
Supplementary information

Metastability of diamond ramp-compressed to 2 terapascals

In the format provided by the
authors and unedited

Peer Review File**Manuscript Title:** Metastability of Diamond Ramp-Compressed to 2 TPa**Editorial Notes:****Redactions – unpublished data**

Parts of this Peer Review File have been redacted as indicated to maintain the confidentiality of unpublished data

Reviewer Comments & Author Rebuttals**Reviewer Reports on the Initial Version:****Referee #1 (Remarks to the Author):**

The manuscript addresses one of old and fundamental questions of high-pressure physics - what comes beyond diamond? Several theoretical predictions based on thermodynamics are known but so far no post-diamond phase of carbon could be experimentally prepared, because of extreme difficulties in reaching ultra-high pressures in TPa range. It is therefore of great interest to explore this region of carbon phase diagram with the most advanced experimental techniques. The authors applied the state-of-the-art technology of ramp compression to reach a pressure of 2 TPa and managed to perform at this extreme pressure also XRD measurements, providing a direct structural probe. The results convincingly show that diamond at 2 TPa does not transform to another phase on the time scale of 10 ns. The authors interpret this by the existence of high enthalpic barriers related to breaking sp^3 bonds which appears the most natural explanation. The persistence of the parent FC8 phase and lack of structural transformation thus point to the necessity of reaching even higher pressures for longer time intervals as well as performing the experiment at higher temperatures in order to reach post-diamond phases. The manuscript is well written and the result is of broad interest. I recommend it to be published in Nature.

Referee #2 (Remarks to the Author):

In the work by submitted by Lazicki et al, the authors report the existence of the FC8 structure of diamond over an extensive pressure-temperature regime, through the use of dynamic ramp compression in conjunction with time-resolved x-ray diffraction. This observation, is in stark contradiction with numerical simulations where other configurations of carbon are found to be more favorable in terms of enthalpy. The author's reasoning for the significant metastability draws from previous theoretical work and also the known analogous behaviour in the graphite-diamond system, whereby strong carbon-carbon bonding can present large energetic-barriers.

The subject matter, carbon at extremes, has a history of producing high-impact papers and is quite rightly justified given its fundamental importance, finding applicability in a wide scope of scientific interests. Within this field, one of the key open questions is the ultimate stability of the diamond structure (FC8), a question Lazicki et al. attempt to resolve. The paper itself is an enjoyable read, as it is clear, well written and presented along with an extensive SI documenting the procedures involved within these experiments. Given the author's can clarify any issues that may arise during review, I feel this article would be appropriate for publication in Nature and ultimately a good addition to the literature on the topic.

In general, I find the results to be robust, taking into account the nature of these experiments, and the conclusions convincing. I have only the following concerns with the paper, which I feel should be addressed:

- (1) Although the authors give reference to another paper (ref. 31) demonstrating the background subtraction routines used in these studies. I feel this paper would be more robust and greatly benefit a presentation of the background subtraction applied to this dataset. Particularly for XRD patterns with unusual peak profiles and reduced signal-to-noise ratios, such as N180214, N170305 and N161211 in figure 2. A figure analogous to ref. 31, fig 13(c) would help the reader make a critical assessment of the data processing.
- (2) In continuation from point (1), the paper would benefit from more transparency on how peak parameters were deduced. For example what peak profile was used? Further, it would be good to present a figure demonstrating a fitting and with residuals, again this would be critical for the (111) and (220) reflections above 1.4 TPa.
- (3) Clarification on how the model for the ideal FC8 peaks are generated is required, I think this is lightly touched upon in the caption to Fig 3, but it should be further described in more detail.
- (4) Do the peak widths for the ideal powder patterns from the structural models, presented as the blue peaks in figure 2, have a physical meaning? Or are they set arbitrarily to resemble the experimental data? If so, I find this to be misleading and if its only possible to generate the peak positions from the structural model, these should be presented as bars instead.
- (5) There is a consistent mismatch between the modeled and measured intensities, particularly at large values of momentum transfer. I assume this is a consequence of not capturing the entire Debye-Scherrer rings, perhaps its possible to make a more sophisticated intensity model to account for this?
- (6) In section VI of the SI, there is the statement “(111) and (112) – which ought to reasonably appear above the background if there were a phase transformation to SC4.” To be critical, particularly of the example presented in figure S10(b), given the expected intensity of the SC4 (111) and (112) reflections I don't believe it would be readily observable from the background.
- (7) In the caption to figure 3 “...reported stress was deduced from measurements (solid markers)...” it is worth clarifying here for the general reader that VISAR measurements were used to obtain the stress.

