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Journal

Journal of Applied Physics, 118(19)

ISSN

0021-8979

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Publication Date

2015-11-21

DOI

10.1063/1.4935295

Peer reviewed

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Citation: *Journal of Applied Physics* **118**, 195901 (2015); doi: 10.1063/1.4935295

View online: <http://dx.doi.org/10.1063/1.4935295>

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Analysis of laser shock experiments on precompressed samples using a quartz reference and application to warm dense hydrogen and helium

Stephanie Brygoo,^{1,a)} Marius Millot,^{2,3,b)} Paul Loubeyre,¹ Amy E. Lazicki,² Sebastien Hamel,² Tingting Qi,² Peter M. Celliers,² Federica Coppari,² Jon H. Eggert,² Dayne E. Fratanduono,² Damien G. Hicks,⁴ J. Ryan Rygg,² Raymond F. Smith,² Damian C. Swift,² Gilbert W. Collins,² and Raymond Jeanloz³

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(Received 24 June 2015; accepted 23 October 2015; published online 16 November 2015)

Megabar (1 Mbar = 100 GPa) laser shocks on precompressed samples allow reaching unprecedented high densities and moderately high $\sim 10^3$ – 10^4 K temperatures. We describe here a complete analysis framework for the velocimetry (VISAR) and pyrometry (SOP) data produced in these experiments. Since the precompression increases the initial density of both the sample of interest and the quartz reference for pressure-density, reflectivity, and temperature measurements, we describe analytical corrections based on available experimental data on warm dense silica and density-functional-theory based molecular dynamics computer simulations. Using our improved analysis framework, we report a re-analysis of previously published data on warm dense hydrogen and helium, compare the newly inferred pressure, density, and temperature data with most advanced equation of state models and provide updated reflectivity values. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4935295>]

I. INTRODUCTION

There is a great interest for measuring the properties of warm dense low-Z molecular systems, motivated by planetary implications and the fundamental understanding of the warm dense matter regime: pressures of a few hundreds of GPa (= a few Mbar) and temperatures $\sim 10^4$ K ~ 1 eV. Complex structural and chemical modifications from the molecular fluids to the warm dense plasma are expected in this domain, such as the existence of a first order transition between the molecular fluid and the plasma state in dense hydrogen (known as the plasma phase transition) or a superionic state in dense H₂O. Since 2003, a new approach combining static and dynamic compression techniques by launching strong shockwaves in precompressed samples^{1,2} has been developed and allows to explore those new extreme conditions of matter. As a few GPa precompression can induce significant density increase in compressible fluids, the locus of shock states (Hugoniot) accessible by the subsequent shock compression reaches lower temperatures and higher densities, as demonstrated on hydrogen and helium.^{3–5}

A typical configuration is sketched in Fig. 1: A sample is precompressed in a diamond anvil cell before being submitted to a strong shock compression generated by direct laser ablation of a thin plastic polymer layer deposited on one of the anvils.^{1,2} The shock wave propagation is monitored with ultrafast Doppler velocimetry⁶ (VISAR) and pyrometry^{7,8} (streaked optical pyrometer (SOP)) through the back anvil. We use a quartz plate precompressed with the sample as an *in-situ* reference for the impedance-matching

procedure that allows obtaining pressure-density equation of state data from the velocimetry measurements. We also use the reflectivity and emission from the shock front during its transit in the quartz as a reference for the reflectivity and temperature measurements.^{8,9} Important progress has been made recently in the characterization of quartz under shock compression^{8,10,11} and its release from shocked states. However, in the case of precompressed targets, the quartz is not following the principal Hugoniot and the higher initial density needs to be accounted for.

In the following, relying on a better understanding of shocked compressed SiO₂,^{8,10,11} we describe an improved analysis framework where corrections to the principal Hugoniot are presented and we show a re-analysis of previously published data on hydrogen^{1,5} and helium^{3,4} to document the changes in the inferred data according to this new framework. An Appendix contains details on the characterization of the initial state and the associated uncertainties.

II. ANALYSIS FRAMEWORK

A. Pressure-density equation of state measurements

1. VISAR velocimetry of strong reflecting shocks

We use a line-imaging streaked velocity interferometer system for any reflector (VISAR): an interferometric technique which records a phase shift proportional to the velocity of fast moving reflectors.^{12–14} When strong enough shock waves propagate in transparent media, such as oxides or low-Z compounds, they can produce a reflecting shock front. In this case, VISAR offers a line-imaging time-resolved record of the shock speed with better than 1% accuracy.⁶ We

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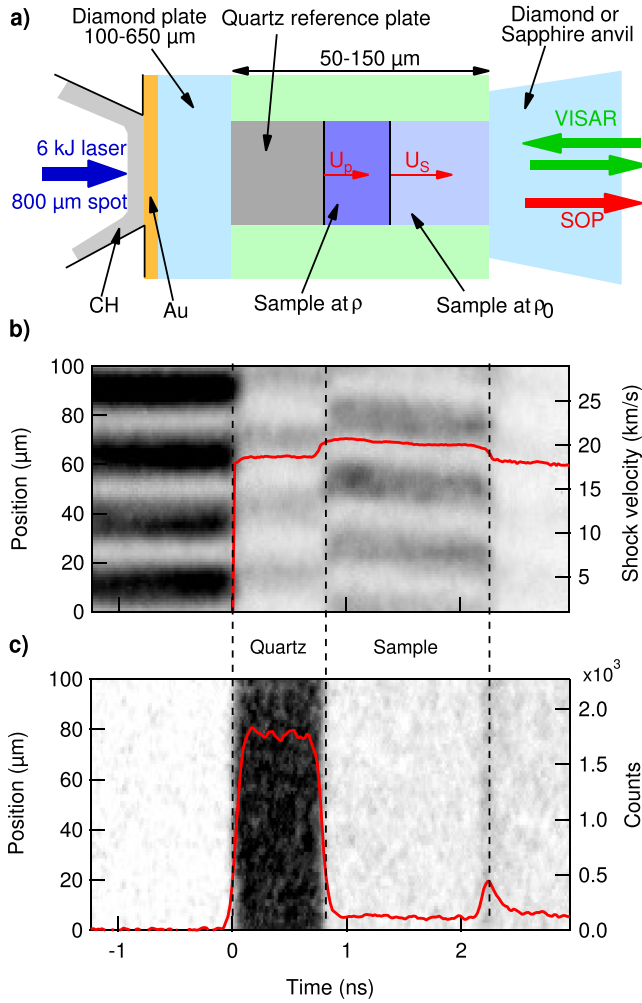


FIG. 1. Experimental setup and raw velocimetry and pyrometry data measured at the Omega laser (LLE, Rochester, NY). (a) Sketch of the diamond anvil cell (DAC). Direct drive laser ablation launches a shock in the precompressed target assembly and its propagation is monitored with velocimetry (VISAR) and pyrometry (SOP) through the back anvil. Typical pulses shapes for the laser are 1 ns with 1–6 kJ. Raw velocimetry (b) and pyrometry (c) images with superimposed shock velocity and counts (right scales) are presented. For clarity, the arrival of the shock in the quartz is chosen as the origin of the time scale. Depending on the anvil thickness, this event occurs ~ 5 –25 ns after the drive laser pulse.

can then obtain the shock velocities just before and after the shock crosses the interface between the quartz reference and the sample: U_S^Q and U_S^S .

Note that the true velocity of the shock front is in fact the ratio of the apparent velocity inferred from the fringe shift to the refractive index of the medium at rest.⁶ The knowledge of the refractive index of the precompressed sample and quartz reference plate is therefore required to obtain the shock velocity in the quartz $U_S^Q(t)$ and in the sample $U_S^S(t)$, and the uncertainty needs to be propagated. The formulas used for quartz are indicated in the Appendix.

