Hugoniot and temperature measurements of liquid hydrogen by laser-shock compression

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Abstract. Hydrogen at high pressure in the fluid state is of great interest for target design of inertial confinement fusion and understanding the interior structure of gas giant planets. In this work, we successfully obtained the Hugoniot data for liquid hydrogen up to 55 GPa under laser-driven shock loading using impedance matching to a quartz standard. The shocked temperature was determined simultaneously by the brightness temperature. The compression and temperature along the principal Hugoniot are in good agreement with theoretical models. High reflectivity of hydrogen was observed at 40 GPa, which suggests the fluid becomes conducting.

The properties of hydrogen at high pressure and high density are of great scientific interest. The equation of state (EOS) of hydrogen at these conditions is essential for modeling of the interior structure of gas giant planets [1]. The EOS of hydrogen isotopes has important practical applications for inertial confinement fusion [2]. Chemical free-energy models [3] and ab initio simulations [4] have been used to predict the properties of warm dense hydrogen, but the results vary widely and have not converged yet. Therefore accurate experimental EOS data for hydrogen are required for evaluation of the theoretical models and for further understanding of the fundamental nature of hydrogen. The principal Hugoniot for liquid deuterium has been measured up to 220 GPa using laser-driven shock waves [5]. For the case of liquid hydrogen, on the other hand, the Hugoniot was studied experimentally only to 10 GPa by a gas gun and explosive method [6, 7]. The metallization of hydrogen on the Hugoniot is expected to occur at much higher pressure. In this work, we carried out laser-shock experiments of liquid hydrogen.
Figure 1. Sketch of the cryogenic hydrogen target. The quartz plates were glued to Kovar flanges attached to a copper cell filled with liquid molecular hydrogen at 15 K. The thickness of the quartz was 50 μm for both of them. The laser-side quartz was deposited by 40 μm aluminum to reduce the x-ray fluence from ablation plasma.

Figure 2. Pressure $P$ versus compression $\rho/\rho_0$ for the principal Hugoniot of liquid hydrogen. Data are from Dick & Kerley [6] (blue triangles), Nellis et al. [7] (green squares), and this work (red circles). Also shown are theoretical predictions from the model EOS of Kerley [3] (solid line), quantum molecular dynamics simulations [4] (dashed line), and the linear mixing model [10] (dot-dashed line). For reference, the Hugoniot data for liquid deuterium are shown by gray diamonds, and gray curve is the EOS model for deuterium.

to pressures exceeding 10 GPa in order to make a quantitative comparison of the hydrogen Hugoniot around the metal transition with the deuterium data.

The experiment was performed on the GEKKO/HIPER laser facility at the Institute of Laser Engineering, Osaka University. Laser energies between 0.8 and 1.4 kJ were delivered using a nominally square pulse of 2.5 ns in duration. The laser focal spot of 600 μm in diameter was smoothed using Kinoform phase plates. This resulted in average laser intensities between 5 and $8 \times 10^{13}$ W/cm². Figure 1 shows the experimental setup and target arrangement. The cryogenic hydrogen targets consisted of two α-quartz plates, by which the hydrogen layer was sandwiched. In this target, the quartz was designed to be used not only as a window material but also as a reference standard for the impedance-matching measurement.

Shock velocities in the laser-side quartz $U_{sQ}$ and hydrogen $U_{sH}$ were measured using a line-imaging velocity interferometer system for any reflector (VISAR) [8]. At the high pressure involved in these experiments, shock waves in the initially transparent quartz and hydrogen are reflecting. Then the VISAR can provide a direct time-resolved measurement of shock velocities in these media. Two VISARs were run concurrently on each shot to resolve the $2\pi$ phase-shift ambiguities at shock breakout. The velocity sensitivities were 4.14 and 14.53 km/s/fringe for quartz, and 5.71 and 20.05 km/s/fringe for liquid hydrogen. The time-resolved VISAR measurements allowed velocities to be tracked continuously during transit through both the standard and sample. Shock velocities immediately before and after the quartz-hydrogen interface were adopted for the impedance-matching analysis. The quartz Hugoniot was previously established to have a linear $U_s-U_p$ relation [9]. Impedance-matching occurs at the intersection of the hydrogen Rayleigh line and the release isentrope of quartz. Because of
the low shock impedance of hydrogen, quartz release curves from several hundreds of GPa down to very low pressure are needed. In this work, the quartz release is approximated by a reflection of the principal Hugoniot in the $P-U_p$ plane.

Figure 2 depicts the principal Hugoniot in the $P-\rho$ plane. These results are shown along with the previous experimental data [6, 7] and theoretical models calculated for hydrogen [3, 4, 10]. The pressure range of hydrogen obtained in this work was 25-55 GPa. The highest pressure is more than 5 times higher than the previous data. The compression $\rho/\rho_0$ of hydrogen ranges from 4.1 to 5.3, which is mostly comparable to the models but slightly softer at 30-40 GPa. The compression of deuterium has been measured as 3.4-4.4 at the same pressure range [11, 5], so that hydrogen exhibits systematically higher compression compared to deuterium. This trend is qualitatively consistent with theoretical predictions [3] and can be seen in the earlier data below 10 GPa [7].

