Observation of Ultrafast Nonequilibrium Collective Dynamics in Warm Dense Hydrogen

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(Received 29 October 2009; published 25 March 2010)

We investigate ultrafast (fs) electron dynamics in a liquid hydrogen sample, isochorically and volumetrically heated to a moderately coupled plasma state. Thomson scattering measurements using 91.8 eV photons from the free-electron laser in Hamburg (FLASH at DESY) show that the hydrogen plasma has been driven to a nonthermal state with an electron temperature of 13 eV and an ion temperature below 0.1 eV, while the free-electron density is $2.8 \times 10^{20}$ cm$^{-3}$. For dense plasmas, our experimental data strongly support a nonequilibrium kinetics model that uses impact ionization cross sections based on classical free-electron collisions.

The frequency shift $\gamma$ of the plasmons with respect to incident radiation is described by the dispersion relation $\omega^2 = \omega_p^2 + 3T_e k^2/m_e$, their intensity ratio by the detailed balance relation $S_{\text{blue}}/S_{\text{red}} = e^{-\hbar \omega_0/T_e}$, where $\omega_p = \sqrt{n_e e^2/m_e \varepsilon_0}$ is the plasma frequency, $k$ the photon wave vector change, $m_e$, $n_e$, $T_e$ electron rest mass, density and temperature (in eV), $e$ the electron charge, and $\varepsilon_0$ the vacuum permittivity. Therefore, from the measured asym-
metry and frequency position of the plasmons we can determine the electron temperature via detailed balance and density via the above dispersion relation.

Our experiments have been performed at DESY using the free-electron laser in Hamburg (FLASH) [16,17] at a photon energy of 91.8 eV. FEL radiation with 5 Hz pulse repetition rate, average pulse energy on target of 15 μJ and duration of ~40 fs is focused to a 25 μm spot using an elliptical mirror, yielding intensities of ~8 × 10^{13} W cm^{-2}. The energy of each pulse is measured using a residual gas ionization detector [18] and fluctuates less than 10%. Figure 1 shows a schematic of the experiment. The FEL pulses hit the liquid hydrogen jet which has a diameter of 20 μm, an atomic density of 4.2 × 10^{22} cm^{-3}, and a temperature of 2 meV, prepared in a liquid helium cooled cryostat [19]. With 60 ms^{-1} flow velocity of the hydrogen jet each pulse scatters from an unperturbed region of the sample. During hydrogen injection the chamber was at a pressure of ~10^{-5} mbar. Scattering from the hydrogen jet is collected at 90° relative to the incident FEL radiation and in the vertical plane since the FEL radiation is horizontally polarized. We use a variable line space grating spectrograph [20] with a resolution of λ/Δλ = 180. A toroidal mirror 25.5 cm from the jet provides a collection solid angle of 1.9 × 10^{-3} sr. Reference measurements of the FEL spectrum (Fig. 1) were performed using a second spectrograph, introduced into the FEL beam during tuning and setup, or a third spectrograph [21] which simultaneously records radiation passing by the liquid jet target.

For these experiments we have chosen 91.8 eV soft-x-ray radiation to be in a regime where the photon energy is well above the plasma frequency for liquid density hydrogen (ℏω_{p} = 7.6 eV) and to match the large penetration depth of 9.4 μm [22] to the target radius (10 μm). While at this photon energy attenuation via photoabsorption has the highest cross section, a small fraction of the incident radiation is Thomson scattered. The ratio between these two cross sections is 3.8 × 10^{4} at 91.8 eV [22]. Thus, the FEL pulses deposit energy in the liquid hydrogen while simultaneously probing the system. Because of the ultra-short pulse duration and the high penetration depth, the liquid hydrogen is heated isochorically and volumetrically by the pulse [23].