2. Equation of state determination by shock impedance matching

When the shock wave encounters the quartz-sample interface, it is partly transmitted due to the shock impedance mismatch and a release wave (or a reshock depending on the

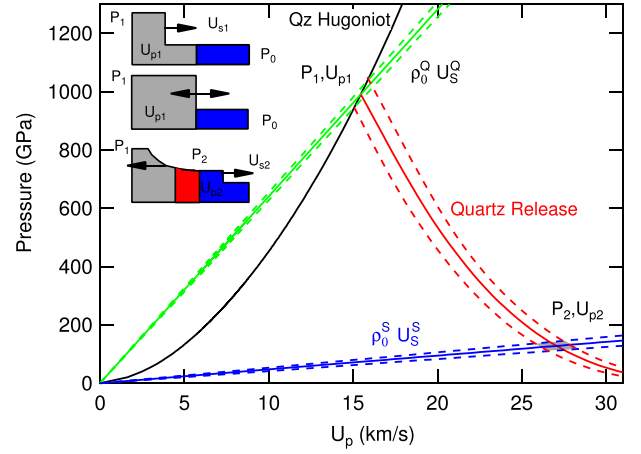


FIG. 2. Impedance matching construction: shock Hugoniot of the quartz standard (black), release isentrope (red), and Rayleigh lines having a $\rho_0 U_S$ slope in the (P, u_p) plane (green and blue). The uncertainty in determining the quartz velocity U_S^Q (green dashed lines) gives a set of different possible first shock states (P_1, u_{p1}) from which we calculate different possible release curves (red) which will intersect the possible Rayleigh lines for the sample (blue lines) yielding an area of possible final states for the sample (P_2, u_{p2}) . Inset: Sketch of the shock wave interaction with the quartz (grey)-sample (blue) interface.

impedance of each sample) is sent propagating backward into the quartz standard to ensure the continuity of the pressure and particle velocity at the interface. A graphical construction (Fig. 2) illustrates the derivation of the transmitted pressure and particle velocity. Knowing the Hugoniot of the quartz standard and using the Rankine-Hugoniot conservation relations

$$\rho = \rho_0 \frac{U_S}{U_S - u_p}, \quad (1)$$

$$P = P_0 + \rho_0 U_S u_p, \quad (2)$$

$$e = e_0 + \frac{1}{2}(P + P_0)(1/\rho_0 - 1/\rho), \quad (3)$$

the measurement of U_S^Q determines the incident shock state (P_1, u_{p1}) . The unknown transmitted shock state in the sample is therefore the intersection of the isentropic release path of the quartz standard from (P_1, u_{p1}) and the Rayleigh line $P_2 = P_0 + \rho_0^S U_S^S u_p$, where P_0 and U_S^S have been measured and ρ_0^S is inferred from P_0 using the sample's equation of state determined at ambient temperature.

3. Available experimental shock data and empirical relationships for precompression correction

The pressure-density relationship of shock compressed quartz in the high pressure fluid regime has been well characterized.^{15–23} We use a weighted, piecewise polynomial U_S - u_p fit of all existing data in the liquid domain:^{15–23} see Table I and Figure 3. The difference with the published fit,¹⁶ obtained using only Z-pinch data, is less than 1%.

Calculated Hugoniot obtained with an analytical equation of state model²⁴ (Sesame) and recent density functional theory based molecular dynamics (DFT-MD) simulations using the AM05 exchange-correlation functional are also

TABLE I. Summary of the equations and fits needed to use quartz as a standard for pressure, density, reflectivity, and temperature relative measurements. Velocities are in km/s and temperature in K.

Initial density: ρ_0	$\rho_0 = 2.649(P_0(4.9/37.7) + 1)^{1/4.9}$
Index of refraction: n_{Q0}	$n_{Q0} = 1.54687 + 0.1461(1)(\rho_0 - 2.649)$
Hugoniot: $U_S(2.65, u_p)$	$U_S(u_p < d) = a + bu_p - cu_p^2$
Piecewise polynomial fit	$U_S(u_p \geq d) = (a + c d^2) + (b - 2 c d) u_p$ $a = 2.124 \pm 0.121$; $b = 1.7198 \pm 0.0258$ $c = 0.01744 \pm 0.00133$; $d = 14.168 \pm 0.731$
Hugoniot shift with initial density	$U_S = U_S(2.65, u_p) + \alpha(\rho_0 - 2.65)$ $\alpha = 2.3(\pm 0.4) - 0.037(\pm 0.027)u_p$
Temperature quartz: $T(2.65, U_S)$	$T_{qz}(K) = 1860(\pm 190) + 3.56(\pm 0.52)U_S(\text{km/s})^{3.036(\pm 0.046)}$
Temperature shift	$T(\rho_0, U_S) = T(2.65, U_S) - \beta(\rho_0 - 2.65)$ $\beta \pm 30\% = -14786 + 1555 U_S$
Reflectivity quartz: $R(2.65, T)$	$R(2.65, T) = \frac{0.11(\pm 0.03)}{1 + (16968(\pm 737)/T)^{3.64(\pm 0.31)}} T^{0.095(\pm 0.029)}$
Reflectivity shift	$R(\rho_0) = R(2.65, T(\rho_0, U_S))$

presented.¹¹ As previously shown, the AM05 DFT-MD simulations capture quite well the pressure-density shock compressibility of warm dense SiO₂ along the quartz and fused silica Hugoniot.¹¹

To describe the Hugoniot of the precompressed reference, we use the experimental data available at standard density 2.65 g/cm³ and apply a small correction. It has been observed that for shock pressures above 150 GPa, the U_S - u_p relationship for various allotropic forms and various porosities of SiO₂ (initial density ranging from 1.15 g/cm³ to 4.31 g/cm³) can be

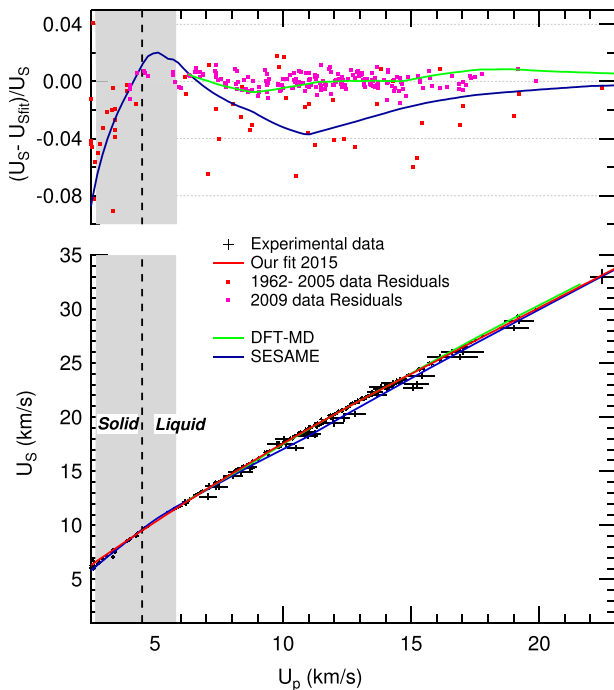


FIG. 3. Quartz principal Hugoniot: shock velocity U_S versus particle velocity u_p . Bottom: experimental data^{15–23} (black crosses), fit to the data (red), and calculated Hugoniot from Sesame (dark blue) and DFT-MD (green). Top: Fit residuals $(U_S - U_{S,fit})/U_S$ as a function of u_p . Red dots corresponds to data until 2005 (Refs. 15–23) and pink dots correspond to the most recent ones.¹⁶ Limit between solid and liquid shocked silica is represented by the vertical dashed line. Gray area: quartz reflectivity is below 2% making direct shock velocity measurement with VISAR challenging.

approximated by a set of parallel lines shifted by an offset that depends linearly on the initial density.^{19,23,25,26} In fact, recent high-precision measurements on fused silica¹¹ ($\rho_0 = 2.20$ g/cm³) and stishovite⁸ ($\rho_0 = 4.29$ g/cm³) shocked to the dense fluid state, as well as the Sesame and DFT-MD models (see

