Dear Dr. Nguyen,

We would like to thank you and the referees for the thoughtful consideration of our work and for providing useful feedback that has improved our manuscript. Below we present a point-by-point response to all the comments and questions raised. Following the suggestions by the reviewers, we have revised our manuscript in order to further clarify our work and the successful operation of liquid cryogenic hydrogen jets. We are confident that we have addressed the comments of the reviewers and that our work meets the standards of JoVE. Thank you for your consideration of our revised manuscript.

Sincerely,
Christopher Schönwälder and Chandra Curry, on behalf of all the authors.

Editorial comments

Changes to be made by the Author(s):
2. Please revise the table of the essential supplies, reagents, and equipment. The table should include the name, company, and catalog number of all relevant materials in separate columns in an xls/xlsx file. Please sort the Materials Table alphabetically by the name of the material.

We have updated the table to follow the format and it is now in alphabetical order.

3. Please incorporate the Principles section into the Introduction as that is not a standard JoVE publication section.

We've incorporated the Principles section into the introduction but left the labeled sub-sections in the text for clarity and quick referencing.

4. Please specify all experimental parameters used. We need a specific experiment with specific values (temperatures/pressures) used in order to film.

All experimental parameters for an example case of 5 µm cylindrical hydrogen jet at 17 K, 60 psig have been listed throughout the introduction and protocol. For filming, we will show the protocol using this example case. Other typical values for extensions of this platform to rectangular apertures and other gases are listed in Table 1. We decided to present it in this way since the protocol is nearly unchanged for these other cases aside from the shift in the phase-diagram of the sample. To clarify this point in the manuscript, we have added the following sentences at the beginning of the protocol:

"The following protocol details the assembly and operation of a 5 µm diameter cylindrical cryogenic hydrogen jet operated at 17 K, 60 psig as an example case. Extension of this platform to other aperture types and gases requires operation at different pressures and temperatures. As a reference, working parameters for other jets are listed in Table 1."

*7. Please discuss some limitations of the protocol in the Discussion.
Please do not abbreviate journal titles.*

As is, the first two paragraphs of our discussion detail the most common failures as well as the limitations of implementing this protocol. Please let us know how this can be further clarified. Reviewer #1 has identified the specific case of high-intensity laser interactions with the jet and the possible damage to the aperture. As explained in our response to the reviewer below, we think that a more detailed discussion is beyond the scope of this manuscript which intends to describe the system operation. We think that a full study of the robustness of this platform in the harsh environment of high-intensity laser plasma interactions with 100 J-class lasers merits a dedicated publication. The authors have already demonstrated the use of this system as a high repetition rate multi-MeV proton source over a range of laser energies which is the basis of the claim made in the abstract.

Report of Reviewer #1 –

We would like to thank Reviewer #1 for the kind and thoughtful comments. Below we present a point-to-point reply to each of the comments.

I have seen a number of talks on this approach (and competing ones) as well as on some of the results achieved at various laser facilities. Two problems that have come up are stability of the jet and operation of the system when high energy laser pulses are used. Jet stability is addressed both in the text and in a nice data Fig., Fig. 6 and significant progress has been made. Successful operation of the jet when high energy pulses are used is not addressed as well. It is mentioned in the Discussion section, but only briefly, line 635: "Another observation is that the aperture can be damaged when the jet is irradiated by an ultra-high-intensity laser too close to the source. Recently, a mechanical chopper blade operating at 150 Hz and synchronized with the laser pulse, has been implemented to protect and isolate the aperture from the laser-plasma interaction." I think a modest amount of additional detail is needed here or it is not possible to evaluate this instrument's operation under a range of conditions of considerable interest. Let me stress that since, for many situations, this system is one of the best or only games in town, the existence of limitations is not a barrier to publication, but it is important to have some idea of what they are; for example, by providing approximate Fig.s of merit such as how far from the tip can pulses with 100 mJ, 1 J, 10 J, 100 J energies be safely used. It would also be interesting to know if use of the chopper degrades operation in some way (jet thickness, position stability, etc.).

