Two-color Thomson scattering at FLASH

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Abstract

We propose a two-color pump-probe Thomson scattering experiment at the FLASH facility in Hamburg to characterize warm dense matter states. The fundamental free electron laser wavelength of 40.5 nm is used to pump a liquid hydrogen jet that is subsequently probed with the third harmonic at 13.5 nm. We have considered the laser-target interaction in the pump and probe phase by using the radiation hydrodynamics code HELIOS. The calculation of the Thomson scattering spectrum is based on the Chihara formula which is evaluated using the Born-Mermin approximation for the free electron dynamic structure factor and the Debye-Hückel static structure factor for the elastic scattering part. We consider the full density- and temperature-dependent Thomson scattering cross section throughout the inhomogeneous target. The results indicate that the electron-ion equilibration rate can be extracted by measuring the electron and ion feature with varying time delays between the pump and the probe pulse.

1. Introduction

X-ray Thomson scattering is a promising tool for the diagnostics of dense strongly correlated plasmas [1]. X-rays emitted from laser produced plasmas [2, 3] can probe the warm dense matter (WDM) region [4, 5] with temperatures of several eV and densities close to solid density [6, 7, 8] up to compressed matter well above solid density at electron temperatures T_e between 0.1 eV and several 10 eV [9, 10, 11, 12, 13]. The study of dense plasmas is also possible with the implementation of free-electron lasers which provide brilliant radiation in the soft x-ray region at the free-electron LASer Hamburg (FLASH) at DESY, Hamburg [14, 15] or in the x-ray region at the Linac Coherent Light Source (LCLS) in Stanford [16], and will be available in the future at the European XFEL in Hamburg [17].

Collective x-ray Thomson scattering experiments yield information on the density and temperature of dense plasmas [8]. Assuming a homogeneous density and temperature throughout the target, these parameters can be determined directly from the plasmon dispersion and the ratio of the plasmon amplitudes via the detailed balance relation [18]. For higher laser wavelengths, i.e., optical lasers and wavelengths of about 40–50 nm accessible with FLASH, the target is overdense, as the absorption is limited to the skin depth, in contrast to excitation by short-wavelength FEL radiation where gradients are smaller,

see [19, 20]. The scattering signal represents an average of the local density- and temperature- dependent scattering cross-sections weighted with the respective density and temperature profiles [21, 22, 23].

In this paper, we propose a new Thomson scattering experiment at FLASH using a two-color pump-probe scheme on liquid hydrogen based on theoretical results obtained recently for an optical pump – FLASH probe setup [23]. This proposed experiment uses the FLASH fundamental as pump and the third harmonic as probe. We calculate the Thomson scattering spectrum of free electrons via the Born-Mermin approximation (BMA) [18, 24]. Furthermore, we determine the elastic scattering signal of bound electrons, the so-called Rayleigh signal, within the simple Debye-Hückel approximation. We find that a realistic description of the light-matter interaction within the proposed pump-probe experiment is of paramount importance, especially to derive reliable density and temperature profiles throughout the inhomogeneous target. We use the radiation hydrodynamics code HELIOS for the characterization of the target [25]. Our results indicate that the full time-resolved Thomson scattering spectrum can be used to extract electron-ion equilibration rates by varying the time delay between pump and probe pulse.

2. Thomson scattering spectrum

The experimentally available scattering power per solid angle $d\Omega = \sin\vartheta d\vartheta d\varphi$ and per unit frequency interval $d\omega$

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is given by the following expression [21]:

$$\frac{d^2 P_{sc}}{d\Omega d\omega} = \frac{\sigma_T}{A_{rad}} \frac{k_f}{k_i} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} G_{\Delta\omega}(\omega - \omega') \times \int d^3\mathbf{r} l(\mathbf{r}) S_{ee}(\mathbf{k}, \omega'; n_e(\mathbf{r}), T_e(\mathbf{r})) n_i(\mathbf{r}), \quad (1)$$

where $\sigma_T = 6.65 \times 10^{-24} \text{ cm}^2$ is the Thomson scattering cross-section. The energy and momentum transfer are given by $\Delta E = \hbar\omega = \hbar\omega_f - \hbar\omega_i$ and $\hbar\mathbf{k} = \hbar\mathbf{k}_f - \hbar\mathbf{k}_i$, respectively. Here k_i and k_f are the initial and final photon wave numbers, and ω_i and ω_f are the initial and final photon frequencies. The momentum is related to the scattering angle θ in the limit $\hbar\omega \ll \hbar\omega_0$ according to $k = 4\pi \sin(\theta/2)/\lambda_0$, with λ_0 being the probe wavelength. $l(\mathbf{r})$ is the $\mathbf{r}$ -dependent power density of incoming photons which is also dependent on the absorption, $n_i(\mathbf{r})$ is the ion density, $n_e(\mathbf{r})$ is the electron density, and A_{rad} is the irradiated surface of the target. The dynamic structure factor $S_{ee}(k, \omega)$ is convoluted with the instrumental function $G_{\Delta\omega}(\omega)$ that models the spectrometer's finite spectral resolution as well as the probe's spectral bandwidth. Usually, a normalized Gaussian distribution is employed with the full width at half maximum (FWHM) $\Delta\omega$, see also Ref. [22]. The dynamic structure factor can be separate into contributions of free electrons, weakly- and tightly-bound electrons, and core electrons as proposed by Chihara [26, 27]:

