Measurement of Preheat Due to Nonlocal Electron Transport in Warm Dense Matter

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This Letter presents a novel approach to study electron transport in warm dense matter. It also includes the first x-ray Thomson scattering (XRTS) measurement from low-density CH foams compressed by a strong laser-driven shock at the OMEGA laser facility. The XRTS measurement is combined with velocity interferometry (VISAR) and optical pyrometry (SOP) providing a robust measurement of thermodynamic conditions in the shock. Evidence of significant preheat contributing to elevated temperatures reaching 17.5–35 eV in shocked CH foam is measured by XRTS. These measurements are complemented by abnormally high shock velocities observed by VISAR and early emission seen by SOP. These results are compared to radiation hydrodynamics simulations that include first-principles treatment of nonlocal electron transport in warm dense matter with excellent agreement. Additional simulations confirm that the x-ray contribution to this preheat is negligible.

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The thermodynamic properties and dynamic behavior of materials at extreme conditions of high energy density states are relevant to many astrophysical objects [1] and inertial confinement fusion (ICF) [2]. A particularly problematic state is warm dense matter (WDM) defined by moderately high temperatures of 0.1-100 eV, solid densities, and pressures above 1 Mbar. Under such conditions, ions are strongly correlated and the electron population is partially or fully degenerate making the theoretical description of WDM very challenging. Thus, robust measurements of the equation of state (EOS), structure, and transport properties of WDM are crucial to the understanding of many processes in the formation and structure of astrophysical objects such as Jovian planets or white dwarfs as well as the dynamics of the ICF implosions [3,4]. Heat and radiative transport through various layers influences the laver structure and convection of astrophysical objects, and electrical conductivity strongly affects magnetic fields generated by planetary core dynamos [5]. Alternative fusion schemes such as fast ignition rely on heating of fusion targets by the energy deposition of electrons [6,7]. Preheat of target components due to x rays and energetic particles in laser-driven high energy density systems is a well-known problem causing changes in initial conditions and multiple hydrodynamic instabilities in EOS and ICF experiments [8,9]. Specifically, electron transport in dense laser-heated plasmas holds the key to understanding many fundamental questions [10].

Despite great challenges, much progress has been made in the theoretical description of the structure, the EOS, as well as the transport properties of WDM [11,12]. Examples of remarkable work include ab initio quantum molecular dynamics (MD) simulations obtaining the thermal conductivity of warm dense hydrogen [13], the resistivity saturation in warm dense Al [14], and the charged particle stopping powers, and transport has been described both by using pure theory as well as with MD simulations [15,16]. The concept of nonlocal electron transport modeling in hydrodynamic simulations was first introduced to compute the delocalization strategy of the classical diffusion approach [17]. This model had a great impact and led to a consequent improvement of the hydrodynamic simulations in experimental data prediction. It was not until much later that the first attempt to include a real nonlocal transport model retaining on a first-principles approach based on kinetics came and addressed the necessity of using the proper physics [18].

In this Letter we present our recent work where we utilize a platform previously developed for direct measurements of temperature and shock velocity in order to study nonlocal electron transport in WDM [19]. The analysis of our data showed that the stronger drive used in this experiment caused nonlocal electrons to preheat the CH sample. A detailed theoretical study confirmed that this effect leads to observed shock velocities and temperatures well above those expected without preheat. In past years, there have been some experiments measuring the transport properties of electrons in dense plasmas [20] and radiation transport in standard materials [21,22]; however, no full characterization including the complete measurement of

plasma conditions with *in situ* measurements of transport coefficients has been achieved to date. A newly established diagnostic technique of x-ray Thomson scattering (XRTS) opens a novel path towards studying structure and transport properties in WDM [23]. XRTS is capable of obtaining information about the temperature, density, ionization state, as well as the microscopic properties of dense plasmas. If combined with other diagnostics such as velocity interferometry (VISAR), streaked optical pyrometry (SOP), or radiography, it can then provide a comprehensive measurement of the thermodynamic properties of WDM [19,24].

