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Inelastic X-ray Scattering from Shocked Liquid Deuterium

S. P. Regan,[1] K. Falk,[2] G. Gregori,[2] P. B. Radha,[1] S. X. Hu,[1] T. R. Boehly,[1] B. Crowley,[1,3] S. H. Glenzer,[4] O. L. Landen,[4] D. O. Gericke,[5] T. Doeppner,[4] C. D. Murphy,[2] T. C. Sangster,[1] J. Vorberger,[5] D. D. Meyerhofer[1*]

[1]*Laboratory for Laser Energetics, University of Rochester, 250 East River Road, Rochester, NY 14623-1299, U.S.A.*
[2]*Department of Physics, University of Oxford, Oxford OX1 3PU, United Kingdom*
[3]*AWE plc., Reading RG7 4PR, United Kingdom*
[4]*Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94551, U.S.A.*
[5]*Centre for Fusion, Space and Astrophysics, Department of Physics, Warwick University, Coventry CV4 7AL, United Kingdom*

[*]*also, Depts. of Mechanical Engineering and Physics and Astronomy University of Rochester*

ABSTRACT

The near-Fermi-degenerate plasma conditions created in liquid deuterium by a laser-ablation driven shock wave were probed with non-collective, spectrally-resolved, inelastic x-ray Thomson scattering (XRTS) employing Cl Ly_α line emission at 2.96 keV. The first XRTS measurements of the microscopic properties of the shocked deuterium, show an inferred mean electron temperature and density of 3±2 eV and $0.4\pm0.2\text{x}10^{23}$ cm^{-3}, respectively. Two-dimensional hydrodynamic simulations using equation of state models suited for the extreme parameters also occurring in inertial confinement fusion research and planetary interiors show that the weighted average of the predicted state of the probed plasma is consistent with the x-ray scattering measurements.

PACS:

Extreme states of matter existing in astrophysical objects (e.g., stars and planetary interiors) can be created in the laboratory with high-intensity laser beams, particle beams, and Z-pinch generators.[1] High-energy-density physics (HEDP) encompasses the research of matter having energy densities of ~10^{11} J/m^3, or more, or equivalently, pressures greater than 1 Mbar.[1,2] A subset of this field involves the study of warm dense matter (WDM),[1,2] with electron temperatures around the Fermi temperature and the ratio of the potential energy to the kinetic energy of the ions greater than unity. The latter can be quantified by an ion–ion coupling parameter[2] that holds $\Gamma_{ii} \equiv (Ze)^2/d_i k_B T > 1$, where Ze is the electric charge of the ion, d_i is the mean ion spacing, k_B is the Boltzmann constant, and T is the temperature. In shock-compressed matter at these extreme conditions, the determination of the system properties, in particular the equation of state (EOS), is complicated by the highly correlated nature of the medium, consisting of a system of strongly coupled ions immersed in a fluid of partially degenerate electrons. Understanding the physical properties (e.g., opacity[3], conductivity[4], EOS[5], and compressibility[6]) of WDM is however very important for inertial confinement fusion (ICF) research[7,8] and the study of planetary interiors[9], because theoretical models differ by factors of several when predicting these quantities. In the past decade, developments in laser produced plasma sources and detector efficiencies has enabled inelastic x-ray scattering to become a powerful diagnostic providing electron temperature (T_e), electron density (n_e) and ionization (Z) for critical EOS measurements in ICF and planetary science research.[10-14] On the other hand, x-ray scattering measurements from warm dense hydrogen (and its isotopes) have still remained elusive due to the very small cross section, and the requirement of collecting the data over a few nanoseconds only.