While we agree with the reviewer that this platform has been most successfully used with free-electron lasers (LCLS, EuXFEL, FLASH), synchrotrons, and up to 100TW-class lasers, there are limitations to the survivability of the aperture when petawatt lasers are focused to ultra-high intensities. Already, the co-authors have conducted experiments at laser facilities using each of the stated energies. Increasing the shooting distance to more than 15 mm from the aperture at the expense of spatial jitter, allowed high repetition rate experiments to be conducted with the Draco PW (800 nm, 20 J, 30 fs) at Helmholtz-Zentrum Dresden-Rossendorf (HZDR). We expect the recently developed mechanical chopper will prevent catastrophic failure of the aperture that we have observed after a single shot when conducting experiments with high-energy (>100 J) PW-class lasers such as the Texas PW (1054 nm, 130 J, 145 fs). We will apply for experimental time through LaserNetUS to test this in CY2021. Further, recent experiments with the Draco PW at HZDR using the mechanical chopper allowed the shooting distance, thus the spatial jitter at the interaction point, to be decreased. The full technical details of the mechanical chopper system, measurements of the jet quality after being chopped, spatial jitter as a function of distance, and tests with the Draco PW will be covered by a future publication.

As the paper attempts to describe the operation of the jet, we choose to limit the discussion about high-intensity laser-plasma interactions which is only one of the many applications of this system.

Shooting distances used in our experiments are always listed in the publications from our collaboration. For example, Gauthier et al. *Applied Physics Letters* **111**, 114102 (2017) by the co-authors demonstrated high-repetition rate multi-MeV proton acceleration using this platform, a shooting distance of 15 mm was used without damage to the aperture. Each curve in the figure shown below (extracted from the aforementioned paper) correspond to 60 shots at 1 Hz at laser energies from highest to lowest of 3 J, 1.2 J, 600 mJ, and 180 mJ.

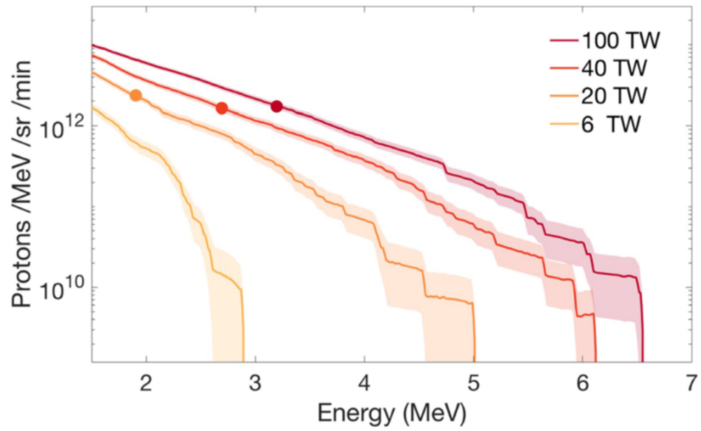

FIG. 3. Integrated proton flux for 6, 20, 40, 100 TW collected at 1 Hz over 1 min. The standard error of the mean is represented by the shaded area. The $0.8\,E_{CO}$ is indicated by filled circles at 1.9 MeV, 2.7 MeV, and 3.2 MeV.

Line 190 has a sign error given line 186. The Bernoulli left hand side (pressure + kinetic energy density + potential energy density) needn't be zero. Explain why it is set to zero here.

We understand that, if quickly read, our representation of the Bernoulli equation might seem confusing. What follows is a more detailed derivation of the final equation and the estimations we made along the way.

Furthermore, we changed our formulation of Bernoulli's principle as follows, hoping that it will cause less confusion:

$$\frac{1}{2}\rho_H v^2 + \rho g z + p = const.$$

Our estimate is that there are no pressure losses along the hydrogen line. So, we are looking at the conservation of flow across the aperture. Because of the size difference between the liquid channel leading up to the aperture and the micron-sized aperture, the flow before the aperture can be estimated as stationary relative to the jet that is injected into the vacuum chamber. Therefore, the term z expresses the difference in elevation between the aperture and the entrance to the vacuum chamber, meaning that z describes the thickness of the nozzle. Therefore, the term $\rho g z$ is very small compared to the other terms.

