nature portfolio

Peer Review File

Data-driven Picosecond X-ray Imaging for Quantitative Plasma-induced Shock Characterization

Corresponding Author: Dr Christopher Campbell

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Communications Physics.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This manuscript presents in-situ X-ray imaging experiments visualizing weak shock wavefronts and cavitation bubbles. The results indicate the formation of the cavitations increase the density behind the shock wavefront, while such effect on the shock wave velocities is found to be minimum. The results provide new insights into the high-energy-density physics communities, and it aligns with the scope of the journal. However, some minor comments listed below needs to be addressed before acceptance.

1. The authors include "picosecond" in the title, but nothing about picosecond is mentioned in the main text. Is it indicating the pulse duration?

2. Considering the broad audience of the journal, describing what Rankin-Hugoniot model is with a few citations would be helpful. Also, it should be noted that the model is usually applied for planar shock fronts.

3. In Section IV, the authors note "λ = ~25 keV". The photon energy should be converted to the wavelength here.

4. In Section VI, the authors describe "This bubble interface expands outward from a relatively small radius (¡10 μm) and high energy density, compressing the post-shock region to a higher density." About this point, the shock front is a smooth spherical shape while the interface between the cavitation bubbles and liquid have more complicated structure. Could the non-spherical bubble-liquid interface shape affect the stress distribution/gradient in the liquid? If so, could it disturb the shock wavefront structure slightly?

5. Please add a description for the bottom image of Fig 6 in its caption.

Reviewer #2

(Remarks to the Author)
In this submission, the authors reported an experimental study of shock waves and their evolution in heptane. To perform these measurements, the authors utilized a rep-rated pulsed power driver at the Advanced Photon Source. While I agree that this work is certainly interesting, in its current state the article is not structured appropriately to clearly present its contents and does not set the work in context.

I believe that this article can be published within Communication Physics after some re-structuring. I will order my comments and questions according to the sections of the paper.

A. Motivation

It is unclear if the aim of the publication is to present a novel target, a more sensitive measurement, or a type of analysis that has not been presented before. The introduction is also missing a large amount of previous work that would set it into context.

1. I advise the authors to discuss previous rep-rated experiments at advanced light sources – these were largely experiments performed with lasers for example:

a. https://doi.org/10.1038/s41467-021-23664-1
b.
https://www.xfel.eu/sites/sites_custom/site_xfel/content/e35165/e46561/e46886/e46963/e83032/xfel_file83043/11_Roadmap_DIPOLE_Wark_eng.pdf

These are just experiments at the EuXFEL (which is pioneering multi-Hz ~100 J laser experiments) – there are certainly examples of such experiments (at a lower rep-rate) at LCLS MEC too.

2. "…we have confirmed to be sensitive to the event's associated shock with superior temporal and spatial resolution compared to prior results…"

– It would be beneficial to reference previous work that this is being compared to, or at least quote relevant numbers.

3. "… highlights the superior sensitivity of this implementation of phase contrast imaging…" – For the reader, it is necessary to mention the key difference which distinguishes this implementation of PCI from the referenced work or other common implementations.

Other minor points:
• "…direct imaging photography remains one of the most impactful and data-rich methods for learning about any physical phenomenon…" – I should highlight that there are many physical phenomena where direct imaging photography will not be of much use. I recommend that this statement be modified to something a little more down-to-the-earth.

• I recommend (but do not insist) that the authors broaden their listing of advanced light sources and user facilities beyond the United States. This might be more representative of the global scientific community – for example by including the European Synchrotron or European XFEL, which have also been active in dynamic compression science. This is particularly stark given that the ESRF has been the first synchrotron world-wide to upgrade its storage ring to achieve higher emittance. This is mentioned as an upcoming upgrade for APS ("…which will have superior brightness…") later in this manuscript, but a similar upgrade has already been done at ESRF (https://doi.org/10.1038/s42005-023-01195-z).

B. Experimental Setup

1. Is it possible to provide a reference for the size of the discharge channel (i.e. ~ 5 micron at peak current)?

2. Comparison between the peak energy per unit mass of the discharge channel and the National Ignition Facility appears to draw a quantitative connection between these two facilities. Is it possible to clarify why this number allows any connection to be made? Do the related physical effects that can be studied on your platform scale appropriately?

This needs to be clarified because, as an absurd example, an average Snickers bar has energy density per unit mass of 20 MJ/kg. This is within three orders of magnitude of your experiment but does not necessarily draw a meaningful connection.

I also believe that the value quoted for NIF, 1TW/kg, should be 1 TJ /kg.