Using a streaked optical pyrometer (SOP), the temperature $T$ of shocked hydrogen was measured simultaneously [12, 13]. We extracted the temperature by fitting the absolute spectral radiance to a gray body Plank spectrum with $T$ as a fit parameter. The radiance was obtained in a single spectral band centered at 455 nm with a bandwidth 38 nm. The emissivity $\varepsilon$ is related to the reflectivity $R$ of the shock front through $\varepsilon(\lambda) = 1 - R(\lambda)$. For the reflectivity at the SOP wavelength, we adopted the reflectivity measured at the VISAR wavelength assuming a weak dependence of $R$ on the wavelength. Absolute reflectivity is determined by comparing the shock reflectivity to that from the aluminum surface which has a known value. The obtained $R$ of shock front in hydrogen was $16\pm2\%$ at the shock velocity $U_{sH} \approx 25$ km/s corresponding to the pressure $P_H \approx 40$ GPa. The result of shocked temperature was $0.78\pm0.10$ eV at $U_{sH} = 24.9\pm0.5$ km/s (see Fig. 3). Theoretical models predicted that Hugoniot temperatures of liquid hydrogen are higher by a factor of $\sim 1.3$ than those of liquid deuterium at the same pressure [3]. Deuterium temperature was measured as 0.5-0.7 eV around 40 GPa by previous shock experiments [14]. Although the uncertainty in temperature measurements is still large, the hydrogen $T$ obtained in this work is higher than the deuterium data. At this temperature, molecular hydrogen begins...
to dissociate and the fraction reaches to $\sim 30\%$ according to the Kerley’s model [3].

Drude-type models are often applied to parameterize the optical properties of liquid metal. Within the Drude description, the complex index of refraction is given by $n^2 = 1 - (\omega_p^2/\omega^2)(1 + i/\omega\tau_e)^{-1}$ where $\omega_p = (4\pi n_e e^2/m_e)^{1/2}$ is the plasma frequency, $n_e$ is the carrier density, $e$ is the electron charge, $m_e$ is the electron mass, and $\omega = 2\pi c/\lambda$ is the optical frequency. Here the electron relaxation time is assumed to be $\tau_e = R_0/v_F$ where $R_0$ is the interparticle spacing and $v_F$ is the electron Fermi velocity. We adopt $R_0 = 0.126$ nm corresponding to a density 0.4 g/cm$^3$. Figure 4 shows the Drude reflectivity of liquid metallic hydrogen derived as a function of $n_e$. High reflectivity is produced when the carrier density exceeds the critical density $n_c$ defined by $\omega_p^2(n_c) = \omega^2$. The critical density is $n_c = 3.9 \times 10^{21}$ cm$^{-3}$ at $\lambda = 532$ nm. Taking $R = 16\%$, the carrier density is given by $1.8 \times 10^{22}$ cm$^{-3}$, which is about 15% of total hydrogen number density. The Fermi energy estimated from this carrier density is 2.5 eV, which is higher than the shock temperature. Based on the Drude model, the reflectivity increases with the carrier density and saturates at $R = 36\%$ for the fully ionized case. It will be interesting to confirm the saturated value of the reflectivity by future experiments.

The hydrogen Hugoniot obtained in this work is strongly dependent on the quartz EOS. Although the shock Hugoniot is relatively well determined, the EOS data along the release curves are still model dependent. For comparison, we calculated a release isentrope from the Kerley 7360 model for $\alpha$-quartz [15] and obtained the corresponding Hugoniot data, $U_{sH}$ and $P_{sH}$. At higher pressure, the mirror curve is higher than Kerley’s model. But just above 50 GPa, they cross and the Kerley model gives a larger particle velocity. The Hugoniot pressure $P_{sH}$ is higher by 2% and the density $\rho_{sH}$ larger by 6% compared to the mirror approximation case. Apparently, improvement of the quartz EOS is essential for the further development of Hugoniot measurements using quartz standards.

In summary, we have obtained the Hugoniot data $P$-$\rho$-$T$ for liquid hydrogen in an unexplored range of pressure up to 55 GPa. The results demonstrate that the hydrogen Hugoniot cannot be scaled by density from the deuterium data. As for the study of planetary interiors, the hydrogen EOS data at much higher pressure are required since the transition to metallic hydrogen is anticipated to be at $P \sim 200$-400 GPa in Jupiter [1]. However, the hydrogen temperature must be kept lower because the Hugoniot temperature at this pressure range is too high to reproduce Jupiter’s conditions. Therefore off-Hugoniot measurements of hydrogen by means of reflection shocks [16] and/or precompressed samples [17] will be an important next step.

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