Let "VC" stand for vacuum chamber and "N" stand for "nozzle".

$$\frac{1}{2}\rho_H v_{VC}^2 + p_{VC} = \frac{1}{2}\rho_H v_N^2 + p_N$$

$$\frac{1}{2}\rho_H (v_{VC}^2 - v_N^2) + (p_{VC} - p_N) = 0$$

Due to the fact that the aperture is very small compared to the channel of the cryo-flange and the pressure inside the chamber is very low compared to the pressure inside the liquid channel leading up to the aperture, the following estimates can be made:

$$v_{VC}^2 \gg v_N^2 \ , \quad p_N \gg p_{VC}$$

What follows is:

$$v_{VC} \approx \sqrt{\frac{2p}{\rho}}$$

Fig. 6B isn't explained in the Fig. caption. Count percentage? Are the units of the horizontal axis correct?

Following this comment, we have added a more insightful caption describing the meaning of “count%”. We also corrected the misprint on the x-axis unit and changed from “mm” to “μm”.

Use scientific notation throughout. Don't represent numbers such as is done on line 165 and elsewhere: 4.2E-3.

We updated to scientific notation throughout our work.

I recommend providing a reference to the Ziegler-Nichols method for setting the PID.

Following the reviewer’s suggestion, we have added a reference for the Ziegler-Nichols method.

On line 167, a max pressure of 1E-3 mBar is referred to. Is this effectively the ambient pressure during operation or is this in some region near the jet? How large? Can a measure be provided of how much gas and over what region will a laser have to propagate through?

This value refers to the average pressure inside the vacuum chamber during operation that is measured at the pressure gauges located near the vacuum pumps. The average pressure of the vacuum chamber, while operating a 4x20 μm^2 hydrogen jet with the catcher, is typically ~10^{-4} mBar as shown in Fig. 7. When the liquid hydrogen solidifies by evaporative cooling, it remains continuous for more than 10 cm with no detectable localized gas density around the jet until it comes in contact with an ambient temperature object, such as the floor of the chamber or the inner surfaces of the catcher.

The cooling power of the cryostat will drop to zero before the vacuum pressure becomes problematic for laser propagation and self-focusing effects. The total gas in the chamber results from full vaporization of the injected flow minus the evacuated gas by the catcher system which depends on the size of the differential pumping aperture on the catcher as well as the effective pumping speed on the catcher vacuum line.

Report of Reviewer #2 –

Line 89 - "substantial gas load" - is there a particular value the author can give to the Pressure increase?

This depends on the total flow which is determined by the backing pressure and size of the aperture. Typical values extracted from Fig. 7, are shown below.

Before cooling down for a hydrogen flow of 23 sccm:

- Pressure in the vacuum chamber without catcher: 6.9x10^{-5} mBar
- Pressure in foreline: 5x10^{-2} mBar

During liquid jet operation for a hydrogen flow of 173 (290) sccm:

- Pressure in the vacuum chamber with catcher: 8.9x10^{-5} (1.2x10^{-4}) mBar

- Pressure in foreline with catcher: 6.3×10^{-2} (8.4×10^{-2}) mBar
- Pressure in the vacuum chamber without catcher: 8×10^{-4} (unknown) mBar
- Pressure in foreline without catcher: 1.6×10^{-1} (unknown) mBar
- Pressure in the catcher line: 7.1×10^{-2} ($1. \times 10^{-1}$) mBar

Line 91 - More specific on the type of pump and its relative pressure, this will have to be pumping at a higher vacuum than the local pressure.