A scattering spectrum for 15 min integration time (4500 pulses) is shown in Fig. 2. It is composed of asymmetric peaks (plasmons) equally blue and red shifted from the incident photon energy by 0.65 eV. The measured spectrum is mainly broadened by the incident FEL bandwidth (1.1 eV full width at half maximum, mainly Gaussian, Fig. 1). Further source broadening effects (0.2 eV corresponding to the 20 μm source diameter) are minimal due to the geometry of the setup and the FEL pointing stability.

The total spectrum is described by the dynamic structure factor S(ω) [24,25]:

\[ S(ω) = Z_f S_e(ω) + Z^2(1 - e^{-2W}) S_i(ω). \]  

(1)

Here, Z is the nuclear charge and Z_f the average number of free electrons per atom. The first term in Eq. (1) describes the high frequency free-electron fluctuations which includes collective plasmon scattering. The second term describes the ionic, nearly elastic response with the ion-ion structure factor S_i modulated by the Debye-Waller factor \(1 - e^{-2W} \approx 2W/T_i^2/T_D^2\) [26,27], which accounts for low frequency ion acoustic fluctuations coupling with ionic thermal vibrations, where \(T_i\) is the ionic and \(T_D\) the Debye temperature. In this experiment \(T_i\) is sufficiently small against \(T_D (T_i \sim 2 \text{ meV}, \ T_D \sim 10 \text{ meV estimated using the Bohm-Staver relation [28]) so that elastic scattering is strongly suppressed. This effect was previously observed in laser based nonequilibrium experiments [29].

Collective TS spectra are calculated in Born-Mermin approximation [30] and convolved with the experimental

![FIG. 1 (color online). Experimental setup showing the liquid hydrogen jet in the focus of the FEL beam with typical time averaged spectrum, and the spectrograph at 90° scattering angle with the lineout region of the CCD, corresponding to the focussed part of the source image, marked by white bars.](image191x231 to 264x265)

![FIG. 2 (color online). Experimental spectrum (red circles) and the best fit of a calculated spectrum with \(n_e = 2.8 \times 10^{20} \text{ cm}^{-3}\) and \(T_e = 13 \text{ eV}\) (solid blue line). Comparison to fits with variation in density (left graph), \(5.6 \times 10^{20} \text{ cm}^{-3}\) (dashed green line) and \(1.4 \times 10^{20} \text{ cm}^{-3}\) (dash-dotted brown line), and in temperature (right graph), 20 eV (dash-dotted brown line) and 5 eV (dashed green line), are shown.](image216x167 to 237x228)
resolution. A synthesized spectrum fits the measurement best using an electron temperature and density of 13 eV and $2.8 \times 10^{20}$ cm$^{-3}$, respectively (Fig. 2). Spectra obtained by varying these parameters by a factor of 2 deviate considerably from the observed spectra. Sensitivity analysis shows that the error in the fitting procedure is 25% in electron temperature and density, respectively.

Furthermore, elastic scattering is practically absent in the experimental spectra and comparing with synthesized spectra we obtain an upper limit for $T_i < 0.1$ eV, where $S_{ij}$ has been estimated using the screened one component plasma model [31]. Consequently, the liquid hydrogen is excited to a nonequilibrium plasma state with different electron and ion temperatures at a very low degree of ionization ($\sim 0.7\%$) but high atomic density ($4.2 \times 10^{22}$ cm$^{-3}$).

The degree of ionization remains low despite the high electron temperature, due to the short time scale of the interaction. The electron-atom, electron-ion, and electron-electron energy transfer time scales for 13 eV electrons are $>13$ ps, $\sim 16$ ps, and $\sim 25$ fs, respectively [32]. Impact ionization takes 0.4–1 fs [33,34], being the fastest process for electrons between 20 eV and 400 eV. This suggests that the electronic subsystem is thermalized within the duration of the FEL pulse [Fig. 3(b)], but no equilibration has been reached between the other components explaining the combination of low degree of ionization and high electron temperature.

