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1 ***In situ* X-ray diffraction of silicate liquids and glasses under dynamic and**
2 **static compression to megabar pressures**

3 Guillaume Morard^{a,b}, Jean-Alexis Hernandez^{c,d}, Marco Guarguaglini^c, Riccardo
4 Bolis^c, Alessandra Benuzzi-Mounaix^c, Tommaso Vinci^c, Guillaume Fiquet^a,
5 Marzena. A. Baron^a, Sang Heon Shim^e, Byeongkwan Ko^e, Arianna E.
6 Gleason^{f,g}, Wendy L. Mao^f, Roberto Alonso-Mori^g, Hae Ja Lee^g, Bob Nagler^g,
7 Eric Galtier^g, Dimosthenis Sokaras^g, Siegfried H. Glenzer^g, Denis Andrault^h,
8 Gaston Garbarinoⁱ, Mohamed Mezouarⁱ, Anja K. Schuster^j and Alessandra
9 Ravasio^c

10 ^a Sorbonne Université, Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, IMPMC,
11 Museum National d'Histoire Naturelle, UMR CNRS 7590, 4 Place Jussieu, 75005 Paris, France.

12 ^b Univ. Grenoble Alpes, Univ. Savoie Mont Blanc, CNRS, IRD, IFSSTAR, ISTERre, 38000 Grenoble,
13 France

14 ^c Laboratoire pour l'Utilisation des Lasers Intenses (LULI), Ecole Polytechnique, CNRS, CEA, UPMC,
15 91128 Palaiseau, France

16 ^d Center for Earth Evolution and Dynamics, University of Oslo, Norway

17 ^e School of Earth and Space Exploration, Arizona State University, Tempe, Arizona, USA

18 ^f Geological Sciences, Stanford University, Stanford, CA, USA

19 ^g SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA

20 ^h Université Clermont Auvergne, CNRS, IRD, OPGC, LMV, Clermont-Ferrand, France

21 ⁱ European Synchrotron Radiation Facility, ESRF, Grenoble, France

22 ^j Helmholtz-Zentrum Dresden Rossendorf, Bautzner Landstr. 400, D-01328 Dresden, Germany

23 **Abstract**

24 **Properties of liquid silicates under high pressure and high temperature conditions are**
25 **critical for modeling the dynamics and solidification mechanisms of the magma ocean in**
26 **the early Earth, as well as for constraining entrainment of melts in the mantle and in the**
27 **present-day core-mantle boundary. Here, we present *in situ* structural measurements by**
28 **X-ray diffraction of selected amorphous silicates compressed statically in diamond anvil**
29 **cells (up to 157 GPa at room temperature) or dynamically by laser-generated shock**
30 **compression (up to 130 GPa and 6000 K along the MgSiO₃ glass Hugoniot). The X-ray**
31 **diffraction patterns of silicate glasses and liquids reveal similar characteristics over a**
32 **wide pressure and temperature range. Beyond the increase in Si coordination observed**
33 **at 20 GPa, we find no evidence for major structural changes occurring in the silicate**
34 **melts studied up to pressures and temperatures exceeding Earth's core mantle**
35 **boundary conditions. This result is supported by molecular dynamics calculations. Our**
36

37 **findings reinforce the widely-used assumption that the silicate glasses studies are**
38 **appropriate structural analogs for understanding the atomic arrangement of silicate**
39 **liquids at these high pressures.**

40

41 *Significance*

42 *Understanding the structural changes silicate melts undergo over the pressure-temperature*
43 *range of the Earth's mantle has been a major, longstanding challenge in the geosciences.*
44 *Experimental studies are extremely difficult, due to required temperatures exceeding 4000 K*
45 *needed to melt silicates over megabar pressures. To overcome this issue, novel laser-driven*
46 *shock experiments combined with X-ray Free-Electron Lasers were performed to provide*
47 *nanosecond resolution on silicate structural transformations. By comparison with statically*
48 *compressed diamond-anvil cell experiments at ambient temperature, a common high-pressure*
49 *structural evolution of glasses and liquid silicates was revealed. This supports the concept*
50 *that silicate glasses of dominant mantle composition are suitable structural analogues for the*
51 *corresponding liquids at these pressures.*

