[TYPESETTER: websum: Researchers have measured the equation of state of hydrocarbon in a high-density regime necessary for accurate modelling of the oscillations of white dwarf stars.]

A measurement of the equation of state of carbon envelopes of white dwarfs

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White dwarfs represent the final state of evolution for most stars[1–3]. Certain classes of white dwarfs pulsate[4,5], leading to observable brightness variations whose analysis with theoretical stellar models probes their internal structure. Modelling of these pulsating stars provides stringent tests of white dwarf models and a detailed picture of the outcome of the late stages of stellar evolution[6]. However, the high energy density states existing in white dwarfs are extremely difficult to reach and to measure in the laboratory, and, as a result, theory is largely untested at these conditions. Here we report measurements of the relationship between pressure and density of hydrocarbon along the principal shock Hugoniot, i. e., equations describing the state of the sample material before and after the

passage of the shock derived from conservation laws, to within five percent. The observed maximum compressibility is consistent with theoretical models that include detailed electronic structure. This is relevant for the equation of state of matter at pressures ranging from 100 million to 450 million atmospheres, where the understanding of white dwarfs is sensitive to the equation of state and where models differ considerably. The measurements are used in calculations of white dwarfs and inertial confinement fusion experiments[7,8], and we predict an increase in compressibility due to ionization of the inner core orbitals of carbon. We also find that detailed treatment of the electronic structure and the electron degeneracy pressure is required to capture the measured shape of the pressure–density evolution for hydrocarbon before peak compression. The equation of state of the white dwarf envelopes, which are partially ionized, partially degenerate, with non-ideal plasmas, remains one of the weakest links in white dwarf constitutive physics[9].

At pressures of a million to a billion times that of the Earth's atmosphere (megabar to gigabar), core electrons can affect the compressibility of stellar material by altering both the amount of stored internal energy and the plasma pressure through ionization. At these pressures, the excitation energies of the core electrons and the amount of ionization are difficult to predict[10]. Depending on how these details are modelled, the equation of state (EOS) along the shock Hugoniot (for example, pressure versus compression in this work) can vary by nearly 10%[11], which is important for white dwarf modelling[12]. Benchmarking EOS models for a hydrocarbon is directly relevant to the modelling of white dwarf stars of the hot DQ class that is characterized by a degenerate carbon and oxygen core surrounded by an envelope of mostly carbon[13,14]. Uncertainty in EOS and opacity models can affect the extent and properties of mixing within the convection zone, affecting the inferred surface abundances and pulsation properties of pulsating white dwarf stars with carbon-rich envelopes[1,13–15] (see Methods). In addition, instability growth in laboratory inertial confinement fusion experiments[7] has been shown to be sensitive to EOS modelling[16,17], with more compressible models leading to higher instability growth rates. Until now, no laboratory measurements of the shock Hugoniot have been reported at pressures exceeding ~60 Mbar for any material, and theoretical models in this regime have not been constrained by experimental data. A single dataset at gigabar pressures recorded in underground nuclear explosions for aluminium[18] has uncertainty that is too large to distinguish between theoretical models.

We present laboratory EOS measurements in a regime where none currently exist, enabling tests of theoretical models at pressures exceeding 100 million atmospheres. We access the atomic pressure regime

>300 Mbar (see Methods), in which extreme temperatures and densities ionize inner-core electron orbitals and can change the shape of the pressure-versus-density function (P–ρ) along the Hugoniot states. We resolve the shape of the Hugoniot response by measuring a continuous sequence of data points in a single shot, using a spherically convergent geometry[19–22]. The combination of small scatter and precision of the data effectively constrains EOS models. Alternative techniques such as planar dynamic compression experiments[23,24] are restricted to pressures of <60 Mbar and can only access one Hugoniot point in a single shot. Accessing the shock regime in this work required us to increase the laser driver energy relative to ref. 19 and use multiple coalescing shock waves to generate a single stronger shock. Operating in an intense X-ray drive environment required mitigation of sources of preheat and background from electrons and X-rays generated from the radiation drive, as well as symmetric shock generation. To interpret the experimental measurements in this high-pressure regime, in which the opacity at the shock front deviates from the cold opacity, we developed a simultaneous density and opacity analysis method[25] (see Methods).