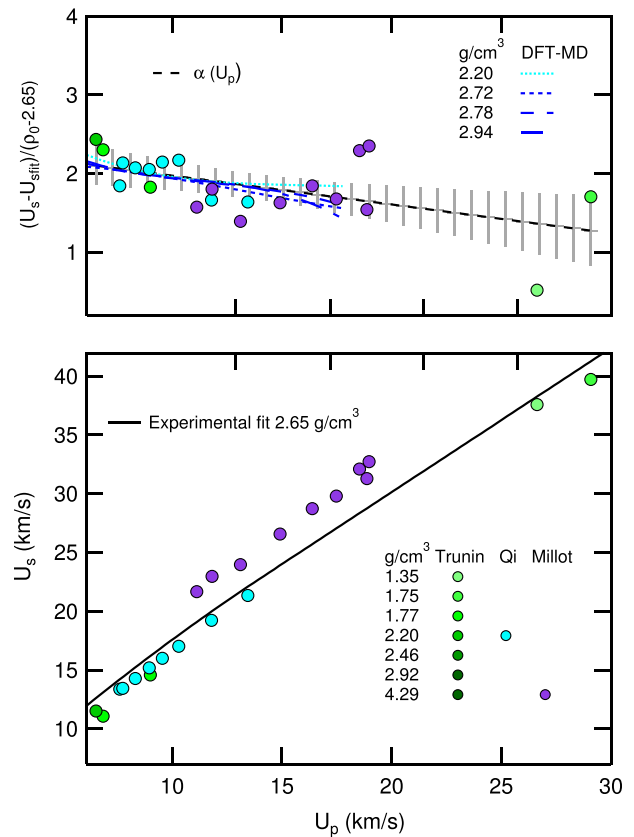


FIG. 4. Precompression correction on the quartz U_S - u_p Hugoniot. Bottom: shock data for different silica starting materials - porous silica^{19,23,25,26} (green), fused silica¹¹ (cyan), and stishovite⁸ (purple) - and fit to the quartz Hugoniot. Top: density scaled deviation from the quartz Hugoniot defined as $(U_S - U_{S,fit,2.65})/(\rho_0 - 2.65)$ for the data shown in the bottom panel and our empirical relationship (dashed black line with gray error bars). Initial densities of 2.72, 2.78, and 2.94 g/cm³ correspond to 1, 2, and 5 GPa pre-compressions, fused silica density is 2.20 g/cm³. Sesame model gives similar results to the DFT-MD (not shown).

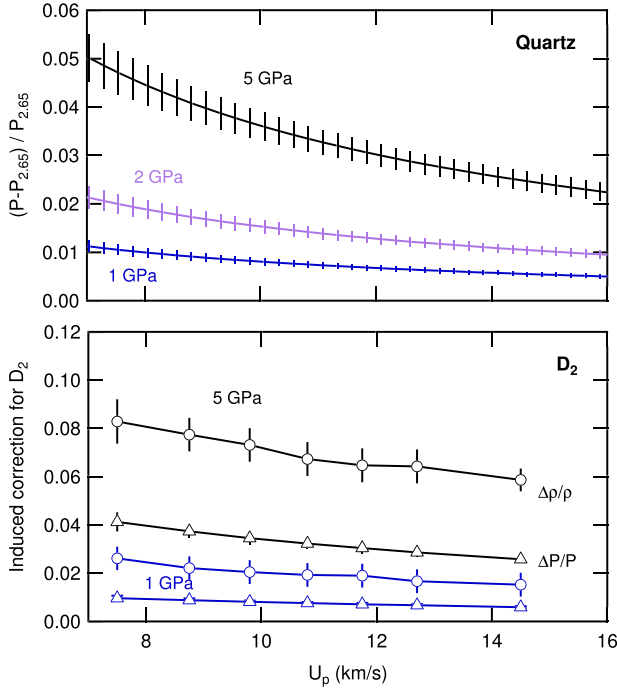


FIG. 5. Magnitude of the precompression correction on the quartz U_S - u_p Hugoniot. (Top) Effect on the inferred shock pressure in the quartz reference $(P(\rho_0) - P(2.65))/P(2.65)$. (Bottom) Effect on the inferred final pressure (triangles) and density (circles) for D_2 precompressed to 1 GPa (blue) and 5 GPa (black). The error bars reflect the systematic uncertainties arising from the uncertainty in determining $\alpha(u_p)$.

Fig. 4) suggest that a slightly more complex correction can describe these data more accurately.

Using the available experimental data for the different silica starting materials, we derive a correction to estimate $U_S(u_p, \rho_0)$ as a correction from the quartz Hugoniot $U_S(u_p, \rho_0 = 2.65)$ (all velocities in km/s and densities in g/cm^3)

$$U_S(u_p, \rho_0) = U_S(u_p, \rho_0 = 2.65) + \alpha(u_p)(\rho_0 - 2.65), \quad (4)$$

with

$$\alpha(u_p) = 2.3(\pm 0.4) - 0.037(\pm 0.027) u_p. \quad (5)$$

Only data in the liquid phase were used for the fit. The density change of the quartz reference (having a bulk modulus of 37 GPa) is however limited: it only amounts for 10% at a challenging precompression of 5 GPa.

We show the magnitude of the correction obtained with our approach in Fig. 5 (Top). For an initial density of 2.72 g/cm^3 ($P_0 = 1 \text{ GPa}$), the relative difference in shock pressure in the quartz $(P(\rho_0) - P(2.65))/P(2.65)$ is less than 2%, increasing to a few % with an initial density of 2.94 g/cm^3 corresponding to a 5 GPa precompression.

Note that due to the large impedance mismatch between the quartz and typical samples, larger changes are observed in the inferred quantities from the impedance matching procedure. In Fig. 5 (bottom), we present the magnitude of the change in pressure and density defined as

$$\frac{\rho_{\rho_{qz}=\rho_0}^{D2} - \rho_{\rho_{qz}=2.65}^{D2}}{\rho_{\rho_{qz}=\rho_0}^{D2}} \quad (6)$$

and

$$\frac{\rho_{\rho_{qz}=\rho_0}^{D2} - \rho_{\rho_{qz}=2.65}^{D2}}{\rho_{\rho_{qz}=\rho_0}^{D2}}, \quad (7)$$

for a deuterium sample at two different precompressions $P_0 = 1 \text{ GPa}$ and 5 GPa (we used Caillabet *et al.*³² equation of state for D_2). The correction of 4% in quartz velocity thus corresponds to a correction of 8% in shock density for deuterium precompressed to 5 GPa.

4. Quartz release model

Once the shock state in the quartz reference P_1 has been determined from the measurement of the shock velocity and the initial pressure, one has to determine the possible final states for the reference by computing the release curve from P_1 . In contrast with the great wealth of data on the principal Hugoniot, there are very few release measurements available for the quartz principal Hugoniot, and none for higher density polymorphs.

Previous studies of laser shocks on precompressed samples^{3,5} estimated the release by calculating the reflected Hugoniot and applying a correction depending on a constant Grüneisen²⁷⁻²⁹ $\Gamma \simeq 0.64$ or 0.66 .

An improved release model has been recently developed based on DFT-MD simulations and shock experiments.¹⁰ The release isentrope is parameterized as a Mie-Grüneisen correction from an effective reflected Hugoniot, using an effective Grüneisen parameter Γ_{eff} . The reflected Hugoniot is defined by a linear U_S - u_p relationship: $U_S = c_1 + s_1 u_p$ which makes the derivation of the release isentrope analytical. The slope s_1 is fixed to 1.197 and c_1 can therefore be uniquely determined from P_1 . The release isentrope can then be obtained using the parameterization of Γ_{eff} provided in

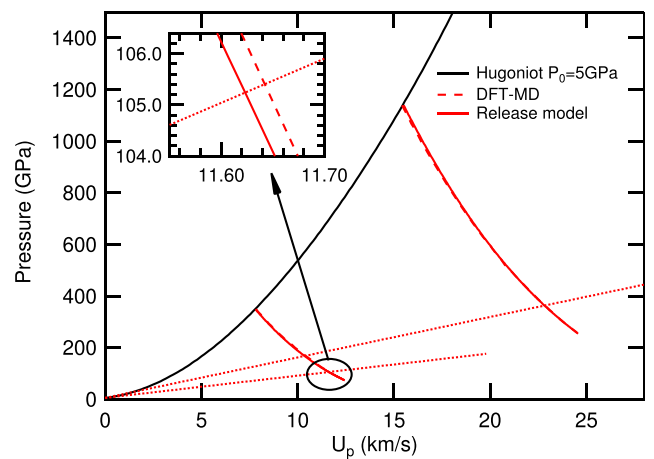


FIG. 6. Release model for precompressed Hugoniot. Example of the impedance matching construction in the P - u_p plane for a 5 GPa initial pressure: precompressed quartz Hugoniot (solid black), release isentropes from 348 GPa ($U_{S_{Qz}} = 15 \text{ km/s}$), and 1139 GPa ($U_{S_{Qz}} = 25 \text{ km/s}$) obtained from the DFT-MD (dashed red) and the release model from Ref. 10 (solid red) modified to take into account the initial pressure and Rayleigh lines for the two corresponding shocks in liquid deuterium (dotted red). Inferred u_p for the deuterium is determined at the intersection of the Rayleigh line and the release curves.