52

53 **Main text**

54 **Introduction**

55 Terrestrial planet interiors have silicate-based mantles and iron-rich cores. It is
56 generally accepted that this structure is inherited from early differentiation stages occurring in
57 concert with radiogenic decay of short-lived nuclides and multiple shock events inducing
58 temperatures high enough to sustain a global magma ocean. Hence, the physical properties of
59 liquid silicates over a wide pressure-temperature range are crucial for understanding early
60 differentiation events that occurred in Earth and other rocky planets (1). Those properties,
61 especially the density, control the extent over which silicate melts can be stably entrained in
62 the mantle (2). In addition, liquid silicates exist in our present-day upper mantle (3–5) and
63 potentially at the core-mantle boundary (CMB) (6, 7). The exact composition and origin of
64 molten or partially-molten domains in the deep mantle remain enigmatic. Direct
65 measurements of molten mantle material properties to relevant pressures and temperatures are
66 required to constrain these geodynamic processes (8, 9) as well as candidate materials
67 contributing to mantle heterogeneities (10).

68 MgSiO₃ is the end member composition for the most abundant mineral of the Earth's
69 mantle, i.e. bridgmanite (11). Liquid MgSiO₃ is arguably the closest proxy for liquid silicates
70 in deep Earth. Its simple chemical composition includes two important features of liquid
71 silicates at ambient pressure, with SiO₄ building blocks and octahedral Mg cation sites. In
72 crystalline MgSiO₃, the pressure-induced transition to bridgmanite above 23 GPa is
73 accompanied by an increase in coordination, from 4- to 6-fold, of Si sites by O atoms. Such
74 dramatic atomic reorganization may also affect the properties of the corresponding liquid (12,
75 13).

76 The experimental study of liquid MgSiO₃ has been a long-term challenge, as MgSiO₃
77 melts at extremely high temperatures even at moderate pressures (melting temperature
78 exceeds 4000 K at 40 GPa (14–16)). Therefore, MgSiO₃ glass has been extensively studied as
79 a possible structural analog for the liquid state at high pressures (13, 17–19). Vibrational
80 spectroscopy identified structural changes (13, 19–21) which were confirmed by *in situ* X-ray
81 diffraction (XRD) studies (22, 23). This led to early constraints on the density of mantle melts
82 at CMB conditions (18). However, the validity of the liquid/glass structural analogy has been
83 a longstanding question. In order to probe lattice-level structural changes of liquid silicates,
84 XRD from laser-driven shock compression with nanosecond resolution were collected with
85 femtosecond X-ray pulses from a Free Electron Laser. These results were compared to static
86 compression experiments and Molecular Dynamics simulations.

87

88 **Results**

89 We performed *in situ* XRD measurements on three silicate glasses: MgSiO₃,
90 Mg₃Si₂O₇, and SiO₂. XRD patterns were collected up to 160 GPa at room temperature under
91 static compression using diamond-anvil cells (DACs) and up to 130 GPa and ~6000 K under
92 dynamic compression. Dynamic compression experiments reached the liquid phase along the
93 principal Hugoniot. Integrated XRD patterns for MgSiO₃ glasses measured under dynamic
94 and static compression, as well as synthetic spectra obtained from molecular dynamics (MD)
95 simulations, are shown in **Figure 1**. Similar features were observed for these different
96 approaches as well as for previous results from large volume experiments (17) (**Figure 1**).
97 Amorphous MgSiO₃ displays a first large diffuse feature at 20 nm⁻¹ (Q1) below a few GPa;
98 this is clearly visible in the static compression data at 2.9 GPa, while in the dynamic
99 experiment reference spectrum Q1 is mixed with the signal from the unshocked polystyrene
100 ablator at lower Q-values (**Figure 1**). The signal from the ablator disappears upon shock
101 compression and the recorded patterns capture contributions from both the shocked and

102 unshocked parts due to experimental geometry. Careful background subtraction allows the
103 separation of these two contributions (SI Appendix, Figure S2). A second diffuse diffraction
104 peak (Q2) arises under compression in both static and dynamic data around Q-values of 30-35
105 nm⁻¹ (Figure 1) (17, 24) and marks the transition towards a dense amorphous structure. This
106 spectral evolution is related to the transition from 4- to 6-fold oxygen coordination around the
107 Si atoms (17, 24). As the pressure is further increased, both diffuse scattering peaks shift
108 toward higher Q. The relative intensity of the second (Q2) compared to the first peak (Q1)
109 increases, as qualitatively illustrated by intensity plots (Figure 1) and quantitatively confirmed
110 by the structure factors S(Q) (SI Appendix, Figure S3).