Referee #3 (Remarks to the Author):

This well-written study concerning in situ X-ray diffraction on diamond, ramp compressed to pressures of ~2 TPa is an impressive piece of work, pushing the boundaries of X-ray diffraction measurements at extreme conditions, and will gain a significant amount of attention in the materials science, geophysical, and fusion energy science communities.

That being said, I note a few areas of improvement in the discussion of the data, request several points of clarification, and a few minor adjustments. Following said corrections, I would highly recommend this manuscript for publication.

1. Line 18 and throughout – I find the notation of crystal structure to be quite odd. I would expect FC8, or cubic diamond, to instead be cF8, SC1 to be cP1, as is more common in the literature. I note that BC8 is not in Pearson notation and instead is in a commonly used notation, therefore a change here in notation of the diamond and simple cubic structures are not necessary but

preferred.

2. Line 28 – the statement that diamond “starts melting” above 0.6 TPa is based on a study which relies on velocimetry and pyrometry and so phase transformations are inferred. Ref. 18 study does not directly observe the loss of crystallinity or the onset of melting and therefore the text should reflect that this is an inference.

3. Figure 1 – the authors should plot $F_{TQ} = 0.9$ as that is the value discussed in the text instead of $F_{TQ} = 1$

4. Lines 64 and 168 – the authors mention use of a “slurry” (supplementary information) target composed of a mix of epoxy and diamond powder. Due to the significant amount of epoxy in the sample – 47% by packing fraction – one would expect that due to the significant impedance mismatch between the epoxy and the diamond crystals that a very different compression path would be followed by these targets. I have not found a discussion on this within the manuscript. What effect does the presence of the epoxy have on the temperature or pressure reached for the same ramp pulse, if any? A discussion on the different behavior of the epoxy targets and the microcrystalline targets is needed.

5. Additionally, the authors should specify which diffraction patterns in Fig. 2 correspond to an epoxy target and which correspond to microcrystalline.

6. Furthermore, what is the starting crystallite/grain size of the microcrystalline samples? What is the spread in grain size? Are they CVD diamond or manufactured by another method?

7. Line 72 – do the image plates have any filtering for e.g. the ablation plasma?

8. Figure 2. Following point 4. Which diffraction patterns correspond to which target type explicitly. Why is the texture of the diamond arcs in the top right image significantly different from the other two image plates? It would be useful if the authors describe the significant differences in texture that are shown in this figure. Is the texture at 2 TPa related to target or related to proximity to the melting line?

9. Figure 2 – for the top two azimuthal integrations there appear to be peaks below 4 inverse angstroms (below the identified (111) reflection). What are these reflections due to? The authors should mark them with a symbol and identify them in the text.

10. Figure 2 – topmost line out. I do not find the presence of the (220) reflection here convincing as there are other features of the same intensity that are not identified. Can the authors show a smaller region of integration showing this? Furthermore, there appears to be a lot of structure around the (111) reflection and I have difficulty believing that the pattern is due to the presence of a single peak. The authors should describe this pattern more clearly since it is arguably the most important pattern in the manuscript, and clearly identify all peaks present in the main manuscript.