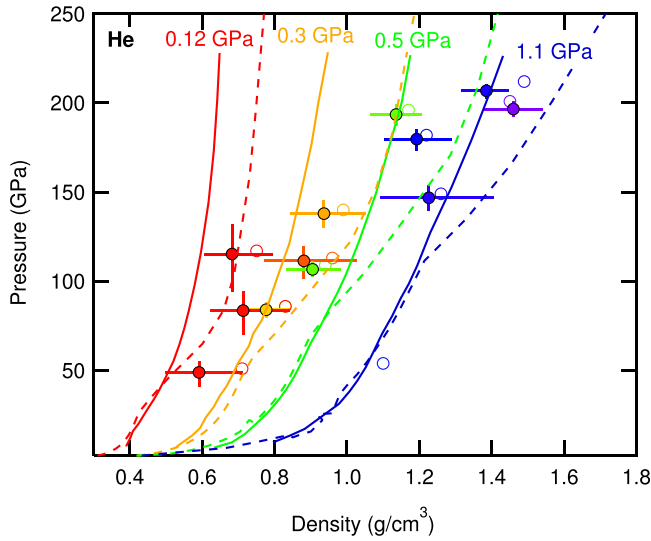


FIG. 7. Shock pressure versus density data for warm dense helium: open and solid symbols are, respectively, the published data³ and the re-analyzed one. The solid and dashed lines are DFT-MD calculations from Militzer *et al.*³⁰ and the chemical SCVH model.³¹ Red, orange, green, and blue indicate the initial pressure of 0.12 GPa (0.123 g/cm³), 0.30 GPa (0.225 g/cm³), 0.50 GPa (0.296 g/cm³), and 1.10 GPa (0.412 g/cm³), respectively.

Ref. 10. Note that in the precompression case, the derivation of c_1 needs to account for the initial precompression $P_0 > 0$:

$$c_1 = \frac{P_1 - P_0}{\rho_0^0 u_{p1}} - s_1 u_{p1}, \quad (8)$$

and P_1 and u_{p1} are determined using the precompressed Hugoniot.

We tested the validity of using this model (which had only been tested¹⁰ when $P_0 = 0$) to compute the release states of quartz shocked from a precompressed state by comparing the calculated release paths with isentropic releases computed from DFT-MD simulations. Note that the DFT-MD simulations were found to be in good agreement with the experiments in building the release model in Ref. 10. Fig. 6 presents the difference in inferred u_p along the isentrope for D_2 at a 5 GPa precompression for two shocks 15 km/s and 25 km/s in quartz. The difference is smaller than 1% independently of the initial pressure or the shock along most of the isentrope. The overall difference being smaller than the numerical noise introduced by interpolating the DFT-MD pressure and energy, we conclude that this model seems appropriate to describe release states from precompressed shock states. In the future, improved equation of state might confirm this assumption or provide a more accurate way of obtaining the quartz release paths.

5. Re-analysis of helium, deuterium, and hydrogen pressure-density data

We present in Figure 7 the shock equation of state data on warm dense helium³ first reported in 2008. Three contributions account for the difference between the values originally reported³ and the new ones. First, the function α was set to a constant value of 2.42 instead of a varying function with u_p . The precompression being below 1.25 GPa, the

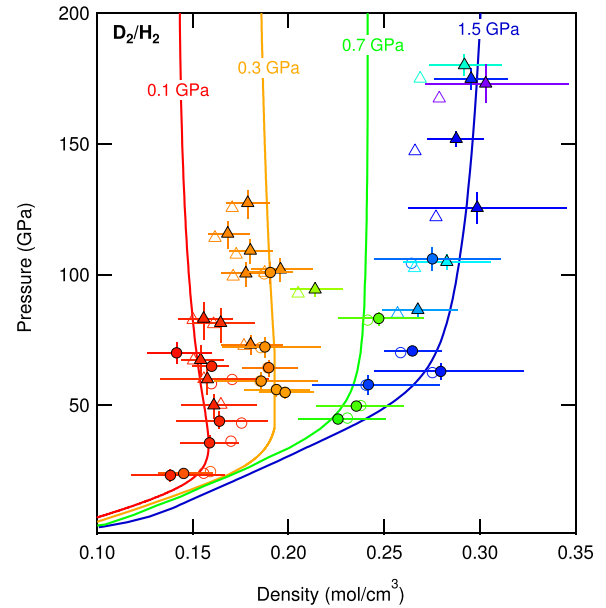


FIG. 8. Shock pressure versus molar density for warm dense hydrogen and deuterium: open and solid symbols are, respectively, the published data³ and the re-analyzed one. Triangles are D_2 data and circles H_2 data. The solid lines are DFT-MD calculations from Caillaud *et al.*³² Red, orange, green, and blue indicate the initial pressure of 0.13 GPa (0.029 mol/cm³), 0.30 GPa (0.044 mol/cm³), 0.70 GPa (0.061 mol/cm³), and 1.50 GPa (0.079 mol/cm³), respectively.

difference is below 0.35% which is negligible. As a comparison, for a precompression of 5 GPa, the difference would be around 1%. Using the new fit for the Hugoniot has a larger effect on the final density which, on average, *decreases* by 10% and by 4% in pressure. In contrast, the new release model has the opposite effect contributing to an *increase* in density of around 6%. So overall, only a 4% *decrease* in density is observed. With the improved analysis, at low precompressions, the experimental data are in better agreement with both DFT-MD calculations³⁰ and the chemical model SCVH (Saumon Chabrier Van Horn),³¹ but at higher precompressions, DFT-MD calculations reproduce better than the experimental data.

We present in Figure 8 the shock equation of state data on warm dense hydrogen and deuterium⁵ first reported in 2012. The difference between the previously reported values and the new ones comes mainly from the new release model. Depending on the shock impedance of the sample, the final density is either increased or decreased by a few % relative to the initial report. The agreement between the data and the latest DFT-MD calculations on hydrogen isotopes³² is improved. This seems to lift the small systematic discrepancy between the simulations and the data.

6. Evolution of the random and systematic uncertainties

Random uncertainties coming from technical limitations in the experimental accuracy of the observables—shock velocities, initial pressure—are sometimes important but are generally getting smaller as improved diagnostics are used and better experimental procedures are developed.

Systematic uncertainties arise from the models and empirical relationships used to determine the initial density and

TABLE II. He Hugoniot data: pressure (P), compression ratio (ρ/ρ_0), temperature, reflectivity, and variation of internal energy $E - E_0$ using the present model for the quartz reference and the shock velocity data from Eggert *et al.*³ and Celliers *et al.*⁴ For all quantities, the total errors (which include the systematic errors due to the quartz standard and random errors due to experiment) are given in parentheses and correspond to the error on the last digits. For example, $\rho/\rho_0(-, +) = 4.97 (58, 76)$ means $\rho/\rho_{0min} = 4.97 - 0.58$ and $\rho/\rho_{0max} = 4.97 + 0.76$.

Shot	P (GPa) (-, +)	$\rho/\rho_0(-, +)$	T (kK) (-, +)	R (-, +)	$E - E_0$ (kJ/g) (-, +)
33488	99 (3,3)	1.89 (6,6)	8.8 (0.4,0.3)	0.0 (1,1)	57 (3,3)
34833	111 (10,8)	4.96 (62,82)	45.7 (3.2,2.7)	0.6 (2,2)	251 (11,10)
34836	107 (4,4)	3.06 (24,27)	25.9 (1.6,1.4)	0.2 (2,2)	122 (5,5)
36177	84 (5,4)	3.41 (26,29)	28.3 (1.7,1.4)	0.0 (1,1)	131 (5,5)
36178	138 (8,7)	4.34 (43,53)	39.5 (2.4,1.8)	0.11 (6,6)	247 (10,9)
38993	115 (21,17)	5.59 (64,92)	60.5 (3.3,2.7)	0.17 (4,4)	388 (17,16)
38994	84 (13,11)	5.48 (70,99)	46.4 (3.3,2.8)	0.09 (5,5)	263 (11,11)
40131	197 (5,4)	3.40 (20,19)	30.3 (3.0,2.9)	0.05 (2,2)	163 (6,6)
41452	147 (7,7)	2.94 (32,43)	19.4 (0.9,0.7)	0.02 (2,2)	117 (5,5)
41453	194 (6,6)	3.88 (24,24)	44.2 (1.8,1.3)	0.12 (3,3)	246 (9,9)
41454	180 (6,6)	2.90 (22,23)	23.8 (1.2,0.8)	0.05 (2,2)	144 (6,6)
41455	49 (8,6)	4.52 (72,91)	30.4 (2.6,2.6)	0.01 (1,1)	146 (7,7)
43301	207 (4,4)	3.34 (17,15)	40.5 (5.3,5.9)	0.10 (5,5)	176 (6,6)

refractive index of both the quartz and the sample, as well as describing the behavior of the quartz under shock and release (including the precompression corrections).