These experiments were performed at the National Ignition Facility (NIF)[26] where 1 MJ of 351-nm laser light was delivered to the inside of a gold cylinder (hohlraum) creating an X-ray radiation bath with a maximum radiation temperature (T_r) of 294 eV (nearly 3.5 million degrees Kelvin); see Fig. 1. The X-ray drive is absorbed by a spherical sample mounted in the centre of the hohlraum, in an outer region called the ablator. The ablator heats and expands, which launches inwardly converging shock waves via the rocket effect towards the centre of the solid sphere. The shocks coalesce into a single stronger shock reaching near-gigabar pressures at radii of ~100 μm. We measure the Hugoniot at the shock front as it travels inward, where the shock-front pressure continuously increases owing to convergence. The shock travels inward faster than the converging material behind it, enabling continuous tracking.

The density and pressure at the shock front are determined from temporally and spatially resolved streaked X-ray radiography measurements. The shock-front speed is determined from the radiograph by tracking the shock-front radial position as a function of time[21]. The radial density profile was extracted from the transmission of a 9-keV X-ray source (back-lighter) and analysed by profile matching[20]. At these pressures, the opacity decreases relative to the cold opacity as a result of carbon K-shell ionization. The opacity is simultaneously unfolded from the measured back-lighter transmission[25]. The mass density is further constrained by conserving the total mass contained within a higher-opacity fiducial layer which is visible in the radiograph (radiographic Ge marker layer; see Fig. 1b) and by matching the density profile to the known density of the unshocked material ahead of the shock. Using Hugoniot jump relations, we calculate the pressure, P, from the measured shock-front speed, D, and the mass density, ρ (refs. [21,25]):

$$P = P_0 + D^2\rho_0^2(1/\rho_0 - 1/\rho) \qquad (1)$$

where ρ_0 is the initial mass density and P_0 is the initial pressure of the unshocked material (see Methods).

The experimental data are shown in Fig. 1d, where the shock front is labelled on the image. The shock travels at speeds of up to 150–220 km s^{-1}, traversing the 1-mm sample in ~9 ns. We verify that the shock converges spherically by measuring the shock-front symmetry to be within the radiographic resolution of 25 μm at a shock radius of 200 μm through tracking of the radial density profiles on both sides of the sample in the equatorial plane. We also determine the symmetry through imaging of X-ray self-emission at ~9 ns when the shock wave collapses at the centre of the sample (shock flash). The symmetry of the shock flash was measured using penumbral imaging[27] to be within 0.5 ± 0.3 μm at a shock radius of 12 μm in the equatorial direction.

Figure 2 shows the measured opacity at the shock front normalized to the cold material opacity (red curve) as a function of shock-front pressure. Here, the red shaded region and outer red lines correspond to uncertainty contours of $\pm 1\sigma$ in the measurement. Also shown are calculations of the normalized opacity with and without trace amounts of fluorine present in the samples (see Methods), using detailed configuration accounting (DCA)[28] (black curves). Calculations of the carbon K-shell (inner shell) occupation fraction by DCA (blue circles) show that the calculated drop in opacity is directly correlated to the K-shell occupation, or ionization of the carbon K-shell. The measured opacities in these experiments are consistent with the modelling at pressures up to ~300 Mbar and slightly higher than theory at higher pressures. The measured drop in opacity indicates distortion of the carbon inner shell with ~63% occupation at 450 Mbar.

The measured sequence of data points along the shock Hugoniot is shown in Fig. 3 (red curve) with errors that correspond to uncertainty contours of 1σ (red shaded region). Also shown are previous shock Hugoniot measurements[19,29–31], theoretical calculations made with molecular dynamics based on Kohn–Sham density functional theory (KS-DFT)[32] (orange curve), and models using an average-atom (AA) single ion-in-jellium description with the electronic structure based on Kohn–Sham density functional theory (AA-DFT)[33–36] (solid black curve) and on Thomas–Fermi–Dirac theory (AA-TFD)[37] (dot-dashed black curve); see also Methods. These models are commonly used to generate EOS tables for, inertial confinement fusion experiments, for example.