This Letter describes the first experimental observation of non-collective, inelastic x-ray Thomson scattering from liquid deuterium driven by laser-produced shock wave near 10 Mbar. The average electron temperature, electron density, and ionization are inferred from spectral intensity of the elastic (Rayleigh) and inelastic (Compton) components of the scattered Cl Ly_{α} emission at 2.96 keV. Two-dimensional hydrodynamic simulations using equation of state models designed for the extreme conditions found in ICF research and planetary interiors predicted an average state of the plasma that is consistent with the x-ray scattering measurements. This experiment provides a platform for the detailed study of compressed deuterium, and it is an important step toward measuring all the thermodynamic variables needed for EOS research, that is pressure (p), mass density (ρ), electron density (n_e), electron temperature (T_e), and ionization (Z), by combining inelastic x-ray scattering with shock velocity and optical pyrometry measurements.[15-18]

The experiment to study the spectrally-resolved inelastic x-ray scattering from shocked deuterium was developed on the 60-beam, 30-kJ, 351-nm OMEGA Laser System[19]. Inelastic x-ray scattering is predominatly collective or non-collective depending on the scattering parameter $\alpha_s = \frac{1}{k\lambda_s}$, where the wavenumber of the scattered x-ray is given by $k = 4\pi/\lambda_0 \sin(\theta/2)$ with the incident wavelength λ_0 = 4.188 Å and λ_S is electron screening length of the plasmas. For the partially ionized conditions in WDM, the screening length may be calculated from the Fermi distribution via a single integral[20]. However, an easy fourth order interpolation between the classical Debye length and the Thomas-Fermi screening length valid for T=0 yields also the correct results within 2 percent[20]. If α_S < 1, the scattering is dominated by independent electrons and is referred to as non-collective.[21] In this case, the free electron contribution experiences a significant Compton shift $\Delta E_C = \hbar^2 k^2 / 2m_e$ and is also Doppler-broadened. The width of this scattering feature indicates is sensitive to the electron temperature for nondegenerate plasmas. If α_S >> 1, the scattering by the collective modes, which are known as plasma waves or plasmons, is dominant and the scattering is referred to as collective.[2,10] To lowest order, the position of the energy-downshifted plasmon feature is related to the electron plasma frequency $\omega_{pe} = \left(e^2 n_e / 4\pi\varepsilon_0 m_e\right)^{1/2}$, providing an electron density diagnostic. The Compton-downshift for this experiment is 16.5 eV and the plasma conditions and scattering geometry results in a scattering parameter of α_S ~ 0.5 to 0.6. As the electrons are partially degenerate, this implies that this inelastic scattering geometry is sensitive to both electron density and temperature, which is a novel regime for inelastic x-ray scattering.[10] Additional information on the plasma temperature is given by the height of the elastic scattering feature[22].

The experimental setup is shown in Fig. 1(a). The 8-μm-thick plastic ablator containing the planar layer of liquid deuterium is irradiated with a constant intensity UV laser drive with 10^{14} W/cm^2. The laser drive, formed with six pairs of beams staggered in time as shown in Fig. 1(b), is uniform over a 0.5-mm diameter. A laser-ablation driven shock wave is launched through the liquid deuterium creating warm dense compressed matter. Sixteen tightly focused beams irradiate a parylene D backlighter with 10^{16} W/cm^2 generating a plasma source of Cl Ly$_a$ emission (λ_0 = 4.188 Å, hν = 2960 eV).[23] These x rays are then scattered at θ =87.8º and detected with an x-ray framing camera

(XRFC) outfitted with a HOPG crystal spectrometer.[24] The backlighter x rays are collimated with a 200 μm diameter pinhole. The timing of the backlighter beams is shown in Fig. 1(b). The integration time of the x-ray scattering measurements is ~200 ps. A photograph of the cryogenic target with XRTS capabilities mounted on the OMEGA planar cryogenic system are shown in Fig. 1(c), with the main components highlighted. The fill tube directs deuterium gas into the cryogenic cell, where it condenses into liquid. The ruby tooling balls on the top and right side of the Cu cold finger structure are target alignment fiducials. The Au/Fe shield blocks a direct line of sight between the laser produced plasmas and the detector, which is positioned ~90^0 to the laser drive axis.