The experiment was carried out at the OMEGA laser facility at the University of Rochester [25]. The WDM conditions were created by a single shock driven by laser ablation from the surface of the target. Fifteen of the laser beams were overlapped to give a $\sim 7 \times 10^{14}$ W/cm² square drive with a 2 ns duration. The drive beams were frequency tripled to give a $\lambda = 351$ nm output and their spatial profile was smoothed with distributed phase plates [26]. The targets were planar layered structures consisting of a 25 μ m plastic (CH) ablator, a 2–3 μ m Au coating used to shield the x-ray radiation created at the critical surface of the laser-plasma interaction, a 70 μ m Al pusher, and 300 μ m of C₈H₈ polystyrene foam. The density of the foam, measured by soft x-ray transmission at 5.4 keV (Cr K- α source), was found to be $137.27 \pm 3.48 \text{ mg/cm}^3$ [27]. The polystyrene foam was made with the Hipe process and the pore sizes were measured with a scanning electron microscope (SEM) obtaining the average pore diameter of $1.26 \,\mu\text{m}$. The schematics of the target layouts are shown in Fig. 1.



FIG. 1. A schematic of the targets. The driven target sits inside the larger Au cone and consists of a plastic ablator, a Au radiation shield, an Al pusher, and C_8H_8 foam. The XRTS target also includes a second smaller cone containing a Ni backlighter and Ta slit used to collimate the x rays. Laser beams illuminate the ablator from inside of each cone. The VISAR target is identical, but has no backlighter and includes four steps on the back side of the foam. Additional Ta shielding was used to restrict the view of VISAR and XRTS.

The thermodynamic conditions in the shock wave traveling through the C₈H₈ foam were studied with a number of diagnostics developed for the platform including VISAR, SOP, and XRTS [19]. The XRTS and VISAR or SOP measurements were carried separately due to geometrical constraints, but the laser drive was kept the same in both cases and the platform has shown good shot-to-shot reproducibility. The shock velocity was measured by a two interferometer VISAR system operating at 532 nm by detecting shock break-out timing across four 40 μ m steps manufactured on the back side of the foam on the VISAR targets [28]. Supporting measurements of shock break-out timing were obtained from direct emission observed by the SOP system [29]. The average shock velocities at the time of break out were found to be 57.8 ± 3.8 , 64.0 ± 4.9 , and 67.5 ± 5.0 km/s for different shots. These shock velocities were rapidly decaying with significant slowing of the shock waves. SOP observed early emission that came ~ 1 ns before the VISAR signal confirming the presence of preheat in the foam.

The temperature was obtained from analytical fits to the broadening of the inelastic Compton feature in the XRTS spectra in the noncollective geometry [23]. The x-ray probe used for XRTS was He- α line emission at 7.8 keV generated by ten 1 ns backlighter beams with an intensity of $2-3 \times 10^{15}$ W/cm² focused onto 5 μ m thick Ni foil [30]. The x rays were then collimated by a 200 μ m diameter Ta slit placed 500 μ m away from the studied carbon sample, defining the scattering angle at 95° ± 5°. The scattering signal was detected by a spatially and spectrally resolved imaging x-ray Thomson spectrometer (IXTS), which utilizes a toroidally curved Ge (220) crystal coupled with a deep-depletion CCD camera [31]. The xRs code was used to compute analytical fits to the scattering spectra [32–34].



FIG. 2. X-ray scattering data from shocked C_8H_8 polystyrene foam. The best fit conditions were $T_e = 26 \pm 3$ eV, $n_e = 8.43 \times 10^{22}$ cm⁻³ and $Z \sim 2.4$. The error bars were estimated from a χ^2 fit (inserted image).

The XRS fits were calculated using a measured source spectrum and varied input conditions including the electron temperature T_e , density n_e , and ionization state Z and compared with experimental data as shown in Fig. 2. The temperature in each sample was obtained from the best fit to the data and the error bars were estimated using χ^2 fitting and matched to 1 standard deviation of the noise in the Compton peak, see the insert in Fig. 2. Some XRTS spectra had some contamination by direct emission from blow off Ni plasma, which was possible to be reduced from the data thanks to the spatial resolution of the IXTS instrument. In the case of shots 80 373 and 80 377 multiple lineouts could be retrieved from the data that corresponded to slightly different times during the shock wave evolution; i.e. the upper lineout corresponds to an later time. A clear trend is seen in the data: the shock temperature increases with time. The XRTS results are summarized in Table I.