11. Furthermore, have the authors considered the possibility of a mixed phase of cF8 and BC8 to occur at the highest pressures? Mixed phases have been observed following shock and ramp compression of many systems. Of particular relevance to the discussion in this manuscript. Mixed phases between cF8 silicon and its high-pressure phase have been predicted by Moggi et al. *Physics Review B*, 89, 064104 (2014) and observed by McBride et al. *Nature Physics*, 15, 89 (2019). Can the authors clarify why they have not considered the presence of more than one phase, or include a discussion concerning this topic? How do the authors exclude a mixed of cF8 and a high pressure solid, or a mixture of cF8 and the high pressure liquid?

12. Line 115 – missing reference.

13. Figure 3 – The authors state “Reported stress was deduced from measurements (solid markers), or simulations (open markers).” More details on the simulations are required, or direct reader to appropriate section of the main manuscript or supplementary information.
14. Discussion beginning line 110 onwards on temperature – I do not find the argument that solid diffraction is observed therefore the compression path is best described by $F_{TQ} = 0.5$ entirely convincing. Due to the significant discrepancies in the predicted melting lines of diamond – as shown in Fig. 1, and the fact that no experimental studies have directly observed the onset of melting, it is not convincing to me that F_{TQ} could not lie in the region 0.5-0.9. Can the authors clarify this point in more detail as to why $F_{TQ}=0.5$ is the correct path. Furthermore the observation of solid diffraction indicates only that part of the target is solid, can the authors explain how they are certain there is not solid-liquid coexistence at 2 TPa? Eggert et al. Nature Physics, 6, 40 (2010) note there is expected solid-liquid coexistence along the melting line from their inferences from visar and SOP. Without a further constraint it seems only clear that the compression path lies between the two bounds stated. Another note is that both suggested paths lie above the expected sensitivities of SOP. Can the authors comment on why this was not fielded as it would provide an additional constraint.
15. Line 158 what type of phase plates were used?
16. Line 166 What is the estimated photon flux that reaches the target through the pinhole?
17. Line 178 Authors should include MgO as a tamper here
18. Information on the velocity sensitivity of the visar system should be included in the supplementary information.
19. Figure 4 – I don't find this figure particularly illuminating. I would suggest that instead the authors move figure 3 to figure 4 and instead include some of thei velocimetry data in the main text of the manuscript as reaching a pressure of 2 TPa and performing X-ray diffraction is a remarkable achievement, showing both diagnostics up front will be very instructive.

Author Rebuttals to Initial Comments:

We thank the referees for their careful reading and good comments and questions, and for their positive recommendations. We have made the changes to the manuscript suggested by Referees 2 and 3, as detailed in the responses to the individual remarks below. We think these changes have increased the clarity and robustness of the result.

Referees' comments:

Referee #1 (Remarks to the Author):

The manuscript addresses one of old and fundamental questions of high-pressure physics - what comes beyond diamond? Several theoretical predictions based on thermodynamics are known but so far no post-diamond phase of carbon could be experimentally prepared, because of extreme difficulties in reaching ultra-high pressures in TPa range. It is therefore of great interest to explore this region of carbon phase diagram with the most advanced experimental techniques. The authors applied the state-of-the-art technology of ramp compression to reach a pressure of 2 TPa and managed to perform at this extreme pressure also XRD measurements, providing a direct structural probe. The results convincingly show that diamond at 2 TPa does not transform to another phase on

the time scale of 10 ns. The authors interpret this by the existence of high enthalpic barriers related to breaking sp³ bonds which appears the most natural explanation. The persistence of the parent FC8 phase and lack of structural transformation thus point to the necessity of reaching even higher pressures for longer time intervals as well as performing the experiment at higher temperatures in order to reach post-diamond phases. The manuscript is well written and the result is of broad interest. I recommend it to be published in Nature.

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- We have provided two additional figures in the Extended data, one illustrating an example of the background subtraction technique and data processing for shot N170305 (The same background-subtraction method and gaussian line-shape fitting were used for the other shots presented in this paper), and another figure describing the detailed signal and noise features for the 2 TPa N180214 shot, and how we went from the background-subtracted data to the lineout shown in the main manuscript (we have now updated that lineout in response to comments 9 and 10 from referee #3).