Two-dimensional hydrodynamics simulations of the experiment were performed with the DRACO code, which uses the SESAME EOS, a 3D laser ray trace model to calculate the laser absorption via inverse bremsstrahlung, a flux-limited thermal transport approximation with a flux limiter of 0.06, and a multi-group diffusion radiation transport approximation using opacity tables created for astrophysics.[25] The simulation results shown in Fig. 2 predict at peak compression a mass density of ρ ~ 0.8 g/cm^3, a temperature of T_e ~ 5 - 15 eV, and an ionization stage of Z ~ 0.5 – 0.8 for the shocked liquid deuterium. Since the measured spectrum of the scattered x rays is spatially integrated, it includes contributions from both the shocked and unshocked liquid deuterium. An equivalent spatial integration was applied to the 2D simulation results to calculate the average predicted plasma conditions as seen by the scattering measurement. Given that the total number of scattered photons is proportional to the number of particles in the scattering volume, the average thermal velocity is obtained by

$$\bar{v}_{th} = \frac{\int \sqrt{k_B T_e(r,z)/m_e}\,\rho(r,z) r\, dr\, dz}{\int \rho(r,z) r\, dr\, dz}$$

which can be related to the observed Doppler broadening of the spectrum ($\Delta E / E \sim k\bar{v}_{th}$). From the DRACO simulations, we then obtain an electron temperature of T_e=3 eV. The mass averaged quantities of n_e, and Z from DRACO are 0.5 x 10^{23}cm^{-3} and 0.3, respectively.

The scattered spectrum of the Cl Ly$_\alpha$ emission is shown in Fig. 3(a) and incident spectrum is shown in Fig. 3(b). The incident spectrum is measured by irradiating a parylene D foil target on a separate laser shot. The scattered spectrum has a strong Rayleigh peak around 2960 eV and a Compton-downshifted feature. The splitting of the Cl Ly$_\alpha$ emission is observed in the scattered spectrum, but not in the incident spectrum, due to differences in the amount of source broadening in the different settings. In particular, the backlighter plasma is considerably larger than the scattering volume. Scattered x-ray spectra were calculated using the x-ray scattering code (XRS) code, which uses the finite temperature random phase approximation with static local field corrections to obtain the spectral shape of the inelastic (Compton) feature due to scattering at free electrons[26]. The elastic scattering intensity strongly depends on the degree of ion-ion correlations in the plasma via the structure factor S_{ii}.[22] To constrain the value for S_{ii}, density functional theory molecular dynamics (DFT-MD) simulations were performed using the VASP package.[27,28] The simulations indicate weak ionic correlations for the conditions similar to the average of the plasma probed. This means the ion-ion structure factor S_{ii} at the relevant scattering wavenumber is close to unity for most conditions probed. With this information, the elastic scattering feature can be used to restrain the temperature and the ionization degree of the system. Structure factors close to unity are also found for the unshocked deuterium liquid. In addition to Doppler broadening, the width and position of the inelastic feature are also dependent on the density for $\alpha_s \sim 1$. This fact allows us to bracket the electron density and estimate the ionization charge based on the initial mass density of the sample. The simulated scattering spectra computed using XRS provided the best fit to the measured spectrum for the following plasma conditions: T_e = 3±2 eV, Z ~ 0.35±0.15 and n_e = 0.4±0.2 x 10^{23}cm^{-3}. These values are in agreement with DRACO simulations (see Fig. 3(b), where the calculated spectra bracket the measurement). Additional experiments are ongoing to resolve spatially shocked and unshocked portions of the target.

In conclusion, this Letter reports the first experimental observation of non-collective, inelastic x-ray scattering from shocked liquid deuterium. An electron temperature of T_e = 3±2 eV, an electron density of n_e = 0.4±0.2 x 10^{23}cm^{-3} and an average ionization degree of Z ~ 0.35±0.15 are inferred from the shapes and intensities of the elastic (Rayleigh) and inelastic (Compton) components in the scattering spectra. These plasma conditions are nearly Fermi-degenerate with similar electron and Fermi temperatures ($T_e/T_F \sim 1$). Two-dimensional hydrodynamic simulations utilizing equation of state models suited for the extreme conditions obtained indicate that the predicted average state of the probed plasma is consistent with the x ray scattering measurements. This experimental result is a significant step toward achieving accurate measurements of all thermodynamic variables needed to provide stringent tests of equation of state models, which would require at least three thermodynamic variables like pressure, mass density, and temperature. However, additional information on the electron density or the ionization degree in WDM states is also highly valuable for comparison with theoretical predictions and can be obtained by combining inelastic x-ray scattering observations with shock velocity measurements.

