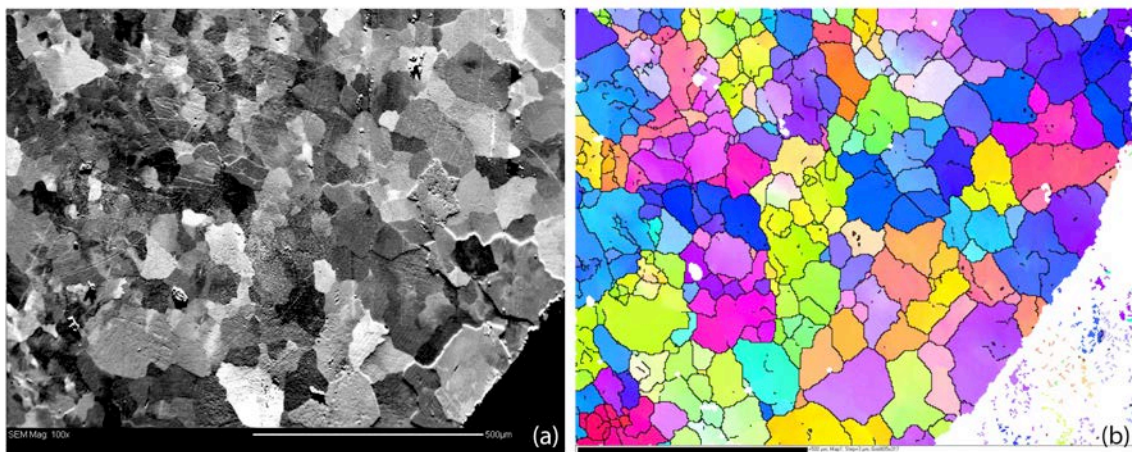
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434 **FIGURE 4**

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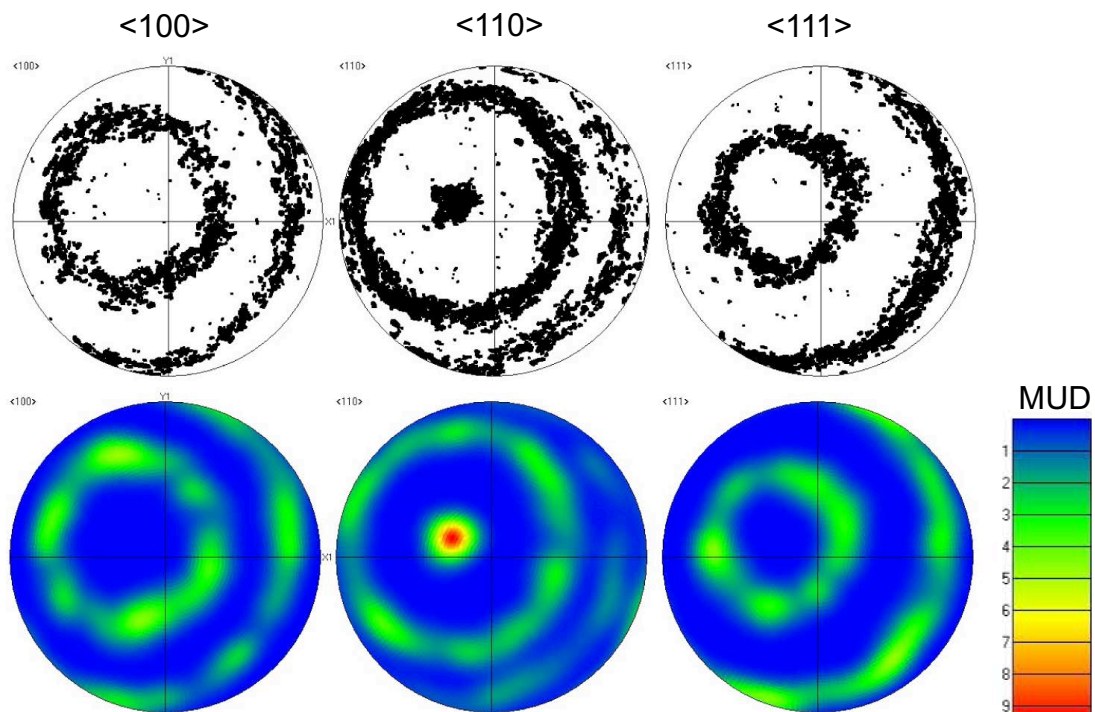


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437 **FIGURE 5**

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441 **FIGURE 6**

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