111 Shock-compressed MgSiO₃ glass does not show evidence for recrystallization at the
112 nanosecond timescale up to 129(±35) GPa and 6000 K, well above the melting along the
113 Hugoniot (SI Appendix, Figure S5). This observation highlights a fundamental difference
114 with fused silica SiO₂ which is found to crystallize into stishovite in a few- to 100s of
115 nanoseconds (25, 26) under shock compression. The absence of recrystallization of MgSiO₃
116 glass along the Hugoniot may be related to the presence of Mg affecting the degree of
117 polymerization, thus leading to an increased stability of the glass structure at nanosecond
118 timescales. The dense disordered structures obtained in both static and dynamic compressions
119 can therefore be compared along the entire pressure range reached in the shock experiment,
120 which includes equilibrium liquid conditions. Indeed, according to previous works, melting of
121 MgSiO₃ glass samples along the principal Hugoniot is expected to occur between 90 and 110
122 GPa (15, 16) (SI Appendix, Figure S5). In our dynamic experiments, we used velocimetry
123 records via a velocity interferometer system for any reflector (VISAR) diagnostic to
124 determine the peak pressure in our shots. Four out of the six datasets presented in Figure 1 can
125 be assigned to pressures below 90 GPa, considering the existing measurements of the
126 Hugoniot of liquid MgSiO₃ from shocked MgSiO₃ glass (15, 16, 27–29) and the Hugoniot of
127 MgSiO₃ glass as calculated from density functional theory (DFT) MD simulations (30), and
128 are therefore in a glassy solid state. The two data points at 124 and 129 GPa are above the
129 melting curve and are representative of a high-pressure liquid (SI Appendix, Figure S5). To a
130 first approximation, the data show no significant difference between the diffraction of the
131 glass acquired along the Hugoniot and the corresponding liquid at similar pressure conditions.

132 The limited Q range available for dynamic compression XRD patterns precludes a
133 more detailed analysis, specifically preventing a real space analysis of the diffraction patterns.
134 This limitation was intrinsic to the experimental setup available (see Methods). The partial
135 structure factors for MgSiO₃ glass highlight the fact that this portion of the total structure

136 factor is mainly related to contributions from cation-anion (Mg-O and Si-O) bonds (22, 31). A
137 further consequence of the limited Q-range intrinsic to the dynamic experiments is the
138 inability to accurately track structural differences between statically and dynamically
139 compressed samples at the atomic level. However, differences in atomic arrangements should
140 be reflected in a change in the structure factor, with the appearance of new diffuse peaks (e.g.
141 between 2.5 and 20 GPa in [Figure 1](#)) or by a difference in the Q1/Q2 ratio. In this regard, we
142 find that the XRD signal obtained from shock compression of MgSiO₃ glasses follows the
143 same Q1/Q2 trend as the one from DAC-compressed glasses ([Figure 2](#)). Moreover, another
144 silicate glass with an Mg-enriched composition (Mg₃Si₂O₇) also follows a similar Q1/Q2
145 trend ([Figure 2](#), [SI Appendix](#), [Figure S7](#)). Our experiment also provides evidence that liquid
146 SiO₂ has a similar structure because the Q1/Q2 ratio lies on the trend found for statically
147 compressed SiO₂ glass ([Figure 2](#)). The difference in absolute position of the two features of
148 amorphous MgSiO₃ and SiO₂ samples can be attributed to the presence of Mg. Based on this
149 data, it can be stated that silicate glasses at high pressures and room temperature have
150 remarkably similar structures to their corresponding liquids at high temperatures. In addition,
151 no modification of the amorphous structure with increasing SiO₂ content (from 50%
152 (MgSiO₃) to 60% (Mg₃Si₂O₇)) was observed, which agrees with reported results of glasses at
153 ambient pressure (32).