(2) In continuation from point (1), the paper would benefit from more transparency on how peak parameters were deduced. For example what peak profile was used? Further, it would be good to present a figure demonstrating a fitting and with residuals, again this would be critical for the (111) and (220) reflections above 1.4 TPa.

-In all cases, a gaussian wave profile was used, with a polynomial background. The peak fitting with residuals are shown for 2 of the 1.4TPa+ shots in the added figures. The third shot above 1.4 TPa was treated similarly but we did not make a dedicated figure.

(3) Clarification on how the model for the ideal FC8 peaks are generated is required, I think this is lightly touched upon in the caption to Fig 3, but it should be further described in more detail.

-The model ideal FC8 crystal structure includes the structure factor, multiplicity, Lorentz-polarization factor (for a flat powder sample in transmission), and an instrument broadening tuned to match the data. The peak width FWHM for these shots is between 1 and 2 degrees, which is consistent with expectations for the contributions from various sources, as described in Rygg et al. RSI (2019). The peaks widths are expected to vary somewhat from shot to shot since they are proportionate to the source size, which is the spatial extent of the emitting plasma cloud around the x-ray source and this varies slightly based on the detailed characteristics of the particular x-ray source foil and the delivered laser pulse. We do not attempt to model texture or temperature. This description has now been added to the Extended Data Figure 2 caption and referenced in the main text.

(4) Do the peak widths for the ideal powder patterns from the structural models, presented as the blue peaks in figure 2, have a physical meaning? Or are they set arbitrarily to resemble the experimental data? If so, I find this to be misleading and if its only possible to generate the peak positions from the structural model, these should be presented as bars instead.

The instrument broadening due to the finite source and collimating pinhole sizes is expected to be near 1 degree, as described in Rygg's RSI, and that's approximately consistent with our observation, but there will be some shot-to-shot variation, as described in the response to comment (3). We prefer to model the peak widths and intensities (acknowledging that some important effects have been left out - our structural model does not currently include the effects of texture or temperature, which are both obviously at play in this data) rather than just the predicted positions, to qualitatively illustrate the likelihood that a particular peak would not be observed, because its intensity would not be sufficient to rise above the noise, and because consistent peak widths also help us to rule out spurious background features which sometimes appear as peaks in a lineout.

(5) There is a consistent mismatch between the modeled and measured intensities, particularly at large values of momentum transfer. I assume this is a consequence of not capturing the entire Debye-Scherrer rings, perhaps its possible to make a more sophisticated intensity model to account for this?

We should note that the lineouts shown are averages rather than integrations (I have corrected one place in the manuscript where I mistakenly called them integrations), because of the varying azimuthal coverage. High sample temperature is one cause of the intensity mismatch between the real and ideal diffraction data at high angle, due to the Debye-Waller effect. We have not tried to include this effect in our structural models for this study, because sample texture is also playing a role in several shots where diffraction from the polycrystalline sample is overlapping with diffraction from the single-crystal diamond window, which is largely at pressure equilibrium with the sample. The limited azimuthal coverage of the higher-angle line means that the intensity of a textured line will be inaccurate. A more sophisticated model is beyond the scope of this publication, but we are indeed pursuing your idea of matching intensity and texture models to

dynamic diffraction data, to infer temperature and phase relations and transformation mechanisms across phase transitions.

(6) In section VI of the SI, there is the statement “(111) and (112) – which ought to reasonably appear above the background if there were a phase transformation to SC4.” To be critical, particularly of the example presented in figure S10(b), given the expected intensity of the SC4 (111) and (112) reflections I don’t believe it would be readily observable from the background.

We agree with the reviewer. We did not have a rigorous basis for making this statement and it has been removed. It is sufficient to consider the relative peak positions to rule out the existence of an SC4 phase.

(7) In the caption to figure 3 “...reported stress was deduced from measurements (solid markers)...” it is worth clarifying here for the general reader that VISAR measurements were used to obtain the stress.