The cryogenic jet system is highly adaptable and has been operated in a number of vacuum systems. The total pumping speed required will depend on the total flow of sample gas introduced, the vacuum system volume, and the efficiency of the jet catcher system. For this reason, we have decided to remain general by stating that the high vacuum in the vacuum chamber should be achieved by conventional means of a scroll-backed turbo pump while the catcher should be pumped with a roots pump as shown as shown in the P&ID diagram in Fig. 1. The reviewer is correct that the pressure does increase in the foreline when operating the jet. This flow-dependent foreline pressures are listed in the response to your previous comment and can be seen in Fig. 7.

Line 108 - Fig. 2 does not show indium on the diagram, this may be worth illustrating? Also the numbered features in the text don't quite match up to the order from left-to-right on the Fig..

Following the reviewer's comments, the indium was added to the diagram.

Line 110 - "Liquid reservoir" - specify this is the target (jet) liquid reservoir and not a Liquid-helium cooling reservoir for example.

We have updated terminology from "liquid reservoir" to "sample reservoir" on line 110 for clarity.

Line 111 - Reasoning behind the specific 76um of Indium. Why is this thicker than that of the source-cold finger interface (51um)

The difference in the thickness of the indium rings is primarily for the ease of assembly. A thinner layer of indium between two pieces of copper will be better for thermal conduction. However, a thicker layer of indium will provide more room for deformation and will make the aperture installation easier. We've edited the following sentence starting on line 112 to provide explanation for this:

"A thicker, 76 µm-thick indium ring is placed between the aperture and the liquid channel to increase the deformation length and reliably seal the aperture."

Line 153 to 167 - Consideration: Higher local pressure will increase the sublimation point temperature of the cryogen, and potentially allow operation of the jet at higher temperatures. Does gas conduction from higher gas pressures in the chamber outweigh this increase in operating window?

Unfortunately, we are not entirely certain we understood the question correctly. More specifically, whether or not "cryogen" refers to the cooling medium (liquid helium) or the sample gas (hydrogen).

In case "cryogen" refers to the helium used to cool down the cryostat, this will not be an issue. The liquid helium within the cryostat is isolated by a shroud flow and additional vacuum isolating shielding. An increase in gas pressure inside the chamber as we are experiencing it should not affect the flow of the cooling medium thus the jet.

In case "cryogen" is referring to hydrogen as the sample gas, the pressure used in the phase diagram (at which the hydrogen liquefaction occurs) is the sample gas backing pressure. Consequently, we do not expect the residual hydrogen gas inside the vacuum chamber to have any effect on the phase transition. Furthermore, as explained in detail in the answer to Reviewer #1, the pressure differential between the hydrogen line and the vacuum chamber forces the liquid hydrogen into the chamber. This operating pressure is significantly above atmosphere pressure (see Table 1), therefore we do not expect any increase of the vacuum chamber pressure to affect the flow ($p_N \gg p_{VC}$). As stated in the introduction of the paper, the only effect from a local hydrogen gas cloud is an increase in conductive heating which requires more cooling power from the cryostat to maintain a given temperature.

Line 160 - 17K in a vacuum of 10-3mbar is considerably above the boiling point for H2 - do you have a number on the boiloff/sublimation rate at this P/T?

The gray box in Fig. 4 describes the backing pressure and not the pressure inside the vacuum chamber. This value is usually in the range of tens of psig. We altered the caption of Fig. 4 and stated the range of parameters covered by the grey box to further illustrate this and avoid confusion.

For a 5 µm cylindrical jet operated at 17 K, 60 psig injected into vacuum, evaporative cooling on the surface of the liquid leads to rapid solidification after 100's of micron. The co-authors have performed an experiment at the Coherent X-ray Imaging (CXI) instrument at the Linac Coherent Light Source (LCLS) to visualize the ultra-fast crystallization of a 5 µm hydrogen jet which will be detailed in a future publication. We do not have a direct measurement of the sublimation rate under these conditions however over 1-50 mm (8-400 µs for jet velocity of 121 m/s) from the aperture we do not observe any change in the jet dimensions.