The experimental results were first compared to EOS tables including SESAME 7593 [35] and FPEOS [36] in Hugoniot calculations confirming that CH temperatures in the range of 20-30 eV would require higher shock velocities. We tested this with simulations carried out by the high-energy density code CASSIO developed at LANL combining the radiation adaptive grid Eulerian (RAGE) code [37] with an implicit Monte Carlo treatment [38]. For these calculations, we used a 72 energy group structure, using Rosseland binning for the SESAME opacities [39]. Although the plastic ablator region heats up beyond 100 eV, the simulations show that no x-rays leak through to the foam target and there is no significant x-ray preheat. No explicit treatment of electron transport is included in the code. With no preheat included the shock velocities and temperatures in the simulation are lower than observed in the experiment. By introducing an artificial preheat of 1–10 eV (assuming the electrons stream through the target, heating uniformly), the simulation results better match the higher temperatures, but never reproduce the observed shock velocities. The effect of possible preheat was further studied by using spectral simulations. First, the radiation emission of the CH pusher during the laser irradiation $(T = 2 \text{ keV}, \rho = 7 \times 10^{-3} \text{ g/cm}^3, L = 750 \ \mu\text{m})$ in the range of 0.1-10 keV was modeled by using the FLYCHK code and this radiation was multiplied by the transmission

TABLE I. Summary of XRTS results.

Shot no.—lineout	XRTS delay	Temperature (T_e)	Ζ
80 375	5.5 ns	$17.5 \pm 2.5 \text{ eV}$	2.5
80 373 (lower)	6.0 ns	$24.5\pm4.5~\mathrm{eV}$	2.2
80 373 (upper)	6.0 ns	$27.0\pm2.5~\mathrm{eV}$	2.9
80 377 (lower)	6.0 ns	$23.0 \pm 3.0 \text{ eV}$	2.8
80 377 (middle)	6.0 ns	$25.5\pm2.5~\mathrm{eV}$	2.8
80 377 (upper)	6.0 ns	$26.0 \pm 3.0 \text{ eV}$	2.4
80 376	6.5 ns	$35.0 \pm 5.0 \text{ eV}$	2.2

of the Au and Al layers [40,41]. It was found that a flux of only 19 J/cm³ was absorbed in the foam. Another possible source of preheat is the heated Au radiation shield ($T = 35 \text{ eV}, \rho = 1.7 \text{ g/cm}^3, L = 30 \mu \text{m}$). This was evaluated by a similar approach and it was found that <110 J/cm³ would be absorbed in the foam.

In order to study the contribution of the nonlocal electron transport to the observed preheat we used the plasma Euler and transport equations hydro code (PETE), which is a Lagrangian fluid model [42]. In PETE, the plasma fluid (ions) is modeled by Euler equations, while the population of free electrons relies on the first-principles based kinetic model represented by the Bhatnagar-Gross-Krook (BGK) transport equation [43]. Such a treatment allows us to describe the nonlocal transport of electrons with respect to their mean free path (mfp), before being thermalized within the plasma fluid, thus providing the transport of energy. The results of the OMEGA shock wave simulations in 1D planar geometry are shown in Fig. 3. The propagating shock in the foam layer can be recognized by a set of arrows, which point to the sequence of positions of three steps manufactured on the back side of the target and show a very good agreement between the simulated and experimentally measured shock velocities and the strong decay in time. This is a natural consequence of the nonlocal BGK electron transport, which leads to a higher temperature when the shock propagates. We were unable to reproduce this behavior with the classical heat conduction model [44]. In every time step, the foam shock wave exhibits finite preheat. Its maximum thickness can be seen at the position $z = 250 \ \mu m$ in Fig. 3. Physically relevant conditions for such a preheat to occur are high temperature or, more precisely, a sufficiently high Knudsen number Kn = mfp/L, where L is the



FIG. 3. Hydrodynamic simulation of a laser driven shock with the PETE code. The position of the propagating shock can be seen, where the shock velocities corresponding to the break-out times at the three steps on the rear side of the target are shown. The preheat due to nonlocal electrons can be observed and the corresponding thermodynamic conditions are in excellent agreement with the elevated temperature measured by XRTS.