I have made this change.

Referee #3 (Remarks to the Author):

This well-written study concerning in situ X-ray diffraction on diamond, ramp compressed to pressures of ~2 TPa is an impressive piece of work, pushing the boundaries of X-ray diffraction measurements at extreme conditions, and will gain a significant amount of attention in the materials science, geophysical, and fusion energy science communities.

That being said, I note a few areas of improvement in the discussion of the data, request several points of clarification, and a few minor adjustments. Following said corrections, I would highly recommend this manuscript for publication.

1. Line 18 and throughout – I find the notation of crystal structure to be quite odd. I would expect FC8, or cubic diamond, to instead be cF8, SC1 to be cP1, as is more common in the literature. I note that BC8 is not in Pearson notation and instead is in a commonly used notation, therefore a change here in notation of the diamond and simple cubic structures are not necessary but preferred.

We agree with the reviewer that the notation used is not standard. However, we would like to keep it as it is, to maintain consistency with the notation used in the majority of the theory work to describe the predicted high pressure structures (SC1, SC4, BC8), which motivated this study. We have now added the Hermann-Mauguin space group notation at first introduction of the structures.

2. Line 28 – the statement that diamond “starts melting” above 0.6 TPa is based on a study which relies on velocimetry and pyrometry and so phase transformations are inferred. Ref. 18 study does not directly observe the loss of crystallinity or the onset of melting and therefore the text should reflect that this is an inference.

We agree and have modified the text to read as follows: “While such high pressures can be obtained with shock compression, this highly entropic process starts melting diamond above 0.6 TPa, according to a study of changes in entropy manifested in decaying shock waves [18].”

[Redacted]

3. Figure 1 – the authors should plot $F_{TQ} = 0.9$ as that is the value discussed in the text instead of $F_{TQ} = 1$

We have made this change.

4. Lines 64 and 168 – the authors mention use of a “slurry” (supplementary information) target composed of a mix of epoxy and diamond powder. Due to the significant amount of epoxy in the sample – 47% by packing fraction – one would expect that due to the significant impedance mismatch between the epoxy and the diamond crystals that a very different compression path would be followed by these targets. I have not found a discussion on this within the manuscript. What effect does the presence of the epoxy have on the temperature or pressure reached for the same ramp pulse, if any? A discussion on the different behavior of the epoxy targets and the microcrystalline targets is needed.

We have added a section in the Methods section addressing this question. The compression path followed by the diamond crystallites embedded in epoxy is not in the end significantly different from the single crystal diamond, because the wave reverberations within the sub-micron-scale regions of diamond and epoxy are extremely rapid compared to the ~25-ns duration of the ramp drive. The effect on the epoxy matrix on the sample temperature is not well-known, but we have estimated some bounds in the Extended Data.

5. Additionally, the authors should specify which diffraction patterns in Fig. 2 correspond to an epoxy target and which correspond to microcrystalline.

We have added the numbers of the specific shots with the slurry sample to the main text.

6. Furthermore, what is the starting crystallite/grain size of the microcrystalline samples? What is the spread in grain size? Are they CVD diamond or manufactured by another method?

We have added some description of these samples to the Methods section, and a reference to a paper describing the material in further detail. We do not have detailed characterization of the spread of grain size for every individual sample.

7. Line 72 – do the image plates have any filtering for e.g. the ablation plasma?

Yes, and a more complete description of the image plate filtering has been added to the Methods section.

8. Figure 2. Following point 4. Which diffraction patterns correspond to which target type explicitly. Why is the texture of the diamond arcs in the top right image significantly different from the other two image plates? It would be useful if the authors describe the significant differences in texture that are shown in this figure. Is the texture at 2 TPa related to target or related to proximity to the melting line?