The new extreme-pressure measurements of the P–ρ Hugoniot presented here access the Hugoniot structure associated with the ionization of the carbon core (K-shell) electrons. The shape of the Hugoniot in the range ~100–1,000 Mbar is a result of the ionization of the core electrons of carbon (hydrogen being fully ionized under these conditions). First, ionization absorbs energy from the shock, and the material becomes more compressible by an amount that depends on the interplay between the ionization energy and interactions in the plasma. Then, at higher pressures, the curve swings back to lower compression, owing to reduced conversion of shock-wave energy to internal degrees of freedom. EOS models that include the electronic shell structure (for example, AA-DFT) show a sharper bend in the Hugoniot at these pressures and higher maximum compression than models that lack electronic shells (for example, AA-TFD) and are in better agreement with the measurements. Path-integral Monte Carlo calculations are in general agreement with AA-DFT for hydrocarbons[11], and thus also accurately predict the shape of the high-pressure Hugoniot and maximum compressibility for this hydrocarbon.

We provide data that access the conditions deep in the convection zone of hot DQ white dwarf stars. Figure 4 shows a sequence of interior models for the cooling of white dwarfs with a carbon–oxygen core and pure carbon envelope. As the star cools, its structure moves leftward in the diagram. Convectively unstable regions (in red) are associated with the partial ionization of carbon. The experimental points at the highest pressures and temperatures reach the conditions of partial ionization of the core electrons of carbon that are similar to those at the bottom of the convection zone of hot DQ stars. This is the region most responsible for the driving of unstable pulsation modes[5,14] and where EOS models show the greatest range of variability. By constraining EOS models in this regime, these data can contribute to more accurate models of hot DQ stars and of their complex origin, thought to be from stellar mergers or through the late helium flash of asymptotic giant branch stars.

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Fig. 1 | Experimental configuration. a, Schematic of the target showing laser beams incident on the inside of a gold hohlraum, a solid spherical sample inside the hohlraum, and the X-ray back-lighting configuration. **b**, Diagram of the sample configuration, i.e. a portion of the sphere from (a), showing layer thicknesses and level of Ge dopant (in atom per cent) in the GDP, glow discharge polymer, ablator. **c**, Laser drive (blue) and back-lighter (red) power profiles versus time. The calculated radiation temperature versus time is also shown (black curve). **d**, Streaked X-ray radiography data showing the shock front and shock flash at the core. The spatial fiducial line is used for diagnostic warp correction.

Fig. 2 | Opacity of shock-compressed $C_9(H)_{10}$ at 9 keV. Measured opacity at the shock front as a function of pressure along the shock Hugoniot (central red curve). The shaded region and outer red curves correspond to uncertainty contours of ±1σ in the measurement. Also plotted are theoretical curves of the normalized opacity versus pressure with and without 1 at% F (black curves), along with the fraction of C K-shell occupation (blue circles) calculated through detailed configuration accounting (DCA)[28].

Fig. 3 | $C_9(H)_{10}$ shock Hugoniot measurements: Measured pressure versus mass density (ρ) normalized to the initial density (ρ_0) along the shock Hugoniot (red curve and shaded region). Also plotted are previous experimental data (blue line, ref. [17]; squares, ref. [29]; crosses, ref. [28]; triangles, ref. [27]) and theoretical modelling of the Hugoniot using Thomas–Fermi–Dirac theory (black dashed curve), AA-DFT (black curve) and KS-DFT (orange curve).

Fig. 4 | Regime of white dwarf stars accessed by measurements: Density–temperature diagram for the evolution of a white dwarf (WD) star with a mass of $0.6M_{\mathrm{sun}}$ composed of a carbon/oxygen core surrounded by a pure carbon envelope. The surface of the star occupies the region of the curves in the lower left and the core occupies the region of the curves in the upper right. Models start from hot and young state (right) and evolve leftward to older and colder structures, with the bold lines corresponding to hot DQ stars[3]. Convective regions in the stars are shown in red. The regime probed by the experiment is shown by the thick black line, with temperatures estimated from a model EOS[33–35].