The uncertainty in the initial density ρ_0 is a combination of the uncertainty in measuring the initial pressure P_0 by ruby luminescence ($\Delta P = 0.03$ GPa independently of the initial pressure and in the absence of pressure gradients within the precompressed sample) and the uncertainty in the static compression equation of states. Similar reasoning applies to the refractive index. Since the main contribution comes from

the constant uncertainty in the initial pressure, as the pre-compression is increased from 0.1 GPa to a few GPa, the importance of these uncertainty sources strongly decreases.

The reduction of the uncertainty on quartz shock and release behavior is the most important progress of the approach described in this work compared to the previous analysis of early data on hydrogen and helium. In addition, the improved correction for the higher initial density contributes to more accurate data. For example, for hydrogen, on average, the error in pressure drops from 7% to 5% and in

TABLE III. H₂ Hugoniot data: pressure (P), compression ratio (ρ/ρ_0), temperature, reflectivity, and variation of internal energy $E - E_0$ using the present model for the quartz reference and the shock velocity data given in Table I of Loubeyre *et al.*⁵ For all quantities, the total errors (which include the systematic errors due to the quartz standard and random errors due to experiment) are given in parentheses and correspond to the error on the last digits. For example, $\rho/\rho_0(-, +) = 4.97 (58, 76)$ means $\rho/\rho_{0min} = 4.97 - 0.58$ and $\rho/\rho_{0max} = 4.97 + 0.76$. For the shots in the lower part of the table, the shock velocity in H₂ could only be obtained with transit time measurement hence not accurately enough for determining the compression ratio or the energy variation.

Shot	P (GPa) (-, +)	$\rho/\rho_0(-, +)$	T (kK) (-, +)	R (-, +)	$E - E_0$ (kJ/g) (-, +)
34834	56 (3,2)	4.40 (37,40)	10.8 (2.0,1.9)	0.34 (7,9)	247 (11,10)
34835	24 (2,2)	3.85 (36,39)	6.1 (0.2,0.2)	0.11 (3,3)	118 (6,6)
36174	50 (2,2)	3.87 (35,41)	6.6 (0.8,0.7)	0.24 (6,8)	154 (8,8)
36176	45 (2,2)	3.64 (34,40)	5.9 (0.7,0.6)	0.15 (4,4)	133 (9,9)
38326	70 (4,4)	4.91 (54,64)	24.0 (3.1,3.0)	0.34 (5,5)	486 (54,63)
38991	83 (3,3)	4.11 (35,39)	12.4 (1.2,1.1)	0.32 (4,4)	264 (14,14)
38997	44 (3,4)	5.09 (70,79)	12.0 (1.3,1.4)	0.22 (5,6)	275 (26,36)
39000	63 (3,3)	3.55 (44,55)	5.6 (1.2,1.2)	0.21 (4,4)	147 (14,15)
41451	58 (4,4)	3.10 (37,48)	5.5 (1.0,1.1)	0.12 (3,4)	129 (15,15)
41458	23 (3,2)	4.30 (64,88)	5.3 (0.10,0.9)	0.03 (1,1)	139 (8,8)
43297	101 (4,4)	4.38 (26,27)	26.0 (4.0,5.2)	0.38 (9,10)	449 (15,14)
43298	106 (5,4)	3.58 (39,47)	10.4 (1.10,2.6)	0.40 (21,19)	252 (20,20)
47716	36 (2,3)	4.93 (48,47)	10.8 (1.0,1.1)	0.18 (4,5)	222 (11,15)
47719	64 (3,3)	4.57 (33,37)	15.8 (1.3,1.3)	0.36 (6,6)	305 (12,11)
52250	55 (2,2)	4.44 (30,33)	15.6 (0.9,0.8)	0.36 (4,4)	240 (8,8)
53835	59 (3,3)	4.27 (57,68)	13.4 (1.1,1.1)	0.30 (5,5)	263 (19,19)
53838	65 (2,2)	4.97 (31,29)	22.1 (1.6,1.4)	0.28 (4,4)	404 (29,33)
55003	72 (4,4)	4.32 (52,67)	18.4 (1.5,1.6)	0.35 (4,5)	321 (22,21)
56366	71 (2,2)	3.37 (19,19)	6.3 (0.3,0.3)	0.23 (5,6)	162 (7,6)
50377	16.8 (1.8,1.8)	0.38 (6,7)	...
53471	8.8 (0.7,0.6)	0.14 (2,1)	...
53472	7.8 (0.4,0.4)	0.13 (2,2)	...
53478	13.3 (1.2,1.1)	0.28 (5,4)	...

TABLE IV. D₂ Hugoniot data: pressure (P), compression ratio (ρ/ρ_0), temperature, reflectivity, and variation of internal energy $E - E_0$ using the present model for the quartz reference and the shock velocity data given in Table I of Loubeyre *et al.*⁵ For all quantities, the total errors (which include the systematic errors due to the quartz standard and random errors due to experiment) are given in parentheses and correspond to the error on the last digits. For example, $\rho/\rho_0(-, +) = 4.97(58,76)$ means $\rho/\rho_{0min} = 4.97 - 0.58$ and $\rho/\rho_{0max} = 4.97 + 0.76$. For the shots in the lower part of the table, the shock velocity in D₂ could only be obtained with transit time measurement hence not accurately enough for determining the compression ratio or the energy variation.

Shot	P (GPa) (-, +)	$\rho/\rho_0(-, +)$	T (kK) (-, +)	R (-, +)	$E - E_0$ (kJ/g) (-, +)
40133	116 (6,5)	3.95 (25,27)	26.4 (3.2,3.0)	0.29 (3,3)	254 (10,10)
41449	102 (5,4)	4.45 (35,39)	21.9 (2.5,3.8)	0.41 (10,13)	225 (9,9)
41459	73 (4,4)	4.29 (36,40)	18.3 (2.4,2.2)	0.44 (2,1)	168 (8,8)
47715	105 (3,3)	3.79 (30,31)	...	0.36 (10,12)	131 (7,6)
47718	180 (4,4)	4.03 (25,27)	18.8 (2.3,3.3)	0.36 (10,13)	236 (9,9)
47720	100 (5,5)	4.17 (30,35)	22.8 (2.3,2.5)	0.39 (8,8)	225 (10,9)
47721	50 (5,4)	4.95 (52,69)	15.2 (1.5,1.6)	0.26 (6,7)	154 (8,8)
50369	94 (3,2)	3.72 (22,25)	14.7 (1.1,1.2)	0.41 (5,6)	151 (6,6)
50370	86 (3,2)	3.49 (24,27)	9.5 (1.0,0.9)	0.32 (4,4)	102 (5,5)
50372	82 (8,6)	4.90 (44,53)	22.7 (1.8,1.8)	0.36 (4,3)	242 (10,10)
50378	125 (6,6)	3.71 (45,58)	13.7 (1.4,1.9)	0.46 (8,10)	144 (12,12)
52253	152 (3,3)	3.57 (19,18)	19.2 (1.3,1.2)	0.46 (4,3)	172 (6,5)
53473	109 (5,4)	4.10 (25,27)	26.6 (1.5,1.2)	0.39 (2,2)	235 (8,7)
53474	173 (7,7)	3.61 (38,51)	18.1 (1.8,1.8)	0.51 (5,5)	189 (14,14)
53839	127 (6,5)	4.12 (26,27)	33.2 (5,8,5)	0.40 (13,17)	279 (10,9)
56360	175 (4,4)	3.71 (24,24)	19.8 (1.6,1.7)	0.42 (4,4)	203 (9,8)
56370	83 (8,6)	4.79 (41,47)	25.1 (1.6,1.6)	0.26 (3,3)	253 (9,9)
55005	60 (6,5)	4.70 (73,96)	17.0 (1.9,1.9)	0.29 (3,3)	177 (16,16)
58084	67 (6,6)	4.74 (32,37)	22.5 (2.3,2.3)	0.37 (4,3)	205 (7,6)
47723	18.4 (2.1,2.7)	0.45 (8,10)	...
50371	6.9 (0.6,0.5)	0.0 (0,0)	...
50374	4.9 (0.4,0.3)	0.0 (0,0)	...
50376	6.2 (0.9,1.0)	0.13 (6,7)	...
50381	14.4 (1.4,2.2)	0.39 (9,13)	...

compression, from 12% to 10%. All re-analyzed data are presented in Tables II–IV.