FIG. 4. The shock propagating in the foam layer can be seen in the hydrodynamic simulation profiles of density ρ , electron temperature T_e , and pressure p corresponding to the time of the XRTS measurement in Fig. 3. The preheat zone is highlighted in the inset further accompanied by the profile of electron energy flux density q_H . Since the Knudsen number Kn within the highlighted zone reaches a high value 0.1 (diffusive limit 0.001), the nonlocal electron transport becomes significant $[q_H \sim u(E + p)]$, thus forming the preheat.

temperature length scale. If we compare the jump along the shock of the electron energy flux density q_h to the hydrodynamic energy flux density u(E + p), where u is the fluid velocity, E the total energy density of the plasma, and p the total pressure, we find that $\Delta q_h = -0.16\Delta u(E+p)$, which confirms the importance of the nonlocal transport in the shock dynamics. It also qualitatively describes that we are in an off-Hugoniot regime. We have performed the Hugoniot jump condition analysis and the simulated shock velocity is in excellent agreement in every moment of the shock propagation. Corresponding plasma quantities are pictured in Fig. 4, where the inset corresponds to the XRTS region highlighted in Fig. 3 and the simulated thermodynamic conditions are again in a very good agreement with experimental data shown in Fig. 2 and Table I. The highly localized increase in q_H along the shock clarifies that the nonlocal electrons originate from a thin postshock layer and are thermalized within the finite preheat region. The effect of a consequent heating gives the absorbed flux 2.7×10^6 J/cm³, which surpasses the calculated heating effect of x rays by several orders of magnitude.

In conclusion, this experiment demonstrated the first measurement of the nonlocal transport of electrons through WDM causing significant preheat in the target. Direct measurements of the temperature and shock velocity by independent diagnostics including XRTS, VISAR, and SOP were matched with the results from PETE simulations that include the first-principles based BGK model providing the nonlocal transport of free electrons. According to these simulations, the nonlocal electron transport allows additional transport of energy apart from the hydrodynamic shock resulting in increased temperature, pressure, and consequently, higher shock velocity. Additional simulations including the FLYCHK and CASSIO codes confirmed that the x-ray contribution to this preheat is negligible compared to the heating effect due to nonlocal electrons.

These findings enable the benchmarking of electron conduction models in conditions relevant to ICF, such as those employed in the modeling of experiments performed at the National Ignition Facility (NIF), and convection phenomena in white dwarfs [45]. Additional information about the experiment and data analysis is included in the Supplemental Material [46].

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- S. Lebedev, *High Energy Density Laboratory Astrophysics* (Springer, New York, 2007).
- [2] R. L. McCrory et al., Nucl. Fusion 45, S283 (2005).
- [3] T. Guillot, Science **286**, 72 (1999).
- [4] O. Hurricane et al., Nature (London) 506, 343 (2014).
- [5] P. Olson, Science 342, 431 (2013).
- [6] S. Humphries Jr., Nucl. Fusion 20, 1549 (1980).
- [7] M. Tabak, J. Hammer, M. E. Glinsky, W. L. Kruer, S. C. Wilks, J. Woodworth, E. M. Campbell, M. D. Perry, and R. J. Mason, Phys. Plasmas 1, 1626 (1994).
- [8] C. A. DiStefano, E. C. Merritt, F. W. Doss, K. A. Flippo, A. M. Rasmus, and D. W. Schmidt, Phys. Rev. E 95, 023202 (2017).
- [9] R. S. McWilliams, D. K. Spaulding, J. H. Eggert, P. M. Celliers, D. G. Hicks, R. F. Smith, G. W. Collins, and R. Jeanloz, Science 338, 1330 (2012).
- [10] V. N. Goncharov et al., Phys. Plasmas 13, 012702 (2006).
- [11] K. P. Driver and B. Militzer, Phys. Rev. Lett. 108, 115502 (2012).
- [12] D. Saumon, C. E. Starrett, J. Kress, and J. Clerouin, High Energy Density Phys. 8, 150 (2012).
- [13] V. Recoules, F. Lambert, A. Decoster, B. Canaud, and J. Clerouin, Phys. Rev. Lett. **102**, 075002 (2009).
- [14] G. Faussurier and C. Blancard, Phys. Rev. E 91, 013105 (2015).
- [15] C. K. Li and R. D. Petrasso, Phys. Rev. Lett. 70, 3059 (1993); 114, 199901(E) (2015).
- [16] P. E. Grabowski, M. P. Surh, D. F. Richards, F. R. Graziani, and M. S. Murillo, Phys. Rev. Lett. **111**, 215002 (2013).
- [17] J. F. Luciani, P. Mora, and J. Virmont, Phys. Rev. Lett. 51, 1664 (1983).
- [18] W. Manheimer, D. Colombant, and V. Goncharov, Phys. Plasmas 15, 083103 (2008).