Line 232 - Why must the vacuum be better than 5x10-5 specifically? Explain the reason - to remove contaminants, for thermodynamic reasons, jet-dynamics reasons?

The jet is strongly affected by the vacuum conditions, which impact the cooling power. This is clearly described in the discussion of residual gas conduction beginning on line 155. While contaminants are not the primary concern linked to the vacuum pressure, the possibility of residual gases inside the vacuum chamber freezing out at the aperture has been observed especially when the local humidity is high which we pointed out in line 419. An average baseline pressure of $5x10^{-5}$ mBar has been empirically shown to lead to stable, reproducible jet operation in a number of experimental facilities. We have edited line 256 to read:

"Perform a vacuum test to determine the baseline vacuum pressure which, we have found, must be better than ~$5x10^{-5}$ mBar"

Line 239 - Is it worth mentioning the appropriate t-sensor cabling (4-lead, high AWG, heatsinking etc)?

Following the reviewer's comments, we specified the type of sensor used for the temperature sensor (4-lead silicon diode sensor) on line 101.

Line 247 - Maybe specify that the vacuum grease must be cryo compatible (Apiezon-N?)

We agree with the reviewer's comment and included a note, that the vacuum grease must be cryogenically compatible now on line 273.

Line 355 - Is this step to be done at ambient, or does each iteration require a warmup/cooldown cycle?

At the beginning of the protocol we've added a sentence which states which protocols are performed at ambient P/T and at what step the chamber is pumped down to high vacuum for jet operation (protocol 4.1) on line 244-245.

Line 387 - Suggest changing "sublimate" here to deposit. The gas will sublime and then deposit onto the cryogenic source.

We made the suggested change on line 420.

Line 451 - May be worth mentioning the pressure at this transition temperature (<20K).

The value of 20 K *for hydrogen* is now specified. In Fig. 4, the grey box illustrates the ranges of temperatures and backing pressures while operating a 5 μm diameter cylindrical hydrogen jet. Additionally, the right y-axis of Fig. 7 A. shows the changes we make to the sample backing pressure when liquefying and optimizing a cryogenic liquid jet.

Line 454 - Whereabouts in this process does the jet begin extruding from the aperture?

A mixture of gas and liquid will leave the aperture as soon as the phase transition for a given temperature and backing pressure is crossed. It will stabilize as soon as the liquefication process is completed. We have edited the sentence on line 489-490 to include this detail:

"At the onset of liquefaction, the gas flow will increase up to the maximum and a mixture of gas and liquid will spray from the aperture"

Line 551 - Confusion using the term "positioning". Is this actually positioning and correcting the cryostats position, or just determining the jets position in space?

This is highly dependent on the setup and application of the jet. If the user chooses to motorize the jet assembly, the jet would be moved. An alternative would be to motorize the imaging system and catcher.

We've updated Protocol 5.5 to avoid confusion on this point:

"5.5. Optional: If an application or experiment has a pre-determined location for the sample (e.g. detectors aligned to the same position in space), translate the cryogenic source using a multi-axis manipulator on the cryostat flange or motorized push-pin actuators in the vacuum chamber."

Fig. 6 - What is the time frame for this measurement plot? Is it an average of many jets over a long period of time, or a small sample from a particular experiment? Also mention the Reynold's number for the cryogen which produced this jet. Description of 6B is a little unclear. "...corresponds to motion perpendicular to the long axis" might be simpler to say "parallel to short axis"?

We have modified the discussion of Fig. 6 to provide more details regarding the stability in line 594-596. While these measurements were acquired during a single test and each point in 6 (a) is computed from 49 images saved at 10 Hz over 5 seconds, they are typical during stable 2x20 μm^2 jet operation. The co-authors have shown a more detailed study of the jet stability in the following plot extracted from Obst et al. *Scientific Reports* **7**, 10248 (2017).