3. Looking at Figure 1, it appears that your capacitor is 1nF in capacitance and is charged to 10 kV. This means that the capacitor holds $E_{tot} = \frac{1}{2} C V^2$ = 50 mJ of energy. In text, you state that the energy input into the discharge is ~ 100 mJ, in Figure 1 I see ~70 mJ. Both numbers appear to suggest > 100% coupling efficiency from the capacitor into the experiment. Is it possible to clarify this discrepancy?

C. Energy Balance in Post-Shock Region

1. Is it possible to explain how the total electrical energy input was calculated from the current and the voltage measurements?

2. Is it possible to provide more intuitive representation of U_{shock}? Do I understand correctly that this is a fraction of the total deposited energy that is converted into thermal energy by the shock wave produced by the discharge channel?

3. Related to previous question, given the error bars on your voltage and current measurements. Do you believe that 2.8% is a significant number, or is this number consistent with no significant excess heating?

4. Is there any context why one should care about this number? This would hopefully be clarified within the introduction.

D. Rankine-Hugoniot Shock Analysis

1. If understood correctly, your PCI analysis method assumes a cylindrical sample oriented perpendicular to the X-ray axis. How does this affect the results, especially when your shock waves are effectively spherical (radius at late times > interelectrode distance).
2. Is this "bubble effect" relevant in any other system of interest to science or engineering? For example, underwater explosions / earthquakes in marine context?

Overall, I recommend that you focus on a single message within the publication. I assumed that this manuscript was meant to present a novel approach to HEDP science, but this message was not very clear. If re-structured to clearly communicate the utility of large data sets to your analysis, I am certain the content of this manuscript can form a nice publication.

Reviewer #3

(Remarks to the Author)
The paper "Data-driven Picosecond X-ray Imaging for Quantitative Plasma-induced Shock Characterization" by Christopher S. Campbell et al. investigates shockwaves created by high-energy-density processes using phase contrast X-ray imaging (PCI) with fourth and fifth generation X-ray light sources. The paper makes significant contributions to fields such as materials science, high-energy-density plasma physics, and inertial confinement fusion by providing novel imaging data of shockwave phenomena at initiation timescales, demonstrating the advantages of phase contrast X-ray imaging, and revealing previously unobserved deviations in shockwave behavior due to cavitation interactions.
In my opinion, after improving the clarity of some figures, the paper is well-suited for publication. Please see my specific comments below:
1. In Figure 1, the labels should be enlarged to enhance readability.
2. In Figure 2, the time labels should also be enlarged and corrected to improve visibility.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have revised the manuscript accordingly. This is an interesting study I recommend to publish as is.

<u>Reviewers' comments:</u>

Reviewer #1 (Remarks to the Author):

This manuscript presents in-situ X-ray imaging experiments visualizing weak shock wavefronts and cavitation bubbles. The results indicate the formation of the cavitations increase the density behind the shock wavefront, while such effect on the shock wave velocities is found to be minimum. The results provide new insights into the high-energy-density physics communities, and it aligns with the scope of the journal. However, some minor comments listed below needs to be addressed before acceptance.

1. The authors include "picosecond" in the title, but nothing about picosecond is mentioned in the main text. Is it indicating the pulse duration?

Yes, Reviewer #1 is correct in assuming that "picosecond" is referring to the pulse duration of the synchrotron source, which defines the temporal resolution of the XPCI data presented in this work. The authors thank Reviewer #1 for pointing out that this particular aspect was not discussed in the body of the previous manuscript version; the "Motivation" section has been edited to mention the APS's picosecond resolution explicitly (pulse width ~50ps).

2. Considering the broad audience of the journal, describing what Rankin-Hugoniot model is with a few citations would be helpful. Also, it should be noted that the model is usually applied for planar shock fronts.

Per Reviewer #1's comment, the discussion introducing the Rankine-Hugoniot model has been edited to include relevant references and a bit more background discussion. Within the context of this work, the Rankine-Hugoniot model simply refers to one-dimensional conservation of mass, momentum, and energy across the shock front.

3. In Section IV, the authors note "$\lambda = \sim 25$ keV". The photon energy should be converted to the wavelength here.

The authors thank Reviewer #1 for pointing out this minor error, it has been corrected: $\lambda = \sim 50$ pm.

4. In Section VI, the authors describe "This bubble interface expands outward from a relatively small radius (<10 μm) and high energy density, compressing the post-shock region to a higher density." About this point, the shock front is a smooth spherical shape while the interface between the cavitation bubbles and liquid have more complicated structure. Could the non-spherical bubble-liquid interface shape affect the stress distribution/gradient in the liquid? If so, could it disturb the shock wavefront structure slightly?