- [19] K. Falk, E. J. Gamboa, G. Kagan, D. S. Montgomery, B. Srinivasan, P. Tzeferacos, and J. F. Benage, Phys. Rev. Lett. 112, 155003 (2014).
- [20] J. Pasley et al., Phys. Plasmas 14, 120701 (2007).
- [21] E. Falize, C. Michaut, and S. Bouquet, Astrophys. J. 730, 96 (2011).
- [22] P. A. Rosen, J. M. Foster, B. H. Wilde, P. Hartigan, B. E. Blue, J. F. Hansen, C. Sorce, R. J. R. Williams, R. Coker, and A. Frank*et al.*, Astrophys. Space Sci. **322**, 101 (2009).
- [23] S. H. Glenzer and R. Redmer, Rev. Mod. Phys. 81, 1625 (2009).
- [24] K. Falk, L. A. Collins, E. J. Gamboa, G. Kagan, J. D. Kress, D. S. Montgomery, B. Srinivasan, P. Tzeferacos, and J. F. Benage, Phys. Plasmas 21, 056309 (2014).
- [25] T. R. Boehly et al., Opt. Commun. 133, 495 (1997).
- [26] S. P. Regan et al., J. Opt. Soc. Am. B 22, 998 (2005).
- [27] N. E. Lanier, C. Hamilton, and J. M. Taccetti, Rev. Sci. Instrum. 83, 10E521 (2012).
- [28] P. M. Celliers, D. K. Bradley, G. W. Collins, D. G. Hicks, T. R. Boehly, and W. J. Armstrong, Rev. Sci. Instrum. 75, 4916 (2004).
- [29] M. C. Gregor *et al.*, Rev. Sci. Instrum. 87, 114903 (2016).
- [30] J. Workman and G. A. Kyrala, Proc. SPIE Int. Soc. Opt. Eng. 4504, 168 (2001).
- [31] E. J. Gamboa, C. M. Huntington, M. R. Trantham, P. A. Keiter, R. P. Drake, D. S. Montgomery, J. F. Benage, and S. A. Letzring, Rev. Sci. Instrum. 83, 10E108 (2012).
- [32] G. Gregori, A. Ravasio, A. Höll, S. H. Glenzer, and S. J. Rose, High Energy Density Phys. 3, 99 (2007).

- [33] G. Gregori and D. O. Gericke, Phys. Plasmas 16, 056306 (2009).
- [34] D. O. Gericke, J. Vorberger, K. Wünsch, and G. Gregori, Phys. Rev. E 81, 065401(R) (2010).
- [35] S. P. Lyon and J. D. Johnson, Los Alamos National Laboratory Report No. LA-UR-92-3407, 1992.
- [36] S. X. Hu, L. A. Collins, V. N. Goncharov, J. D. Kress, R. L. McCrory, and S. Skupsky, Phys. Rev. E 92, 043104 (2015).
- [37] M. Gittings et al., Comput. Sci. Discovery 1, 015005 (2008).
- [38] T. J. Urbatsch and T. M. Evans, Los Alamos National Laboratory Report No. LA-14195-MS, 2005.
- [39] J. Colgan, D. P. Kilcrease, N. H. Magee, M. E. Sherrill, J. Abdallah Jr., P. Hakel, C. J. Fontes, J. A. Guzik, and K. A. Mussack, Astrophys. J. 817, 116 (2016).
- [40] H.-K. Chung, M. H. Chen, W. L. Morgan, Y. Ralchenko, and R. W. Lee, High Energy Density Phys. 1, 3 (2005).
- [41] B. L. Henke, E. M. Gullikson, and J. C. Davis, At. Data Nucl. Data Tables 54, 181 (1993).
- [42] M. Holec, J. Limpouch, R. Liska, S. Weber, Int. J. Numer. Meth. Fluids 83, 779 (2016).
- [43] P. Bhatnagar, E. Gross, and M. Krook, Phys. Rev. 94, 511 (1954).
- [44] J. L. Spitzer and R. Harm, Phys. Rev. 89, 977 (1953).
- [45] G. Fontaine, P. Brassard, and P. Bergeron, Publ. Astron. Soc. Pac. 113, 409 (2001).
- [46] See Supplemental Material at http://link.aps.org/ supplemental/10.1103/PhysRevLett.120.025002 for additional information about the experiment, setup and history. More detailed description of the data analysis and hydrodynamic simulations is provided.