154 The pressure dependence of the first sharp diffraction peak (FSDP) Q1 is often used as
155 a proxy for pressure induced changes in coordination number (33–36). For example,
156 correlation between pressure increase in Q1 and evolution of oxygen-packing fraction is a
157 common feature for a large variety of glasses (33). [Figure 3](#) illustrates the Q1 dependence on
158 pressure for our data, without evidence for sharp discontinuities in both the liquids and
159 glasses, indicating monotonic structural changes (24) with increasing pressure up to the liquid
160 state. In particular, the MD simulations (see Methods for details) reproduce the correlation
161 between Q1 and Q2 values found in our experiment, for both intensity distribution ([Figure 1](#))
162 and quantitative Q1/Q2 ratios ([Figure 2](#)). The simulations show a correlation between Q1 and
163 Q2 values very similar to the experimental one, and the coordination number of O around Si
164 smoothly increases from 6 to 7 (between 6.1 and 6.7; [SI Appendix](#), [Table S8](#)).

165 It is challenging to discriminate between temperature effects and structural
166 rearrangements, when comparing the pressure dependent peak positions of Q1 and Q2 under
167 static compression at room temperature and dynamic compression along the Hugoniot.
168 Therefore, peak positions are also plotted as a function of density ([Figure 3 and 4](#)). For static
169 compression, density values were extracted from recent experimental studies of amorphous

170 SiO₂ and MgSiO₃ (18, 37). For dynamic compression, they were deduced from VISAR
171 measurements and a known equation of state (see Methods). The error propagation involved
172 in this analysis results in large uncertainties (up to 30%) for the density of the shocked state.
173 The plots clearly show no significant difference in peak position between statically
174 compressed glass and high temperature disordered silicates with similar densities, suggesting
175 that no structural changes occur over the entire examined density range. This analysis can be
176 extended to the second diffraction peak Q₂, which appears above 10 GPa (Figure 4).
177 Theoretical calculations (31), indicate that Q₂ is related to the evolution of cation-anion bonds
178 and the appearance of 5- and 6-fold coordinated Si-O and Mg-O polyhedra. The pressure
179 dependence of the Q₂ position is similar for dynamic and static compression. Moreover, the
180 SiO₂ and MgSiO₃ samples show comparable Q₂ features as a function of pressure, suggesting
181 similar packing mechanisms.

182

183 **Conclusions**

184 Dynamic compression experiments allowed us to probe the structure of liquid silicates
185 with compositions relevant to the Earth's lower mantle (over 130 GPa and 6000 K). Although
186 the limited Q range does not allow for a full structural description, no evidence for major
187 structural changes between liquid and cold compressed silicate glass was found. In particular,
188 the ratio of Q₁/Q₂ measured for cold compressed MgSiO₃ glass at 90 GPa and room
189 temperature is very similar to that of liquid MgSiO₃ along the Hugoniot at 130 GPa and 6000
190 K. Similar conclusions can be drawn for liquid and statically compressed SiO₂. Therefore,
191 XRD measurements of cold compressed silicate glasses can be used to infer structural
192 properties of liquid silicates at Earth's interior conditions. Our experiments further
193 demonstrate the possibility of high-quality XRD diffraction measurements of liquid silicates
194 to previously inaccessible pressure-temperature regimes. In addition, signatures of chemical
195 reactions between the shocked liquids and the target chamber are minimized in the *in situ*
196 XRD spectra due to the nanosecond timescale of the shock compression experiment (or the
197 femtosecond probing timescale of the XFEL) (38).

198 The absence of major structural change under pressure up to ~130 GPa in liquid
199 MgSiO₃ is expected to be important for modeling partial melts at depth in the Earth. Similar
200 mechanisms are likely to occur in rocky exoplanets with Earth-like compositions (39).
201 Finally, our results indicate that combined static and dynamic datasets could be used to infer
202 silicate liquid properties. Future dynamic compression experiments combined with XFEL

203 sources are complementary tools suitable for probing liquid silicates under extreme
204 conditions.

205

206 **Methods**

207 **Target preparation and characterization**

208 Targets for shock compression consisted of a 50 μm -thick polystyrene ablator glued to
209 a silicate sample (between 35 to 90 μm thickness) that was polished on both sides, and coated
210 with 300 nm Ti on the drive laser side prior to gluing (SI Appendix, Figure S9). Glass starting
211 materials with MgSiO_3 and $\text{Mg}_3\text{Si}_2\text{O}_7$ compositions were synthesized by laser levitation in an
212 inert gas flux of argon. Fused silica starting material was manufactured by Asphera company.
213 The thickness of each sample as well as its planarity were individually measured using a 2D
214 interferometer. This characterization has been used for a relevant estimate of the mean shock
215 velocity and hence of the induced pressure.