We have simulated the evolution of the electron kinetic energy distribution during and after the FEL irradiation (Fig. 3), using a model [35–37] based on kinetic equations. Because of the almost uniform target conditions (target radius matches penetration depth) [23] we can simulate a smaller target, a cluster of only $\sim 10^5$ hydrogen atoms. The simulation includes the following predominant interactions: photo- and collisional ionization, three-body recombination, elastic electron-ion and electron-atom scattering, and screened electron-electron interactions. Our model follows the full dynamics of an irradiated sample, from the nonequilibrium up to the thermal equilibrium phase for electrons. The electron temperature from the simulation $T_e' = \frac{3}{2} \langle E_{kin}' \rangle$ is 12 eV, averaged over the FEL pulse duration, and the degree of ionization is 0.5%, where $\langle E_{kin}' \rangle$ is the instantaneous ensemble average of the free-electron kinetic energy. These results compare well to the TS measurement. Furthermore, the electron energy distribution approaches a Maxwell-Boltzmann (MB) distribution at 0 fs before (dashed blue line), and 20 fs after (dashed red line) the FEL pulse peak. A Maxwell-Boltzmann distribution fit at 0 fs (solid black line) and the photoelectron (78.2 eV) and ionization (13.6 eV) energies are shown for comparison.

Simulating the target interaction we can study the influence of different cross sections for impact ionization in dense plasmas by comparing with our measurement. We used the National Institute of Standards and Technology (NIST) database for molecular hydrogen [34] as well as an expression from Lieberman and Lichtenberg [33]. The latter approximates the ionization via the low angle scattering of two free electrons and was employed in Fig. 3. NIST uses the binary encounter Bethe (BEB) model taking atomic and molecular structure into account. The models deviate up to a factor of 4 in the relevant electron energy range. Simulations with the classical model [33] yield an electron density and temperature at the FEL peak (0 fs) which matches our measurement significantly better than BEB (Fig. 4). Even assuming a 4 times higher FEL incident power cannot explain the observed values. A possible interpretation is that the atomic structure (as treated in BEB) does not play a significant role in the context of dense plasmas where, due to screening and correlation effects,
FIG. 4 (color online). Measurements (colored circles) compared to the simulated evolution of the target’s free-electron density and temperature using different impact ionization cross sections and FEL pulse energies: Lieberman and Lichtenberg (solid red line, 15 µJ) and the BEB model (15 µJ dashed blue line, 60 µJ dotted green line). The FEL full width half maximum duration is indicated in gray scale.

high lying atomic states are removed and the electron interaction is more properly described with a classical ionic background.

In conclusion, we were able to obtain Thomson scattering spectra with an unprecedented signal to noise ratio due to the high repetition rate of the FEL and compared to single laser pulse driven x-ray sources [8–10]. The ultra-short high intensity soft-x-ray pulses allow us to heat liquid hydrogen isochorically and volumetrically to the observed moderately coupled nonequilibrium state. Our results compare well to nonequilibrium kinetic simulations using classical impact ionization models which show an electron relaxation time scale of ~20 fs. Already available pump-probe techniques [38] will allow the measurement of time scales for the subsequent relaxation channels via electron-ion equilibration. This is a pioneering step towards the investigation of nonequilibrium and strongly-coupled plasmas with implications from astro- to energy physics [1].

We thankfully acknowledge financial support by the Helmholtz Gemeinschaft via the Virtual Institute VH-VI-104, the German Federal Ministry for Education and Research via Project No. FSP 301-FLASH, and the Deutsche Forschungsgemeinschaft (DFG) via the Sonderforschungsbereich SFB 652. T.L. acknowledges DFG support under Grant No. LA 1431/2-1, R. R. F. under Grant No. GRK 1355. The work of S. H. G. and T. D. was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. DE-AC52-07NA27344 and was supported by LDRDs 08-ERI-002, 08-LW-004. S. H. G. and C. F. were supported by the Alexander von Humboldt foundation, G. G. partially by the Engineering and Physical Sciences Research Council (Grant No. EP/G007187/1) and the Science and Technology Facilities Council of the UK. Finally, the authors are greatly indebted to the FLASH team for enabling an outstanding performance.

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