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FIGURE CAPTIONS

Figure 1. (color) (a) Schematic of the x-ray Thomson scattering experiment. An 8-μm CH ablator is irradiated with a constant-intensity, 6-ns UV laser drive, launching a shock wave through a cryogenic cell filled with liquid deuterium and creating warm dense matter. Sixteen tightly focused beams irradiate a parylene D backlighter at 10^{16} W/cm^2, producing Cl Ly$_\alpha$ emission this is scattered at ~90° and detected with an x-ray framing camera outfitted with a HOPG (highly oriented pyrolytic graphite) crystal spectrometer. (b) Timing of the drive and backlighter beams and the x-ray scattering measurements. (c) Photograph of the x-ray. The fill tube directs deuterium gas into the cryogenic cell, where it condenses into liquid. The ruby tooling balls on the top and right side of the Cu cold finger are target alignment fiducials. The Au/Fe shield blocks a direct line of sight between the laser produced plasmas and the detector, which is positioned ~90^0 to the laser drive axis.

Figure 2. (color) Contour plots of (a) mass density, (b) electron temperature, and (c) average ionization of shocked liquid deuterium at 5 ns, predicted using *DRACO.*

Figure 3. (color) Measurement of (a) Cl Ly$_\alpha$ emission incident on shocked liquid deuterium and (b) Cl Ly$_\alpha$ emission scattered from shocked liquid deuterium. The scattered spectrum has a strong Rayleigh peak around 2960 eV and a Compton-downshifted feature. The splitting of the Cl Ly$_\alpha$ emission is observed in the scattered spectrum, but not in the incident spectrum, due to differences in the amount of source broadening in each measurement. The scattered spectrum was fitted with the X-ray

scattering code (XRS) code and T_e = 3±2 eV, Z ~ 0.35±0.15 and n_e = 0.4±0.2 x 10^{23}cm^{-3} were inferred.