216

217 **Static compression**

218 MgSiO_3 glass was synthesized by laser levitation of MgSiO_3 powder in an inert gas
219 flow of Ar. Flakes of the glass were placed in a pre-indented and drilled Re gasket. For the
220 lower pressure (LP) run, a gas driven membrane Le Toullec type DAC was used to compress
221 the sample up to 100 GPa. A small piece of KCl was placed on the side of the sample in order
222 to calculate pressure from the equation of state (EoS) (40). The experimental setup available
223 on the High Pressure beamline ID27 (ESRF, Grenoble, France) (41) included KB mirrors to
224 focus the X-ray beam to $3 \times 3 \mu\text{m}^2$, a MAR CCD detector to collect the diffraction pattern, and
225 a multichannel collimator slit system to reduce the inelastic X-ray contribution from the
226 diamond anvils (42, 43). High-quality data were obtained with this set-up, enabling the
227 extraction of a high Q range structure factor (SI Appendix, Figure S3).

228 The background contribution from the diamond anvils was first measured by placing
229 an empty DAC in the X-ray beam. Then the background was subtracted from the sample
230 diffraction patterns. The diffraction was normalized to obtain the structure factor $S(Q)$
231 following the previously described method (44, 45). For the high pressure (HP) run up to 160
232 GPa, a different experimental setup was used (SI Appendix, Figure S4). The background from
233 the diamonds was collected on the empty cell and removed from the raw signal. The Re-
234 gasket EoS was used to determine the pressure, which is well correlated with the EoS of KCl
235 used in the LP run (46).

236

237 **Dynamic compression**

238 High pressure and temperature conditions in the MgSiO₃ glass samples were obtained
239 via laser-driven shock compression using the two frequency-doubled Nd-glass laser beams
240 available at the Matter in Extreme Conditions (MEC) end-station of the Linac Coherent Light
241 Source (LCLS) at SLAC National Acceleratory Laboratory. Various thermodynamic
242 conditions could be generated by choosing the laser energy for a 10ns square pulse on a
243 250um focal spot, smoothed using a continuous phase plate.

244 *In situ* XRD measurements were performed using quasi-monochromatic (dE/E =
245 0.2–0.5%), 9.0 keV X-ray pulses of 60 fs duration with an average of $\sim 10^{12}$ photons per
246 pulse. The LCLS X-ray free electron laser spot size was set to 50 μm diameter, much smaller
247 than the flat compressed region so as to avoid detecting pressure and temperature gradients.
248 The measurement of uniform thermodynamic conditions during shock propagation were
249 ensured by optimizing the target design and probe timing. Example patterns collected on the
250 CSPADs of the typical diffuse scattering of amorphous silicates under high pressures and
251 temperatures are shown in [SI Appendix, Figure S8](#). The accessible energy and position of the
252 CSPAD were limited at the MEC end-station at the time of the experiment.

253 The time-resolved velocity of the reflecting interfaces in the sample were measured by
254 two Doppler interferometers (VISARs) operating at 532 nm whose VPF were 4.658 km/s and
255 1.814 km/s. For most of the targets, time-resolved particle velocity inside the sample could be
256 measured from the VISAR fringe shift if the sample remained transparent after shock loading.
257 The mean shock velocity inside the sample was instead measured from the shock arrival and
258 exit times recorded on the VISAR images. For some of the targets, the time-resolved material
259 velocity inside the rear-side lithium fluoride window was also measured to investigate the
260 shock stationarity.

261 The shock pressure was deduced by inserting the measured shock and particle
262 velocities into the Rankine-Hugoniot equations, in case they were recorded. For the shots
263 where only the shock velocity could be measured, the known principal Hugoniot curves
264 (fused silica (SESAME 7386), MgSiO₃ glass (27, 29)) were used to estimate the pressure. An
265 independent pressure estimate could be obtained by applying the impedance matching
266 technique between the sample and the lithium fluoride window. SESAME 7270 was used
267 extract the lithium fluoride Hugoniot curve. The uncertainties on pressure estimates were
268 determined via a Monte-Carlo routine which repeated the analysis 5000 times. Each analysis
269 was performed extracting the inputs from a Gaussian distribution whose mean were the

270 measured value and the standard deviation were the estimated uncertainty. The standard
271 deviation of the output distribution has been taken as the uncertainty in pressure. An extensive
272 discussion on the VISAR analysis is presented in the Supplementary information.