METHODS

Atomic pressure regime

The atomic pressure regime is the pressure greater than that required to significantly distort core electron orbitals, P_a, and can be estimated by

$$P_a = E_H/r^3_{\mathrm{Bohr}} = 294 \text{ Mbar} \quad (2)$$

where E_H is the Hartree energy and r_{Bohr} is the Bohr radius.

Simultaneous mass density and opacity unfold method

The measured transmission radiograph of an X-ray back-lighter source is sensitive to the mass density, or compression, and opacity of the sample material (see Extended Data Fig. 1). When the opacity of a material deviates from the known cold opacity, this must be considered in order to extract density information from the radiograph. As there are no existing experimental measurements of the opacity of any material at the pressures in this work, we simultaneously extract the opacity from the radiograph. Here we use radial profile matching and optimization of the density and opacity to find best fits to the experimental data.

First, an initial guess of the density and opacity profiles are chosen based on physical parameters and radial transmission profiles are calculated. The calculated profiles are compared to the measured transmission profiles and are iterated until a good match to the experimental data are found. Uncertainty contours are determined from the quality of the profile fits to the radiograph. Since the relationships between transmission intensity, mass density and opacity are linear, additional information and assumptions are needed to constrain the range of profiles that provide a fit to the data. This is depicted in Extended Data Fig. 1, with a range of possible compressions and opacities that correspond to a given measured intensity (black dashed curves). Additional information for constraining the compression profile includes the known initial density ahead of the unshocked material and known mass of the sample material inside a Ge radiographic marker layer physically located inside the sample. This constraint on density provides a further constraint on the opacity. This is done continuously as a function of time as the shock traverses the sample. An example shock trajectory is shown in Extended Data Fig. 1 (red dashed curve). We also include an additional constraint that the opacity and density at the shock front vary smoothly in time and do not include discontinuous changes; see ref. [25] for more details on the radiographic unfold analysis method and derived uncertainties.

The uncertainty contours of $\pm 1\sigma$ in the measurement shown in Fig. 3 correspond to statistical uncertainty from simultaneously fitting of the parameterized opacity and density profiles to the measured radiograph. The parametrized fits were perturbed around the best fit and a probability distribution was constructed. This analysis includes uncertainty due to noise in the measurement. Systematic uncertainties from the magnification and the sweep speed of the X-ray streak camera correspond to ~2% in compression and ~7% in pressure, and mainly affect the location but not the shape of the Hugoniot. The measurements in Fig. 3 and the additional measurements provided in the next section are consistent in both position and shape of the Hugoniot with AA-DFT.

Reproducibility

Extended Data Fig. 2 shows additional data obtained in this study in a separate experiment on NIF (green curves and green shaded region), included to demonstrate consistency of the analysis approach and to support our conclusions. This experiment (shot N130103-009-999) used the same sample material over a similar pressure range and was fielded at cryogenic temperatures (24 K), with a measured initial density of 1.136 g cm^{-3} consistent with the fielding temperature. The uncertainty contours of $\pm 1\sigma$ are larger than the main experiment shown in Fig. 3 and Extended Data Fig. 2 (red curves and red shaded region) mainly as a result of using fewer laser beams to create the X-ray back-lighter.

Extended Data Fig. 2 shows that a slightly higher-density sample probes approximately the same theoretical shock Hugoniot at cryogenic temperatures as accessed at room temperature for both AA-DFT and AA-TFD over our pressure range. In the pressure range of these experiments the theoretical models deviate by roughly 1% in compression at the low-pressure range and overlay at higher pressures. This additional dataset is consistent with the data presented in Fig. 3 with a difference in the best fit compression of $<$1% at 107 Mbar, ~2% at 300 Mbar and ~3% at 450 Mbar. Both datasets show good agreement with models that include electronic structure in the AA-DFT approach.