B. Temperature of shocked compressed SiO₂

1. Diagnostics and data analysis

When monitoring reflecting shock fronts, the SOP images the thermal emission of the propagating shock front over a small spectral range,^{7,8} first in the quartz and then in the sample (Fig. 1). To determine the temperature from the measured thermal emission, we assume a grey-body approximation for the spectral radiance, $I = A\epsilon(\lambda) [e^{hc/\lambda kT} - 1]^{-1}$, where $\epsilon(\lambda) = \epsilon$ is the emissivity given by $(1 - R)$, R the measured optical reflectivity, A is a system calibration constant that incorporates the transfer function of the optical system and the response of the detector.

Inverting this expression to solve for temperature gives, $T = T_0 [\ln(\epsilon(\lambda) A/I + 1)]^{-1}$, where $T_0 = hc/\lambda_0 kT$ is a calibration parameter related to the wavelength of the spectrometer peak sensitivity ($T_0 \sim 1.9$ eV at $\lambda_0 = 650$ nm). Since the temperature determination is made relative to the quartz reference, the temperature in the sample can be determined from the ratio of the signal levels observed in the quartz and the sample, such that the system calibration constant A drops out of the expression, $T_s = T_0 [\ln(e^{T_0/T_q - 1} (I_Q^*/I_s^* + 1))]^{-1}$ with $I^* = I_{ADU}/(1 - R)$, I_{ADU} being the analog-to-digital counts associated with the observed signal, and R being the reflectivity measured with the VISAR.

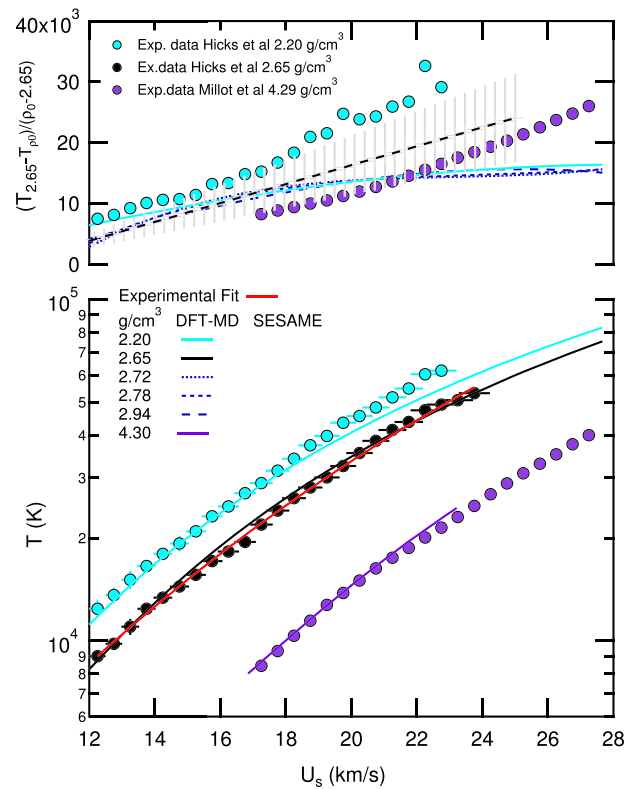


FIG. 9. Shock temperature as a function of the shock velocity for warm dense SiO₂. Bottom: Experimental data for quartz,^{8,29} fused silica,²⁹ and stishovite⁸ and DFT-MD (this work). Top: Precompression correction and density scaled data relative to the quartz Hugoniot (dashed black with grey error bars).

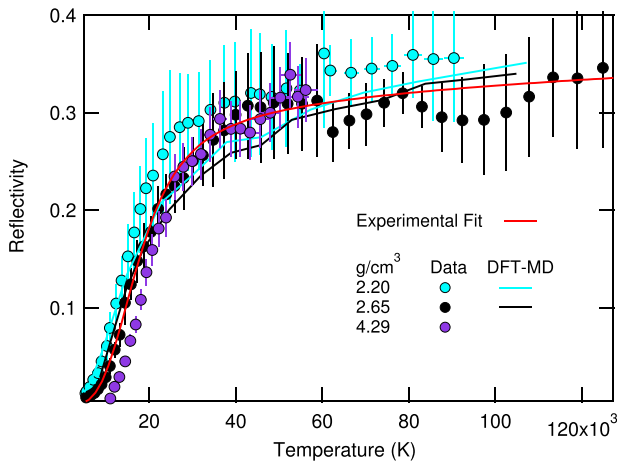


FIG. 10. Shock reflectivity as a function of temperature: black, light gray, and dark gray correspond, respectively, to experimental data on quartz,²⁹ fused silica,²⁹ and stishovite.⁸ The solid dark blue and light blue are reflectivity obtained from the DFT-MD simulations for quartz and fused silica.

Knowing the shock velocity in quartz, the temperature in quartz, T_Q , has to be determined from the calibrated function $T_Q(U_S)$. Then, one can obtain the temperature T_S in the sample.

2. Available experimental shock data and empirical relationships for precompression correction

The temperature along the principal Hugoniot $T_Q(U_S)$ has been measured by Hicks *et al.*²⁹ (black circles in Figure 9) and is well represented between 12 and 23 km/s by a power law

$$T(2.65, U_S)(K) = 1860 + 3.56U_S^{3.036}. \quad (9)$$

Due to its higher initial density,² the shock temperature of precompressed quartz will be lower at a given shock pressure. We propose a simple parameterization of the shock temperature for precompressed quartz based on experimental data on fused silica (2.20 g/cm³) and stishovite (4.29 g/cm³) (light blue circles and light purple circles in Figure 9). The DFT-MD is found to capture well the experimental data for the three starting densities. So does the Sesame EOS (Equation Of State) (Ref. 24) at high pressure once silica is considered fluid and dissociated. As it was done for the principal Hugoniot in the pressure-density plane, several Hugoniots at different densities are calculated and compared to estimate the shift in temperature expected with a higher initial density. The Sesame model and the DFT-MD simulations give very similar results and trends. We observe that a simple density scaling allows us to describe the difference in shock temperature between the quartz Hugoniot and either the fused silica or the stishovite Hugoniot (black curve in Figure 9)

$$T(\rho_0, U_S) = T(2.65, U_S) - (-14786 + 1555U_S)(\rho_0 - 2.65). \quad (10)$$

Given the reduced and sparse set of data, a relative uncertainty on this correction $\sim \pm 30\%$ seems reasonable: this

allows the model to describe relatively well the experimental data for lower and much higher initial densities as well as the DFT-MD results.

C. Reflectivity of shocked compressed SiO₂

1. Diagnostics and data analysis

In addition to extracting the shock velocity from VISAR fringe pattern shifts, we can also measure the reflectivity of the moving reflecting interface from the intensity of the fringes. In the case of a reflecting shock, the reflectivity (at the wavelength of the probe laser) is due to a mismatch of complex refractive index between the shock compressed material (index n) and the precompressed material (index n_0):

$$R = \frac{|n - n_0|^2}{|n + n_0|^2}. \quad (11)$$

A relative measurement of the reflectivity of the shock front in the sample compared to the shock front reflectivity in the quartz reference can be obtained easily from the ratio of intensities of the VISAR fringes in quartz and in the sample. When the shock front is in the quartz, the measured intensity of the VISAR fringes I_Q is given by $I_Q = I_P R_Q(U_S) f_T$, where I_P is the incident probe laser intensity at the shock front, $R_Q(U_S)$ is the calibrated reflectivity of the shock front in quartz, and f_T is the unknown transfer function of the optical system. When the shock is in the sample, the intensity of the VISAR fringes is given by $I_S = I_P R_S f_T$, where R_S is the reflectivity of the sample shock front. Combining these two equations gives $R_S = R_Q(U_S) I_S / I_Q$. A reflection at the quartz/sample interface can come from an index mismatch of the precompressed states and can be easily modeled knowing the influence of the precompression on the respective refractive index. It is usually less than 2% and it has been in this case neglected.