273

274 **Molecular dynamics simulations**

275 MD simulations were performed using the LAMMPS code (47). Pair potentials
276 constructed in Oganov et al. 2000 (48) were used to model the interactions between Mg, Si
277 and O atoms. The simulations contained a total of 112000 atoms and were performed in the
278 isobaric-isothermal (NPT) ensemble with a time step of 1 fs. The first melt structure was
279 created by heating an orthoenstatite structure at ambient pressure up to 4000 K, and
280 equilibrating the resulting melt during 100 ps. Then, we compressed and heated the system to
281 the target pressure-temperature conditions in 100 ps (SI Appendix, Table S6). The cells were
282 then equilibrated during an additional 100 ps at the considered high pressure-temperature
283 conditions. XRD intensities were then calculated using the USER-DIFFRACTION package
284 implemented in LAMMPS (49).

285

286 **Data availability**

287 The different data presented in this manuscript are available in the Supplementary
288 Information.

289

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304

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- 427
- 428

429 **Figure Captions**

430

431 **Figure 1: Diffraction patterns of compressed amorphous MgSiO₃ obtained under static**
432 **and dynamic compression, and from molecular dynamics simulations.** Top left: XRD
433 patterns for MgSiO₃ glass collected during static compression in a DAC. Full S(Q), up to 80
434 nm⁻¹, are presented in **SI Appendix, Figure S3**. Diffraction patterns for MgSiO₃ up to 160
435 GPa are presented in **SI Appendix, Figure S4**. Top right: XRD patterns collected for MgSiO₃
436 amorphous targets compressed along the Hugoniot during dynamic compression experiments.
437 The reported pressures, hence the liquid or glassy solid state of the sample, are determined
438 from VISAR measurements (see Supplementary Materials). Diffraction peaks around 24 and
439 28 nm⁻¹ come from the shocked Ti layer located between the ablator and the sample (see **SI**
440 **Appendix, Figure S1**). The signal from the cold part of the sample has been removed
441 following the procedure presented in Figure S2. There is a clear evolution toward higher Q-
442 values for the two main sample contributions (Q1 and Q2). Bottom left: XRD patterns
443 calculated from MD simulations. Details for the calculations are given in the Methods section.
444 Different pressure-temperature conditions are presented in **SI Appendix, Figure S5**,
445 corresponding to two Hugoniots representing extreme cases (50, 51). Bottom right:
446 Comparison of different XRD patterns at similar density, 4.5 g.cm⁻³. The XRD patterns
447 collected in a previous large volume experiment have been recalculated from the dataset
448 presented (17). The dashed vertical lines serve as guides for the eyes.

449

450 **Figure 2: Correlation between First (Q1) and Second (Q2) Diffraction peaks for**
451 **MgSiO₃, Mg₃Si₂O₇, and SiO₂ glasses under static and dynamic compression (SI**
452 **Appendix, Tables S1-S6).** Round symbols represent data acquired during static compression
453 using DACs (MgSiO₃ glass: with multichannel collimator (MCC) experimental set-up (43)
454 (black circles) and without (red circles); SiO₂ glass: data from literature (24, 34)). Square
455 symbols represent diffraction experiments recorded during dynamic compression (starting
456 with SiO₂ glass, MgSiO₃ glass, Mg₃Si₂O₇ glass, and MgSiO₃ enstatite crystals). In addition,
457 results from our MD simulations of MgSiO₃ glass are shown as filled blue diamonds.

458

459 **Figure 3: Position of the first sharp diffraction peak (FSDP) Q1 for MgSiO₃ and SiO₂**
460 **compressed amorphous samples as a function of density (SI Appendix, Tables S1-S6).**
461 Density of cold compressed glasses is calculated following recent experimental measurements
462 (18, 37). Density of shocked samples is calculated following VISAR measurements (see