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Fig. 1

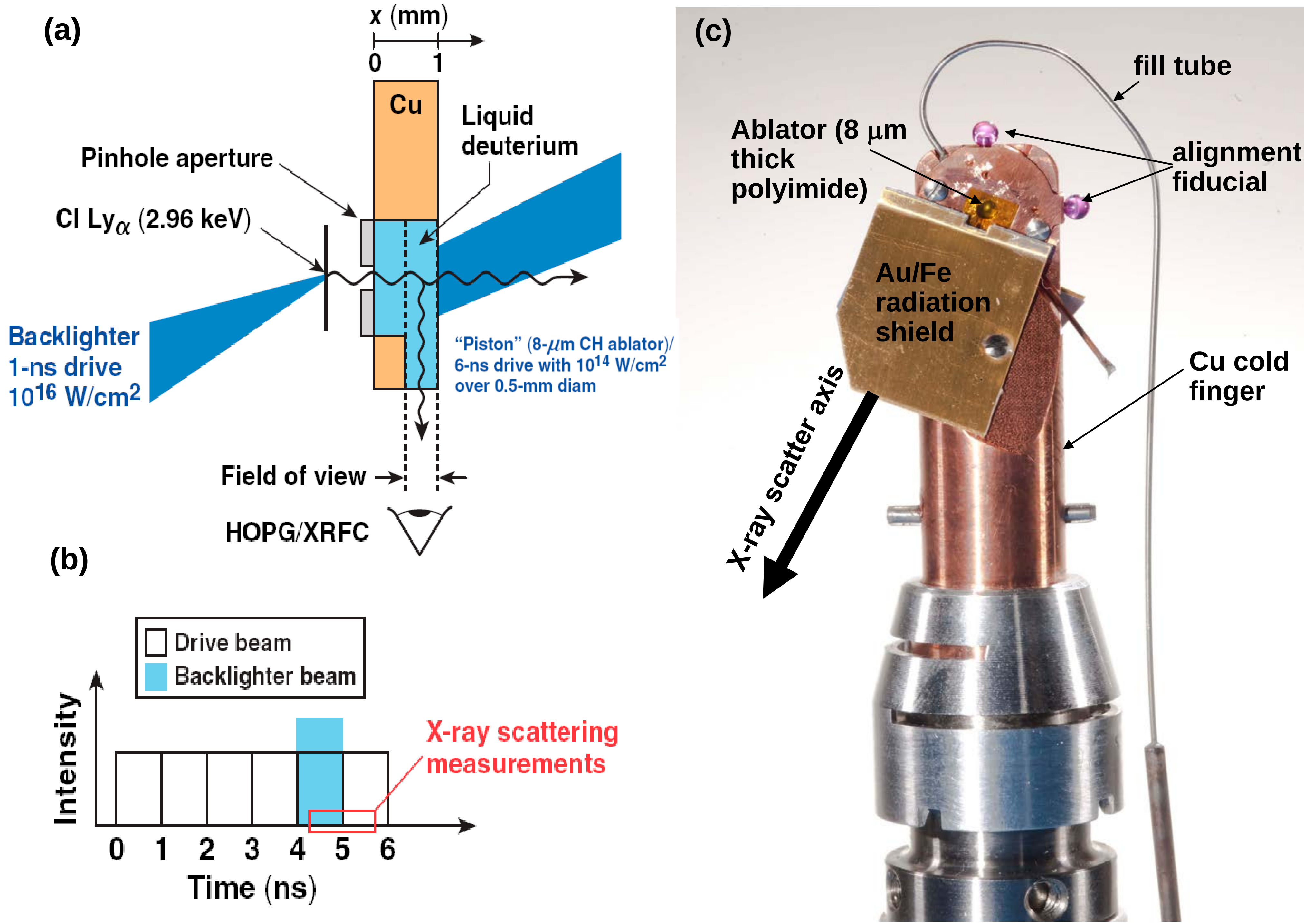
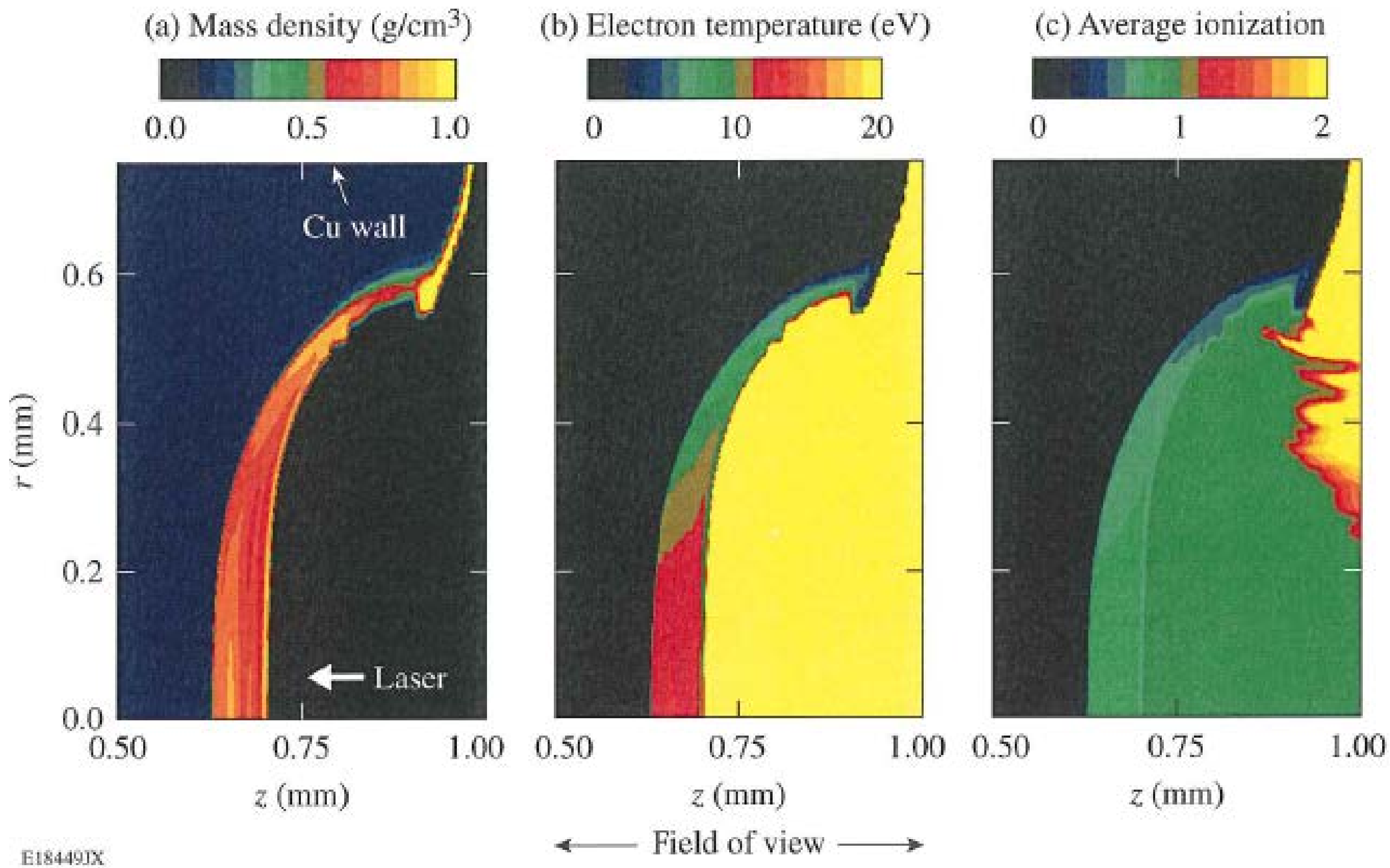