This relative measurement allows for accurate characterization of the optical reflectivity of the shocked sample even in the presence of strong variations of the transparency of the back diamond anvil/window that can be caused by the interaction of the high-energy drive-laser with the diamond anvil cell target and the ablation plasma. The measured reflectivity can then be used to estimate the temperature using the grey body approximation.

2. Available experimental shock data and empirical relationships for precompression correction

We show in Figure 10 the experimental data for shock reflectivity along three different Hugoniot starting with fused silica,²⁹ quartz,²⁹ and stishovite.⁸ A strong dependence in temperature is unveiled: the curves are almost indistinguishable. In particular, the small difference between the quartz (2.65 g/cm³) and the stishovite (4.29 g/cm³) suggests that changes in the temperature dependence of the reflectivity onset induced by the slight density increase for precompressed quartz are most likely negligible given the experimental uncertainties. This is in good agreement with recent DFT-MD results, suggesting that the shock reflectivity for

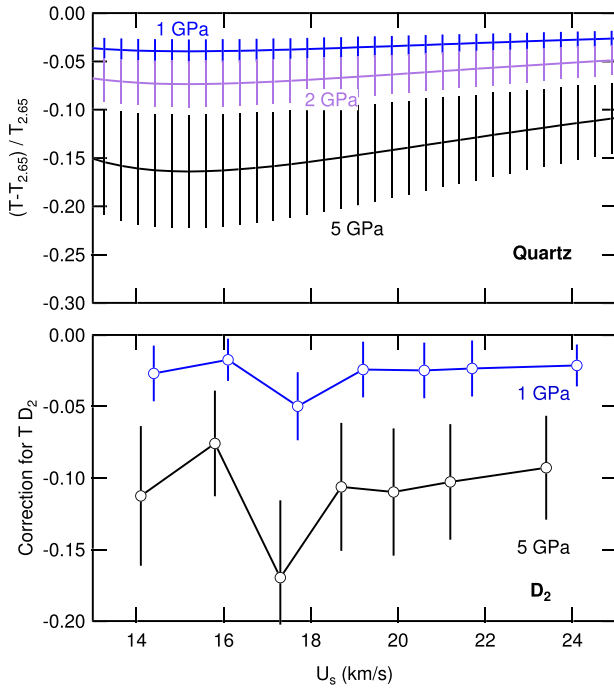


FIG. 11. Magnitude of the correction for the temperature of quartz $(T - T_{2.65})/T_{2.65}$ (upper figure) and for D_2 (lower figure) at $P_0=1$ and 5 GPa. The uncertainties are the systematic uncertainties coming only from the uncertainties in $\beta(U_S)$.

precompressed quartz up to 2.94 g/cm^3 ($P_0 = 5 \text{ GPa}$) depends only on the shock temperature.¹¹

Using the experimentally determined $R(U_S)$ and $T(U_S)$ for quartz, we combine them in a reference curve

$$R_{fit}(2.65, T) = \frac{0.11}{1 + (16968/T)^{3.64}} T^{0.095}. \quad (12)$$

Then, using the previously described parameterization to compute the shock temperature of the precompressed quartz $T(\rho_0, U_S)$, we obtain the reflectivity of the shocked precompressed quartz

$$R(\rho_0) = R_{fit}(2.65, T(\rho_0)). \quad (13)$$

The impact on the corrections in temperature and reflectivity for D_2 defined as

$$\frac{T_{\rho_{qz}=2.65}^{D2} - T_{\rho_{qz}=\rho_0}^{D2}}{T_{\rho_{qz}=2.65}^{D2}} \quad (14)$$

and

$$\frac{R_{\rho_{qz}=\rho_0}^{D2} - R_{\rho_{qz}=2.65}^{D2}}{R_{\rho_{qz}=2.65}^{D2}} \quad (15)$$

obtained using this approach is presented in Figures 11 and 12. An initial density of 2.94 g/cm^3 (5 GPa) gives a temperature correction for the quartz $\sim 15\%$ – 20% which gives for the reflectivity a correction that can be as high as 60% at low shock velocities.

Consequently, the reflectivity corrections for D_2 (lower Figure 12) are of the same order of magnitude. A 15%

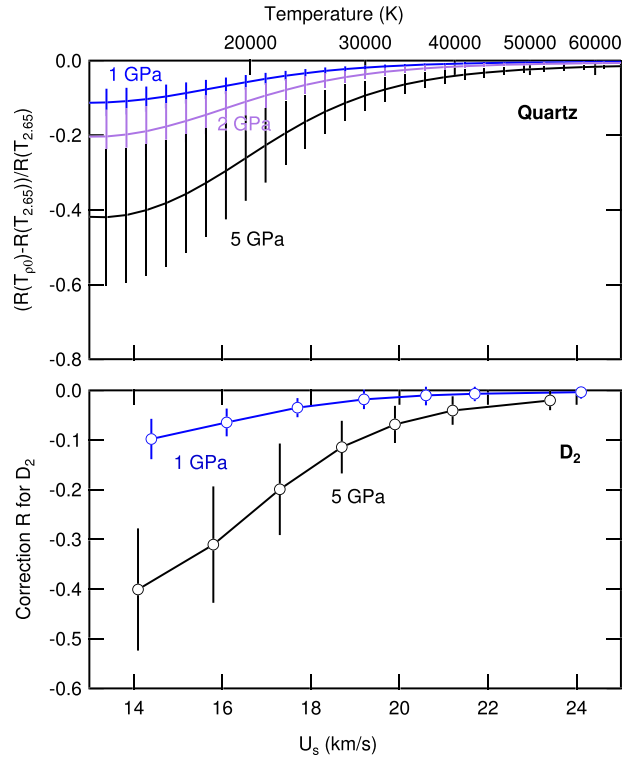


FIG. 12. Magnitude of the correction for the reflectivity of quartz $(R - R_{2.65})/R_{2.65}$ (upper figure) and for D_2 (lower figure) at $P_0=1$ and 5 GPa. The uncertainties are the systematic uncertainties coming only from the uncertainties in T_{qz} .

temperature correction (lower Figure 11) is observed for a 5 GPa precompression. Note that both the precompression correction on the quartz temperature and on the quartz reflectivity affect the final inferred sample temperature.

3. Re-analysis of helium, deuterium, and hydrogen shock reflectivity and temperature data

Figures 13–16 present a comparison between the published data on helium and hydrogen and the data re-analyzed with the new empirical model.

In the previous reports,^{4,5} the precompression was accounted for with a slightly different approach: the temperature shift expected for a precompressed Hugoniot was

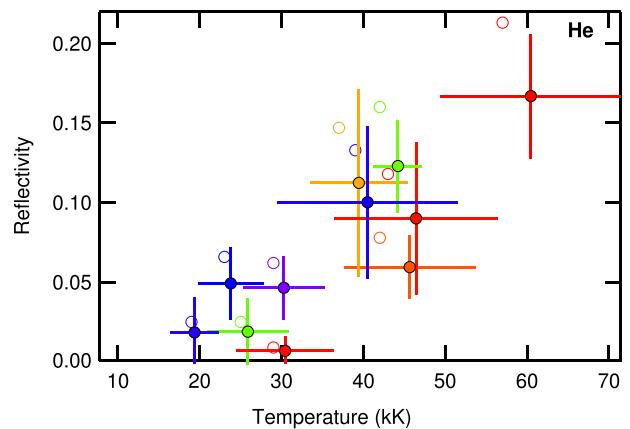


FIG. 13. Helium shock reflectivity as a function of the shock temperature: open and solid symbols are, respectively, the published data⁴ and the re-analyzed ones. The colors show the initial pressure as in Figure 7.