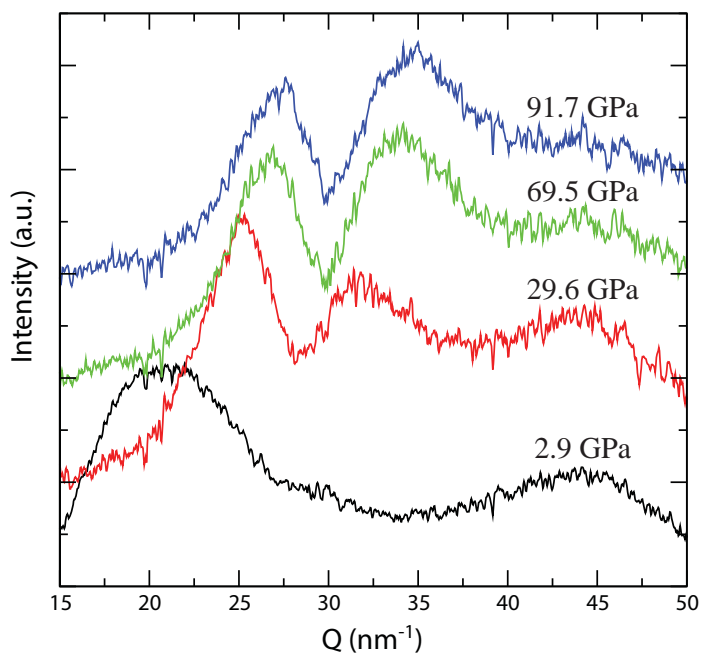
463 Methods). Round, square and diamond symbols are similar to [Figure 2](#). Inset: peak position as
464 a function of pressure. Pressures for shock compressed samples are calculated from VISAR
465 measurements (see Methods).

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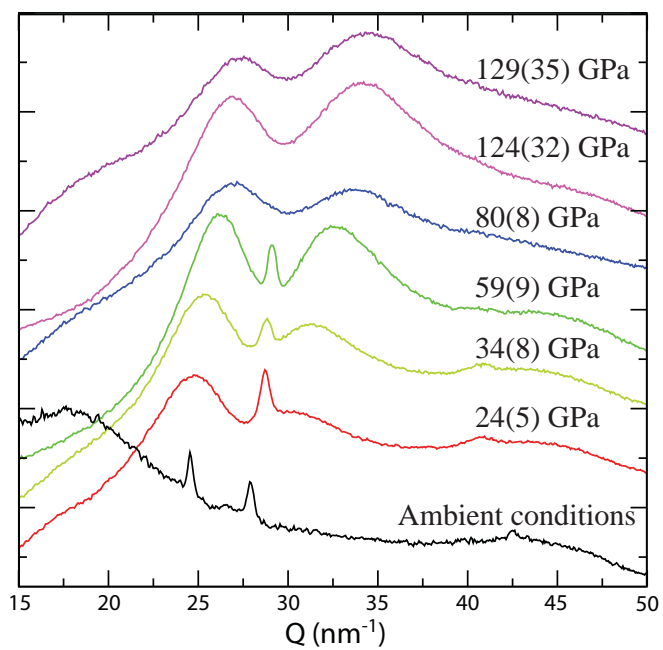
467 **Figure 4: Position of the second diffraction peak (Q2) for MgSiO₃ and SiO₂ compressed**
468 **amorphous samples as a function of density (SI Appendix, Tables S1-S6).** This second
469 peak appears in the diffraction pattern above 10 GPa. Symbols used in this figure are similar
470 to those in [Figure 3](#). Inset: peak position as a function of pressure, following the method used
471 in [Figure 3](#).