(a)
x (mm)
0
1
Cu
Liquid deuterium
Pinhole aperture
Cl Lyα (2.96 keV)
Backlighter
1-ns drive
10^16 W/cm^2
"Piston" (8-μm CH ablator)/
6-ns drive with 10^14 W/cm^2
over 0.5-mm diam
Field of view
HOPG/XRFC
(b)
Drive beam
Backlighter beam
Intensity
X-ray scattering measurements
0 1 2 3 4 5 6
Time (ns)
(c)
fill tube
Ablator (8 μm thick polyimide)
alignment fiducial
Au/Fe radiation shield
Cu cold finger
X-ray scatter axis

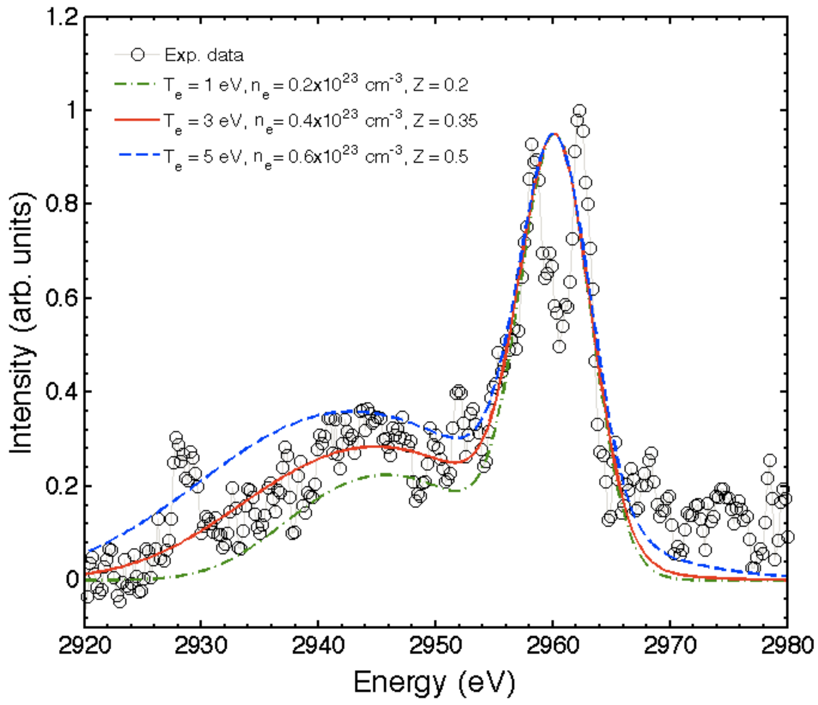
Fig. 2



(a) Mass density (g/cm3)
0.0
0.5
1.0
(b) Electron temperature (eV)
0
10
20
(c) Average ionization
0
1
2
Cu wall
Laser
r (mm)
0.0
0.2
0.4
0.6
0.50
0.75
1.00
z (mm)
Field of view
E18449JX

Fig. 3

(a) Scattered spectrum



(b) Incident spectrum