We judge that the apparent differences in texture mostly arise from the fact that most of our targets contain a polycrystalline sample and a single crystal diamond tamper, and they are largely at pressure equilibrium at the time of the experiment, so we see overlapping diffraction from both materials, to varying degrees shot to shot, depending on the random rotation of the single crystal tamper. At the highest pressures, the textured spots from the diamond window are intense

enough to rise measurably above the extremely high background on the image plate. We have now edited Figure 2 by removing the lowest-pressure image plate and added the VISAR data for the 2 TPa measurement, to highlight this breakthrough measurement. The rest of the data is summarized in Extended Data Figures 2 and 3, and a description of the texture is in the captions of those figures.

9. Figure 2 – for the top two azimuthal integrations there appear to be peaks below 4 inverse angstroms (below the identified (111) reflection). What are these reflections due to? The authors should mark them with a symbol and identify them in the text.

In the 2 TPa shot, the apparent ‘peaks’ below 4 inverse angstroms are due to scanning artifacts as well as fluorescence and shadows from the edge of the fluorescence shield and filter stacks in TARDIS box (the higher-energy x-ray source is scattered at smaller angles, closer to regions on the image plates which are frequently contaminated by such features). The apparent ‘peak’ below 4 inverse angstroms in the 1.74 TPa shot is simply part of the background noise, which is somewhat attenuated right next to the more intense peak in the background subtraction. We have slightly improved the lineout for this shot in the resubmitted manuscript, by narrowing the azimuthal angular range over which we take the average lineout from 250 degrees to 200 degrees, and by trimming off a narrow region at the edges of the top and bottom image plates where the sampling resolution is the lowest, giving very high noise. The data extraction is illustrated in the new Extended Data figure 6, and a reference to the Extended Data has been added to the figure 2 caption.

10. Figure 2 – topmost line out. I do not find the presence of the (220) reflection here convincing as there are other features of the same intensity that are not identified. Can the authors show a smaller region of integration showing this? Furthermore, there appears to be a lot of structure around the (111) reflection and I have difficulty believing that the pattern is due to the presence of a single peak. The authors should describe this pattern more clearly since it is arguably the most important pattern in the manuscript, and clearly identify all peaks present in the main manuscript.

We have added a figure to the Extended Data (Figure 7), to detail the various sources of noise and structure in the highest pressure diffraction data, and to show average lineouts over smaller regions of the image plate (we note that these are average lineouts, not integrations). We agree with the reviewer that the (111) peak as previously displayed, had a lot of structure in it related to coincident features on the image plate unrelated to the Bragg scattering. We have updated the average lineout for this shot by stitching together two narrow lineouts, rather than using one large lineout which encompassed additional noise features.

11. Furthermore, have the authors considered the possibility of a mixed phase of cF8 and BC8 to occur at the highest pressures? Mixed phases have been observed following shock and ramp compression of many systems. Of particular relevance to the discussion in this manuscript. Mixed phases between cF8 silicon and its high-pressure phase have been predicted by Mogni et al. Physics Review B, 89, 064104 (2014) and observed by McBride et al. Nature Physics, 15, 89 (2019). Can the authors clarify why they have not considered the presence of more than one phase, or include a discussion concerning this topic? How do the authors exclude a mixed of cF8 and a high pressure solid, or a mixture of cF8 and the high pressure liquid?

We do not exclude the possibility of a mixed phase (the likelihood of mixed solid and liquid is acknowledged in the discussion of uncertainties in the Methods section). We have not considered the possibility that there may be a fraction of an additional solid phase, because we do not have convincing evidence for diffraction peaks which can’t be explained by the cF8 structure. Of

course, we can't rule out the possibility that there may be a small fraction of an additional solid phase which will not diffract with sufficient intensity to rise above the background.

12. Line 115 – missing reference.

Fixed

13. Figure 3 – The authors state “Reported stress was deduced from measurements (solid markers), or simulations (open markers).” More details on the simulations are required, or direct reader to appropriate section of the main manuscript or supplementary information.

We changed the text to read: “Reported stress was deduced from VISAR measurements (solid markers), or radiation-hydrodynamic simulations (open markers)”, with a reference to a paper describing the particular software used for the simulations.