472

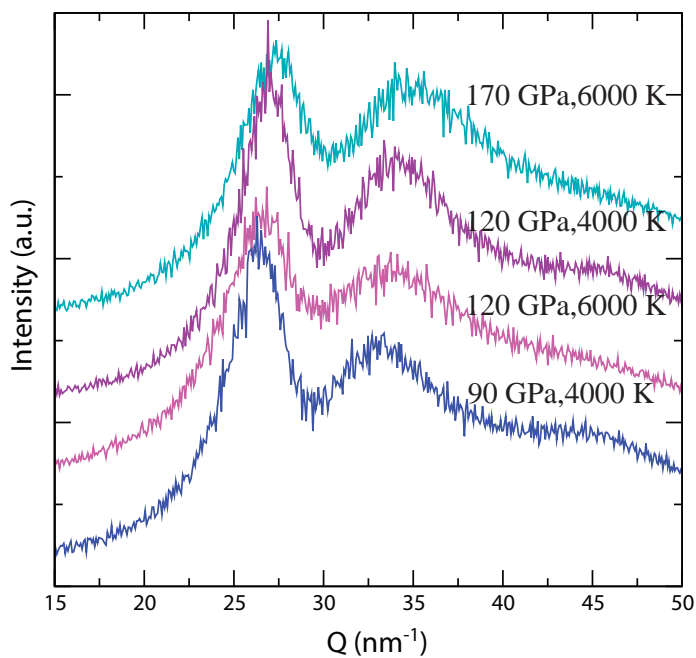
Static cold compression



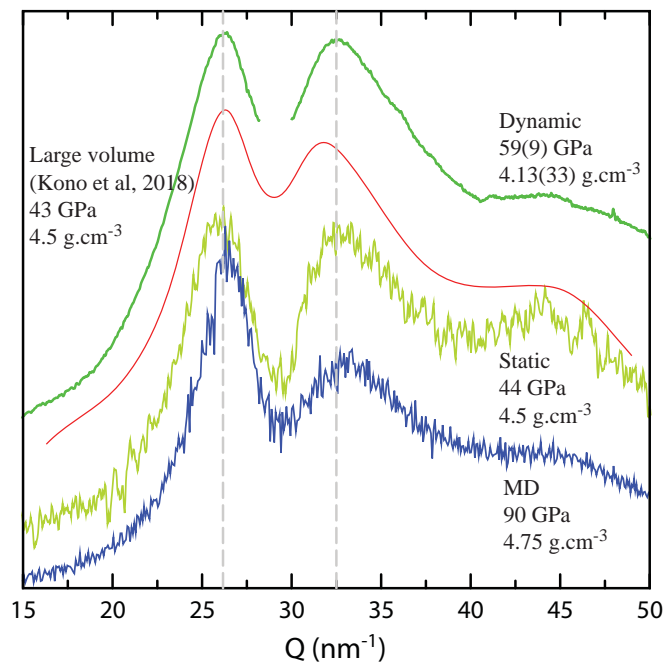
Dynamic compression

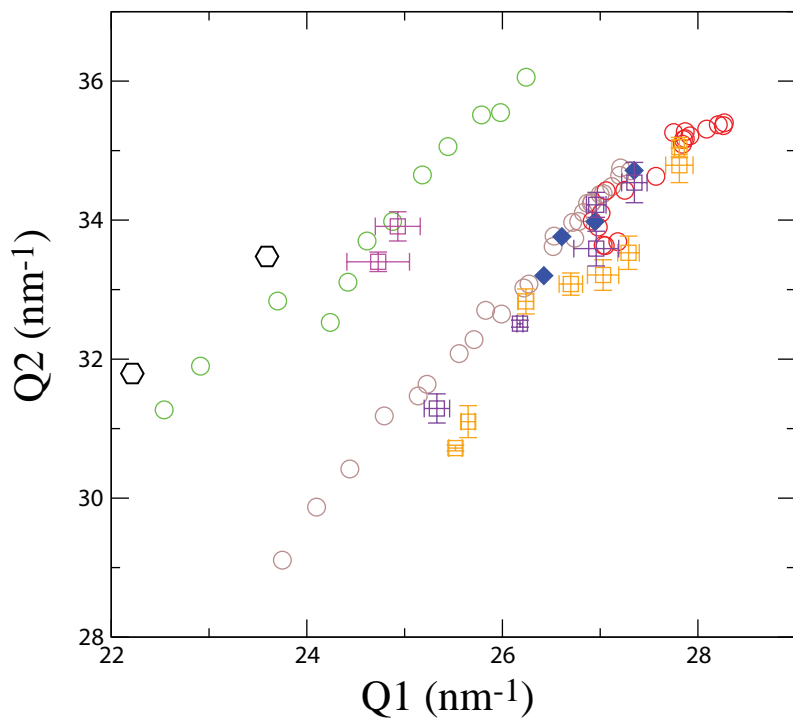


Molecular dynamics



Comparison at similar density





- ◆ MgSiO₃ glass, molecular dynamics
- MgSiO₃ glass, static DAC (Run HP)
- MgSiO₃ glass, static DAC (Run LP)
- MgSiO₃ glass, dynamic compression
- Mg₃Si₂O₇ glass, dynamic compression
- SiO₂ glass, static compression (Prescher et al, 2017)
- ⬡ SiO₂ glass, static compression (Meade et al, 1992)
- SiO₂ glass, dynamic compression