Experimental configuration continued

The sample consisted of a solid sphere of poly(alpha methylstyrene) (PAMS, C_9H_{10}) with trace amounts of fluorine (1 at%) from the fabrication process uniformly distributed throughout the sphere, and a measured density of 1.085 ± 0.009 g cm^{-3} at ambient temperature. The fluorine concentration and uniformity were measured using energy-dispersive X-ray spectroscopy (EDS) and are calculated to have a negligible effect on the theoretical shock Hugoniot (see Extended Data Fig. 3). The spheres were coated with a plastic ablator (glow discharge polymer) that included doped layers of Ge to mitigate X-ray preheat from the hohlraum and act as a radiographic marker for the analysis. The laser drive was a two-shock pulse shape with a total drive energy of 1.1 MJ. The 9-keV, Zn Heα X-ray back-lighter was generated by using up to 16 NIF laser beams incident on a Zn foil delayed in time relative to the drive beams (see Fig. 1c). Our main focus is on the analysis of NIF shot number N130701-002-999, but we also use opacity information from NIF shot N130103-009-999[25]. Radiation hydrodynamic simulations using HYDRA[38] benchmarked to shock timing data[39] calculate the shock coalescence of the two shocks inside the ablator to be before the Hugoniot points are extracted. In these experiments hot electron generation from laser–plasma interactions were inferred from measurements[40] to be more than an order of magnitude lower than previous experiments using higher-pressure gas-fill hohlraums (He density of 0.96 -1.6 mg cm^{-3}), for example compared to shot N1401106 of ref. [41]. This was achieved by using a near-vacuum hohlraum helium gas-fill density of 0.03 mg cm^{-3}.

The calculated preheat of the sample inside the probed region as a result of the hot electrons is $<$1 eV. The estimated bulk sample preheat from the measured hohlraum X-ray background[42] is <1 eV inside the centre of the sphere and $<$2.5 eV at the outer probing radius of the sample, which does not cause the calculated shock Hugoniot to deviate outside of the measurement error from the un-preheated Hugoniot[22] for these shock strengths. Simulations also indicate that radiative heating of the sample ahead of the shock from the shock itself does not cause deviation from the un-preheated Hugoniot outside of the measurement error for pressures up to ~450 Mbar.

This can be seen in Extended Data Fig. 4, which includes extracted shock-front densities and pressures from radiation hydrodynamic simulations of the experimental configuration (red points). Also plotted are curves of the input Hugoniot to the simulations[33–36] and ±2% deviation from the input Hugoniot. At lower pressures, ~100 Mbar, the extracted Hugoniot deviates from the input Hugoniot owing to bulk preheat from the hohlraum radiation drive at the level of ≤2%. Then, as the shock-front pressure increases, this level of preheat becomes a smaller perturbation on the Hugoniot and the extracted points follow the input Hugoniot more closely. At higher pressures of $>$450 Mbar, radiative preheat of material ahead of the shock front from the shock front itself starts to play a role. At 450 Mbar, this preheating causes the Hugoniot to deviate from the input Hugoniot at the level of 2% in the direction of lower compression. We restrict the data record in Fig. 3 to ~450 Mbar for this reason. At 720 Mbar, these calculations suggest lower compressions of 4.0, owing to radiative preheat and deviation from the input Hugoniot of $>$12%. However, the full data range reaching pressures of ~720 Mbar, shown in Extended Data Fig. 5 (red and purple curves and shaded region), does not show this level of reduction in compression at high pressures, which could be an indication that the modelling is overestimating radiative shock-front preheat. Here, the central curves represent the best fit to experimental data, and the shaded regions correspond to $\pm 1\sigma$ uncertainty.

Theoretical EOS models

The theoretical Hugoniot calculations for polystyrene presented in Fig. 3 for AA-DFT and AA-TFD are equivalent to widely used EOS models for ICF experiments (see Figure 9 of ref. [8]). Equal pressure and temperature additive-volume prescriptions were used to mix the C and H contributions. For AA-TFD, both

H and C were treated with Thomas–Fermi–Dirac theory. For AA-DFT, the carbon EOS used ref. [35] and the hydrogen EOS used ref. [36]. Here, the AA-DFT model corresponds to EOS table LEOS 5112, which is the CH version of LEOS 5400[43] (for glow discharge plastic), widely used to model ICF ablators. The AA-TFD model corresponds to SESAME 7593 (see ref. [8] for a more detailed description).