The Reynolds number for the rectangular flow has been estimated using the hydraulic diameter[1], D_H, for non-circular flows and added to the Fig. 6 caption using the following calculation for a 2x20 µm^2 planar hydrogen jet at 18 K, 60 psig:

$$\rho(18K) = 73.765\ kg/m^3$$

$$\eta(18K) = 16.743\ \mu Pa \cdot s$$

$$D_H = \frac{4 \times A}{P} = \frac{4 \times 40}{44} = 3.63\ \mu m$$

$$R_e = \frac{\rho v d_0}{\eta} = \frac{73.765 \times 3.63}{16.743} \times \sqrt{\frac{2 \times 513659.4}{73.765}} = 1887$$

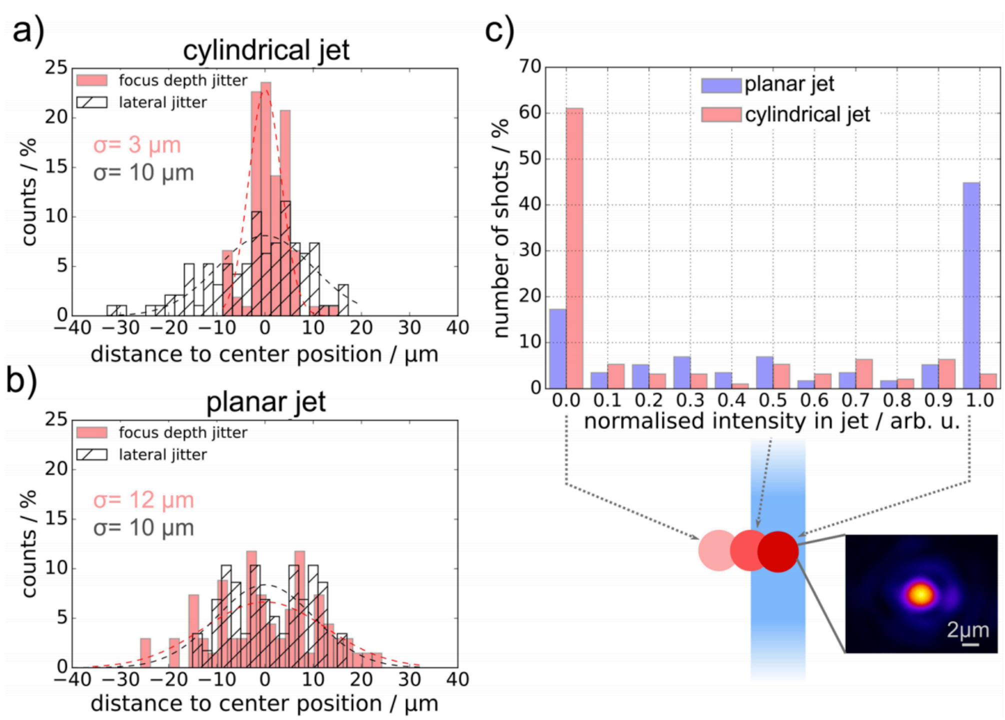

Figure 6. Target position stability study. (**a**) Cylindrical jet: focus depth jitter $\sigma = 3\,\mu$m, lateral jitter $\sigma = 10\,\mu$m. (**b**) Planar jet: focus depth jitter $\sigma = 12\,\mu$m, lateral jitter $\sigma = 10\,\mu$m, distance to nozzle: 10 mm. (**c**) Probability histogram describing the amount of laser intensity expected to be applied to the jet depending on its lateral positioning jitter. The intensity values are normalized to the maximum intensity, which is deposited in the cylindrical jet in the event of a central hit, while the value 0 corresponds to the case that the jet was entirely outside the laser focus. It was calculated from the overlap of a step-like jet profile and a Gaussian laser focus intensity distribution. The average spatial jitter of the laser focus was 1.5 µm and thus negligible compared to the lateral positioning jitter of the jet.

[1] H.E. Merritt, *Hydraulic Control Systems*. John Wiley & Sons. pg. 39 (1991).