The authors appreciate Reviewer #1's thoughtful comment on whether irregularities in the bubble surface could influence the shock wavefront structure. A relevant piece of information to this comment is the speed of the bubble interface, about 0.6 km/s (subsonic), in comparison with the shock speed, about 1.4 km/s (supersonic). Perhaps for early timescales the proposed bubble-shock interaction may be significant, however for the timescales imaged in this work the authors judge the shock front to be unaffected by this higher-order bubble interface structure, based primarily on the speed difference. The main way we conclude that this bubble influences the shock is by displacing the volume of liquid in the post-shock region, the same effect detailed and quantified in this work.

5. Please add a description for the bottom image of Fig 6 in its caption.

The authors thank Reviewer #1 for pointing out this oversight in the caption for Figure 6; it has been edited to include a description of the bottom image.

Reviewer #2 (Remarks to the Author):

In this submission, the authors reported an experimental study of shock waves and their evolution in heptane. To perform these measurements, the authors utilized a rep-rated pulsed power driver at the Advanced Photon Source. While I agree that this work is certainly interesting, in its current state the article is not structured appropriately to clearly present its contents and does not set the work in context.

I believe that this article can be published within Communication Physics after some re-structuring. I will order my comments and questions according to the sections of the paper.

A. Motivation

It is unclear if the aim of the publication is to present a novel {high rate, cheap, portable} target, a more sensitive measurement, or a type of analysis that has not been presented before. The introduction is also missing a large amount of previous work that would set it into context.

The authors have restructured the manuscript's introductory/motivation section for better clarity of this work's goals. The updated introduction clearly presents this work's motivation as two-fold; firstly, the benefits of a this work's dynamic target (high rate, inexpensive, portable) are discussed. Secondly, the benefits of XPCI for the development and results of data-driven quantitative analysis are discussed. The authors trust that this restructuring is an appropriate response to Reviewer #2's concern. Several relevant new references have also been added to support this section.

1. I advise the authors to discuss previous rep-rated experiments at advanced light sources – these were largely experiments performed with lasers for example:
    a. https://doi.org/10.1038/s41467-021-23664-1
    b. https://www.xfel.eu/sites/sites_custom/site_xfel/content/e35165/e46561/e46886/e46963/e83032/xfel_file83043/11_Roadmap_DIPOLE_Wark_eng.pdf
    c. These are just experiments at the EuXFEL (which is pioneering multi-Hz ~100 J laser experiments) – there are certainly examples of such experiments (at a lower rep-rate) at LCLS MEC too.

The authors have added references to prior published laser-driven experiments in this section, while highlighting the relative novelty of the current work: "Typically, these phenomena are investigated at high event repetition rates via laser-based energy deposition [9, 28], however these laser systems tend to be cost-prohibitive and relatively immobile, therefore not conducive for a single research group to use at different light facilities. In contrast to laser-based HED experiments, this work details the development of a pulsed plasma device submerged in ambient liquid heptane which produces several of the physical phenomena of interest at event rates which can exceed 10 Hz, an event rate of high interest to HED physics. This benchtop-scale pulsed plasma target has the potential to broaden availability of HED-relevant experiments, …"

2. “…we have confirmed to be sensitive to the event’s associated shock with superior temporal and spatial resolution compared to prior results…” – It would be beneficial to reference previous work that this is being compared to, or at least quote relevant numbers.

The authors have modified this sentence, deemphasizing sensitivity comparison to prior work while emphasizing that this work’s image sensitivity was sufficient for the development of data-driven quantitative methods of analysis: “…the fact that such a weak shock front is still observable in this work highlights the excellent imaging sensitivity demonstrated in this work, facilitating methods of analysis which can draw more subtle quantitative data directly out of these images.”

3. “… highlights the superior sensitivity of this implementation of phase contrast imaging…” – For the reader, it is necessary to mention the key difference which distinguishes this implementation of PCI from the referenced work or other common implementations.

The authors understand that this sentence’s wording was misleading in the reviewed version of the manuscript; the main intent was to refer to the sensitivity of XPCI in general, not just this work’s implementation. This statement has been adjusted for clarity in the updated manuscript.

Other minor points:
• “…direct imaging photography remains one of the most impactful and data-rich methods for learning about any physical phenomenon…” – I should highlight that there are many physical phenomena where direct imaging photography will not be of much use. I recommend that this statement be modified to something a little more down-to-the-earth.

The authors appreciate this comment. This first sentence of the manuscript has been modified the be more appropriate and down-to-earth: “…direct imaging photography has remained a valuable and data-rich method for exploring and understanding a wide range of physical phenomenon…”.