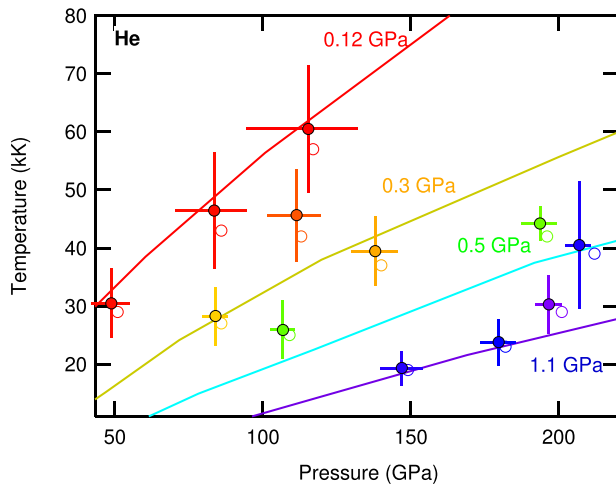


FIG. 14. Helium shock temperature as a function of the shock pressure: open and solid symbols are, respectively, the published data⁴ and the re-analyzed ones. The initial pressure indicated in colors corresponds to the experimental initial pressure. DFT-MD simulations³⁰ for 0.12 GPa (red), 0.35 GPa (orange), 0.85 GPa (light blue), and 1.8 GPa (purple) are also presented (solid lines).

estimated using a Grüneisen model³³ calibrated against the difference in shock temperature at a given pressure between fused silica and quartz. Similarly, the influence on the reflectivity was modeled based on the observed difference in onset and maximum reflectivity between fused silica and quartz.

At the relatively modest precompression achieved in these previously published datasets, the new precompression correction does not strongly affect the results. Instead, the changes observed for the temperature and the reflectivity are mainly arising from the fit used for the quartz reflectivity experimental data along the quartz principal Hugoniot.⁴

The change in helium shock temperature is small but the reflectivity appears lower than previously thought. This suggests lower electronic conductivities in the explored temperature-density domain. We observe that reflectivity saturation has not been reached yet but on-going measurements aiming at higher pressures and densities might reach the expected reflectivity saturation.

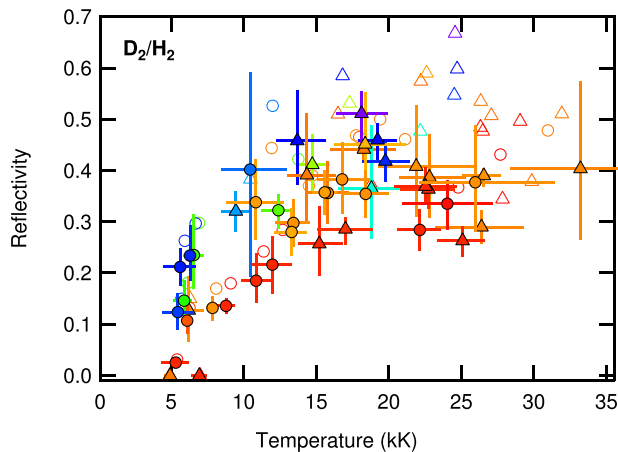


FIG. 15. Hydrogen and deuterium shock reflectivity as a function of the shock temperature: open and solid symbols are, respectively, the published data⁵ and the re-analyzed ones. Triangles are D₂ data and circles H₂ data. The colors show the initial pressure as in Figure 8.

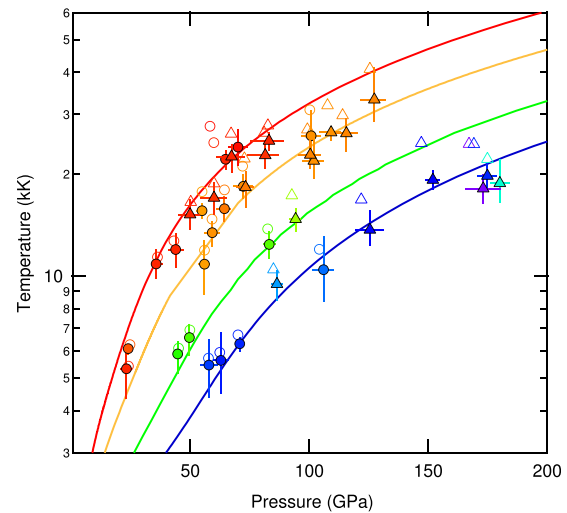


FIG. 16. Hydrogen and deuterium shock temperature as a function of the shock pressure: open and solid symbols are, respectively, the published data⁵ and the re-analyzed ones. Triangles are D₂ data and circles H₂ data. The solid lines are DFT-MD calculations from Caillaud *et al.*³² The colors show the initial pressure as in Figure 8.

For hydrogen, as for helium, the decrease in reflectivity reduces the temperature and yields a better agreement between the data and the DFT-MD calculations³² above 10⁴ K.

III. CONCLUSION

Laser driven shocks on precompressed samples allow reaching completely uncharted territories in the phase diagram of low-Z system and provide solid benchmarks for advanced warm dense matter theories and planetary science models, but require to carefully account for the precompression of the quartz reference, in particular, when the precompression exceeds a few GPa. The essential calibration fits and empirical analytical corrections to use quartz as a standard are summarized in Table I. Future refinements of the quartz standard, and, in particular, knowledge of the release behavior, should improve the precision and accuracy of the past and future experiments.

ACKNOWLEDGMENTS

Authors gratefully acknowledge discussions with Stewart Mc Williams, Florent Occelli, and Dylan Spaulding. Part of this work was prepared by LLNL under Contract No. DE-AC52-07NA27344 with support from LLNL LDRD program and the U.S. Department of Energy through the joint FES/NNSA HEDLP program.

APPENDIX: CHARACTERIZATION OF THE INITIAL PRE-COMPRESSED STATE

1. Initial density

The initial mass density is an important parameter for shock compression experiments. Here, we use the experimentally determined equations of state at room temperature in the GPa pressure range to infer the initial density of the sample and the quartz reference plate.

The high pressure static compression of silica polymorphs has been extensively studied. α - quartz, the stable phase at ambient conditions can be compressed to almost 20 GPa at room temperature without any phase transition, despite the fact that high pressure polymorphs coesite and stishovite become the stable phases above, respectively, 2 and 8 GPa.³⁴ The density of quartz at ambient conditions is $\rho_{00} = 2.649 \text{ g/cm}^3$.^{35,36} Accurate ultrasound measurements up to 1 GPa (Ref. 37) determined α - quartz room temperature bulk modulus $B_0 = 37.5(0.2)$ GPa and its first derivative $B'_0 = 4.7(0.5)$, data that can be used directly in the Birch-Murnaghan equation that determines the final density as a function of pressure. These values are in very good agreement with x-ray diffraction experiments and state-of-the-art *ab-initio* simulations to pressure above 20 GPa giving $B_0 = 37.7(3)$ GPa and its first derivative $B'_0 = 4.9(1)$.³⁸ The relative uncertainty on the initial density $\Delta\rho_0/\rho_0$ is then dominated by the uncertainty on B'_0 and is 0.3% at 6 GPa and 0.6% at 10 GPa.

2. Refractive index

VISAR measurements of reflecting shock velocity in transparent media actually measure an apparent velocity $U_{Sapp} = n_0 U_S$. In order to obtain the shock velocity U_S , we therefore need to know n_0 the refractive index at the VISAR wavelength (532 nm on Omega), room temperature, and initial pressure P_0 for both the reference and the sample.

We use z-cut α -quartz plates. At ambient conditions, the refractive index for the ordinary rays of quartz crystal³⁹ at 532 nm is $n_{Q00,532} = 1.54687$. The refractive index of quartz is known to increase with pressure. We updated the index pressure variation using literature experimental data⁴⁰ and recent elastic constants.^{37,41} A linear fit as a function of density gives the index of precompressed quartz $n_{Q0} = n_{Q00} + 0.1461(1)(\rho_0 - \rho_{00})$.

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⁹Two effects contribute to modify the SOP throughput on each laser shot involving diamond anvil cell: absorption by the diamond anvil and vignetting. Radiations generated by the ablation plasma can cause photo-

ionization of the diamond anvil facing the VISAR/SOP collection optics and attenuate the transmitted thermal emission from the shocked reference and sample. Similarly, optimum geometry of the DAC assembly for mechanical performances (in order to maximize the precompression) can compromise the optical performances and introduce vignetting. Measurements relative to the quartz *in-situ* standard upon breakout of the shock from the quartz to the sample are not affected by these issues.

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⁴¹We recalculate the quartz refractive index pressure dependence from interferometry data up to 0.7 GPa. We use the most recent elastic constant³⁷ instead of Bridgman's values⁴² to infer the quartz plate thickness and the density under pressure. We find a 1.8% lower linear compressibility at max pressure that translates into a slightly slower increase of the index with pressure. We assume that the optical dispersion between 589 nm (wavelength at which the pressure dependence has been measured) and 532 nm is pressure independent in the range explored here ($p < 10$ GPa). A simple linear fit of the index as a function of density⁴³ is then used to extrapolate up to the initial density ρ_0 .

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