14. Discussion beginning line 110 onwards on temperature – I do not find the argument that solid diffraction is observed therefore the compression path is best described by $F_{TQ} = 0.5$ entirely convincing. Due to the significant discrepancies in the predicted melting lines of diamond – as shown in Fig. 1, and the fact that no experimental studies have directly observed the onset of melting, it is not convincing to me that F_{TQ} could not lie in the region 0.5-0.9. Can the authors clarify this point in more detail as to why $F_{TQ}=0.5$ is the correct path.

Furthermore the observation of solid diffraction indicates only that part of the target is solid, can the authors explain how they are certain there is not solid-liquid coexistence at 2 TPa? Eggert et al. Nature Physics, 6, 40 (2010) note there is expected solid-liquid coexistence along the melting line from their inferences from visar and SOP. Without a further constraint it seems only clear that the compression path lies between the two bounds stated.

Another note is that both suggested paths lie above the expected sensitivities of SOP. Can the authors comment on why this was not fielded as it would provide an additional constraint.

We agree that we don't have evidence for $F_{TQ}=0.5$ being the correct path, we simply suggest that it appears more plausible than $F_{TQ}=0.9$, within this simplified model. We have modified the main text to read:

Within this simplified model, f_{TQ} should be nearer to 0.5 for carbon to remain solid at 2 TPa (Figure 1) [Swift], if the predicted melting curve is accurate at these conditions.

The sentence which follows this one was meant to clarify that this Taylor-Quinney model is actually too simple to be expected to accurately predict the ramp-compression pathway anyway, and to suggest that more work needs to be done.

We do in fact have SOP data from all of these shots, but because the diamond loses transparency above the elastic-plastic transition, the diagnostic measures emission from the diamond free surface only. It's difficult to use a free-surface measurement to infer a bulk temperature for several reasons. The ramp has steepened to a strong shock by the time it reaches the free surface so the temperature of the tamper surface may be much different from ramped sample, and the measurement is complicated by the release to zero pressure at the free surface. Additionally, the NIF SOP diagnostic is not absolutely calibrated (temperature is generally inferred by relating the data to thermal emission from within a quartz standard placed on the target).

15. Line 158 what type of phase plates were used?

These were continuous phase plates with a ~1mm circular profile, further smoothed using spectral dispersion and orthogonal polarization. This detail has been included in the Methods section

16. Line 166 What is the estimated photon flux that reaches the target through the pinhole?

The He-alpha fluence at the sample is about 33×10^{18} photons/m², as described in section IV.A.2 of Rygg et al., Rev. Sci. Instrum. 91, 043902 (2020). This estimate has been added to the description of the laser configuration in the Methods section.

17. Line 178 Authors should include MgO as a tamper here

Done

18. Information on the velocity sensitivity of the visar system should be included in the supplementary information.

This has been added to the methods section

19. Figure 4 – I don't find this figure particularly illuminating. I would suggest that instead the authors move figure 3 to figure 4 and instead include some of their velocimetry data in the main text of the manuscript as reaching a pressure of 2 TPa and performing X-ray diffraction is a remarkable achievement, showing both diagnostics up front will be very instructive.

We have taken this advice by adding velocimetry data to Figure 2.

Reviewer Reports on the First Revision:

Referee #2 (Remarks to the Author):

The authors have quite robustly addressed my concerns and therefore I can recommend the manuscript as suitable for publication.

Referee #3 (Remarks to the Author):

This work addresses the question of what is the behavior of carbon at extreme conditions. Carbon, a fundamental system of relevance to planetary science, has many predicted high pressure forms, the direct investigation of which this study addresses. This work pushes the bounds of our knowledge of high pressure solids.

i very much appreciate that the authors have addressed all the points raised in the previous review, and have done so thoroughly. I have no additional points to raise and recommend this for publication following the amendment of the space group for cubic diamond carbon:

Line 15 should read (Fd-3m, here called FC8)