• I recommend (but do not insist) that the authors broaden their listing of advanced light sources and user facilities beyond the United States. This might be more representative of the global scientific community – for example by including the European Synchrotron or European XFEL, which have also been active in dynamic compression science. This is particularly stark given that the ESRF has been the first synchrotron world-wide to upgrade its storage ring to achieve higher emittance. This is mentioned as an upcoming upgrade for APS (“…which will have superior brightness…”) later in this manuscript, but a similar upgrade has already been done at ESRF (https://doi.org/10.1038/s42005-023-01195-z).

The authors understand the importance of global representation in this instance; the manuscript has been updated to include international facilities, especially in light of recently-published improvements. Regarding the status of recent light source upgrades, the “…which will have superior brightness…” sentence mentioned by Reviewer #2 has been modified to reference the ESRF alongside the APS, as well as to change verb tense to the present tense.

B. Experimental Setup

1. Is it possible to provide a reference for the size of the discharge channel (i.e. ~ 5 micron at peak current)?

The ~5µm discharge channel diameter estimation was taken from Figure 5 of the authors' prior work, Campbell et al. 2021 (https://journals.aps.org/prresearch/abstract/10.1103/PhysRevResearch.3.L022021), which imaged pre-breakdown initiation streamers/channels for pulsed plasmas in water, of which ~5µm was the smallest observed diameter; while the liquid media varies from the current work, we consider this value to be an appropriate estimate of the initial channel diameter. A corresponding edit has been made in the updated manuscript.

2. Comparison between the peak energy per unit mass of the discharge channel and the National Ignition Facility appears to draw a quantitative connection between these two facilities. Is it possible to clarify why this number allows any connection to be made? Do the related physical effects that can be studied on your platform scale appropriately? This needs to be clarified because, as an absurd example, an average Snickers bar has energy density per unit mass of 20 MJ/kg. This is within three orders of magnitude of your experiment but does not necessarily draw a meaningful connection. I also believe that the value quoted for NIF, 1TW/kg, should be 1 TJ /kg.

The authors appreciate Reviewer #2's comment about the relevance of comparing this work's results to the NIF; the initial motivation for including this comparison was to indicate the current work's relevance to regimes relevant to high-energy-density physics. In order to retain this point in the manuscript without making a potentially unwarranted comparison to the NIF or any other facility, this comparison has been replaced in the updated manuscript with a cited source stating the range of HEDP-relevant energy densities (>100 GJ/m^3). The authors trust that this modified comparison is more appropriate.

3. Looking at Figure 1, it appears that your capacitor is 1nF in capacitance and is charged to 10 kV. This means that the capacitor holds E_tot = ½ C V^2 = 50 mJ of energy. In text, you state that the energy input into the discharge is ~ 100 mJ, in Figure 1 I see ~70 mJ. Both numbers appear to suggest > 100% coupling efficiency from the capacitor into the experiment. Is it possible to clarify this discrepancy?

The authors thank Reviewer #2 for pointing out this source of confusion; the discrepancy can indeed be clarified. Firstly, note that the voltage probe as shown in Figure 1 is not measuring the stored capacitor voltage, but rather the voltage across the imaging target gap (the probe's connection to electrical ground is implied in the figure); this was done in conjunction with the current probe to quantify energy dissipation within the imaging target. Secondly, the authors have edited the manuscript to mention the actual voltage value to which the capacitors was charged: 14.5kV → 105mJ of stored pre-event energy. The 10kV which Reviewer #2 read from the voltage trace in Figure 1 is in fact a transient value which occurs after the triggered spark gap event but before the imaging target event; this voltage does not reflect the total amount of energy available to the system. The 70mJ shown in Figure 1 is a measurement (via P=I*V integration) of only the energy dissipated within the imaging target. So, the overall approximate energy balance in this case would be:

(~100mJ stored on the 1nF capacitor) =
(~30mJ dissipated by the laser-triggered air spark gap switch) +
(~70mJ dissipated by the imaging target gap, measured via P=I*V integration)

The authors hope that this explanation and corresponding manuscript edit clear up Reviewer #2's concerns.

C. Energy Balance in Post-Shock Region

1. Is it possible to explain how the total electrical energy input was calculated from the current and the voltage measurements?

The authors thank Reviewer #2 for pointing out this gap in explanation; the beginning of this section has been modified to explain this more thoroughly: “As mentioned previously, we quantify the total amount of electrical energy being exerted during the event via simultaneous measurement of voltage across and current through the imaging target, such that cumulative electrical energy input over time is calculated via $U_\text{total}(t) = \int_0^t P(t') dt' = \int_0^t V(t') I(t') dt'$.”