Pulsating white dwarfs

Hot DQ white dwarfs have an envelope composed of mostly carbon mixed with a modest amount of helium[13,14]. They are thought to result either from a late helium-shell flash in the post-asymptotic giant branch evolution of stars or from stellar mergers. Most are highly magnetic, and photometric variability has been attributed to the relatively rapid rotation of stellar spots in and out of view. However, some hot DQ stars are probably pulsators[44,45]. Our data will enable probing of the latter types. These data may also lead to an understanding of the unusual low-mass pulsating white dwarf in the system SDSS J1152+0248 which is likely to have a hybrid carbon-oxygen-helium core[46].

Data availability

Source data are provided with this paper. Additional data are available upon request.

Code availability

Owing to its complexity, the data analysis algorithm that supports the findings of this study is available from D.C.S. upon reasonable request and will be outlined in more detail in a supporting publication.

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Author contributions A.L.K. wrote the manuscript and performed the design calculations. D.C.S. developed the analysis method and analysed the experimental data. B.B., B.M., M.J.M., M.E.M., J.N. and N.K. provided input and feedback to improve the data analysis method. T.D. fielded the experiments and follow on supporting experiments together with B.B., D.K. and A.L. L.X.B., J.L.D., S.H., P.A.S., A.A.C. and H.D.W. provided theoretical calculations of equation of state models included in this paper and/or provided theoretical models for understanding the data. G.F. and D.S. performed calculations of white dwarf stars and worked closely with S.H.G. and A.L.K. to understand the impact of this work for white dwarfs. W.R.J., J.A.G. and J.N.

provided calculations of theoretical opacities. W.S., F.E. and A.N. fabricated and characterized the targets for these experiments. D.C.S., S.H.G, R.W.F., P.N., B.A.R. and G.W.C. proposed the experiments and/or aided in obtaining experimental beamtime at NIF. All co-authors provided input on interpretation of the data and results and/or on their impact for white dwarf modeling.

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Additional information

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Extended Data Fig. 1 | Schematic of the radiographic analysis method. Normalized transmission intensity of an X-ray radiograph versus changes in compression and opacity. A given transmission corresponds to a range of possible compressions and opacities (black dashed curves). Density at the shock front is further constrained through knowledge of the initial material density ahead of the shock front and known mass contained within a region on the radiograph corresponding to a Ge marker layer in the sample. This further constrains the opacity and is included in the analysis as the shock traverses the sample. An example shock trajectory is denoted with a red dashed curve.

Extended Data Fig. 2 | Comparison of two $C_9(H)_{10}$ shock Hugoniot measurements. Measured pressure versus mass density (ρ) normalized to the initial density (ρ_0) along the shock Hugoniot. Green, data from shot N130103-009-999 fielded at 24 K; red (from Fig. 3), shot N130701-002-999 fielded at ambient temperature. Also plotted are the theoretical Hugoniots for AA-DFT and AA-TFD at 24 K (ρ_0 = 1.136 g cm^{-3}), indicating that the initial density conditions are predicted to access approximately the same Hugoniot states at high pressure.

Extended Data Fig. 3 | Sensitivity of the theoretical Hugoniot to fluorine. Calculations show insensitivity of the theoretical Hugoniot (AA-DFT[33–35]) to fluorobenzene solvent (C_6H_5F) for concentrations up to 20%, corresponding to 1% atomic fraction of fluorine (green curve). Concentrations of 0.5% F (red curve) and 0% F (blue curve) are also shown, but not visibly distinguishable.

Extended Data Fig. 4 | Simulations of the shock-front Hugoniot. Extracted shock-front compressions and pressures from radiation hydrodynamic simulations[38] of the experimental platform (red points). The theoretical shock Hugoniot[33–35] input to the simulations is also shown with ±2% deviation in compression from the input Hugoniot (black curves).

Extended Data Fig. 5 | Extended $C_9(H)_{10}$ shock Hugoniot measurements. Measured pressure versus mass density (ρ) normalized to the initial density (ρ_0) along the shock Hugoniot from this work (red and purple curves and shaded region). The purple curve corresponds to the extended dataset that may be impacted by radiative shock-front preheat. Also plotted are previous experimental data and theoretical modelling of the Hugoniot (see Fig. 3).