2. Is it possible to provide more intuitive representation of U_shock? Do I understand correctly that this is a fraction of the total deposited energy that is converted into thermal energy by the shock wave produced by the discharge channel?

The language in this section which introduces and defines U_shock has been adjusted for clarity. Reviewer #2 interpretation is largely correct, though the authors prefer the wording included in the updated manuscript: “…we define U_shock as the cumulative excess internal energy relative to ambient at a given time within the post-shock region”.

3. Related to previous question, given the error bars on your voltage and current measurements. Do you believe that 2.8% is a significant number, or is this number consistent with no significant excess heating?

To address Reviewer #2’s concern about the significance of this number, the authors have modified Figure 5a to include the density vs. time fit line if alternate values of α are assumed, in this case an order of magnitude above and below the fitted value of α. This illustrates that while the method’s sensitivity when measuring α is relatively weak (we have also reduced number of reported significant figures from 2 to 1 for this reason: α = ~3%), this result is indeed significant enough to reject the no-heating null hypothesis ($\alpha = 0 \rightarrow \rho_2/\rho_1 = 1$) which would clearly reside outside of the displayed error bars for ρ_2/ρ_1 determined from XPCI data. The authors trust that this modified energy balance discussion is appropriate and satisfactory.

4. Is there any context why one should care about this number? This would hopefully be clarified within the introduction.

The authors have added additional discussion to the introduction which addresses this particular concern: “More generally, the methods of quantitative thermodynamic analysis and energy balance results presented herein aid in improving understanding and control of this pulsed plasma phenomena, for further utilization of pulsed plasmas in fields such as chemical processing of liquids [34], non-equilibrium liquids sterilization for biomedical applications [35], and other plasma-enhanced engineering applications [36].

D. Rankine-Hugoniot Shock Analysis

1. If understood correctly, your PCI analysis method assumes a cylindrical sample oriented perpendicular to the X-ray axis. How does this affect the results, especially when your shock waves are effectively spherical (radius at late times > interelectrode distance).

Reviewer #2 interprets the analysis method correctly; the computational model was derived by assuming a translationally symmetric one-dimensional geometry, indeed equivalent to a cylinder if the sample cross section is circular and the axis of symmetry is linear. Reviewer #2 is concerned about whether this assumption is sufficiently valid for the longer-timescale shock contours which are more spherical; since the XPCI model is being fit to the average of cutlines perpendicular to a spline fit of the shock contour image (see Figure 4a), and the width of the region being considered is small compared to the overall shock radial position at later timescales, the computational model therefore has negligible sensitivity to the longitudinal curvature of the shock contour. The only situation where this would not be true would be for very small spherical shocks, a case absent from this work's results. For this reason, we judge the "cylindrical" assumption to be sufficient for our purposes here; a more precise name for this assumption would be "one-dimensional" or perhaps "curvilinear" instead of "cylindrical", relevant word choice in the manuscript has been adjusted accordingly.

2. Is this "bubble effect" relevant in any other system of interest to science or engineering? For example, underwater explosions / earthquakes in marine context?

Yes, this "bubble effect" is relevant to multiple different fields of physics and engineering. The most closely related engineering application of the present work is the use of submerged pulsed plasmas for plasma-assisted drilling; a few of our contributing coauthors have a publication in that space, referenced in the updated manuscript: https://pubs.aip.org/aip/jap/article/129/18/183309/1078936. This manuscript's observed "bubble effect" has the potential to improve predictions and controllability for future iterations of this concept. Other examples for which this effect is relevant include tumor treatment using focused underwater shocks as well as in the defense of naval vessels, amongst others referenced in the updated version of this manuscript section.

Overall, I recommend that you focus on a single message within the publication. I assumed that this manuscript was meant to present a novel approach to HEDP science, but this message was not very clear. If re-structured to clearly communicate the utility of large data sets to your analysis, I am certain the content of this manuscript can form a nice publication.

Reviewer #3 (Remarks to the Author):

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In my opinion, after improving the clarity of some figures, the paper is well-suited for publication. Please see my specific comments below:

1. In Figure 1, the labels should be enlarged to enhance readability.

The authors thank Reviewer #3 for this suggestion; to address readability, the updated version of the manuscript spreads Figure 1 across two columns. The authors defer to the editorial board's judgement on whether further adjustment is needed on this matter.

2. In Figure 2, the time labels should also be enlarged and corrected to improve visibility.

The authors thank Reviewer #3 for his suggestion regarding the timestamps on Figure 2; these timestamps have been enlarged in the updated manuscript to enhance readability.