$$S_{ee}(k, \omega) = Z_f S_{ee}^0(k, \omega) + |f_i(k) + q(k)|^2 S_{ii}(k, \omega) + Z_c \int_{-\infty}^{\infty} d\omega' S_c(k, \omega) S_s(k, \omega - \omega'). \quad (2)$$

Here, $Z_f = n_e/n_i$ is the ionization degree of the plasma and Z_c is the averaged number of core electrons. The first contribution of free electrons is calculated via the BMA which contains ion-electron collisions in second order Born approximation. For the calculation of the second term, we use the atomic form factor $f_i(k)$ [28] and the Debye-Hückel ion-ion structure factor for point charges

$$S_{ii}(k) = \frac{k^2 + \kappa_e^2}{k^2 + \kappa_i^2 + \kappa_e^2}, \quad (3)$$

with the inverse screening length $\kappa_c = \sqrt{e_c^2 n_c / (\epsilon_0 k_B T_c)}$ for species $c = e$ (electrons) and $c = i$ (ions). In the Debye-Hückel picture, the screening cloud can be given by $q(k) = \sqrt{Z_f} S_{ei}(k) / S_{ii}(k) = Z_f \kappa_e^2 / (k^2 + \kappa_e^2)$. For the last term in Eq. (2), the contribution of bound-free transitions, we use the formalism of Schumacher *et al.* [28, 29]. This part describes the Raman-type transitions of inner shell electrons to the continuum which can be modulated by the ion's movement contained in $S_s(k, \omega)$ [30]. In this paper for Thomson scattering on hydrogen, the last term can be neglected for the relevant energy range of -10 eV to 10 eV .

Figure 1: *Ab initio* simulation of the inverse absorption length $\alpha(\omega)$ for cryogenic hydrogen ($T = 20 \text{ K}$) at a liquid density of $\rho = 0.086 \text{ g/cm}^3$ as function of the photon energy in comparison with Samson *et al.* [36]. The two red arrows indicates the fundamental wavelength of 40.5 nm and the 3rd harmonic at 13.5 nm.

3. Temperature and density profiles with HELIOS

We describe the new experimental setup for Thomson scattering at FLASH briefly. We propose to use a fundamental wavelength of 40.5 nm ($E = 30.6 \text{ eV}$) for the pump pulse that generates an inhomogeneous plasma in the cryogenic hydrogen droplets of 30 μm diameter, which has an initial state $T_i = 20 \text{ K}$, $\rho = 0.086 \text{ g/cm}^3$. The 3rd harmonic with a wavelength of 13.5 nm ($E = 92 \text{ eV}$) is applied as probe pulse which undergoes Thomson scattering processes in the target. The fundamental and the third harmonic have each a pulse duration of 150 fs and a focal spot radius of 20 μm . The pulse energy of the fundamental is 250 μJ and that of the third harmonic is 2.5 μJ .

For a more detailed description of the absorption process, we calculate the inverse absorption length $\alpha(\omega) = 1/l_{abs}(\omega)$ of cryogenic hydrogen via *ab initio* molecular dynamic simulations using the VASP code [31, 32, 33]. By applying the projector-augmented wave method [34], we are able to evaluate the Kubo-Greenwood formula to obtain optical properties [35], which are compared with experimental data from Samson *et al.* [36], see Fig. 1. The absorption length for 40.5 nm (pump) is about thirty times shorter than for 13.5 nm (probe) so that the energy of the pump pulse is deposited in a layer of only few μm thickness in the front of the droplet. This plasma region may yield a clear plasmon scattering signal when probed with the shorter wavelength of 13.5 nm, similar to the results given in Ref. [23].

In Fig. 2 and Fig. 3, we show the results for the free-electron density and the temperature profiles for the middle layer of the hydrogen droplet using the radiation-hydrodynamic simulation code HELIOS [25]. HELIOS features a Lagrangian reference frame, separate ion and electron temperatures, and flux-limited Spitzer thermal conductivity. The laser energy is deposited via inverse bremsstrahlung as well as bound-bound and bound-free transitions using

Figure 2: Free-electron density profiles $n_e(r)$ for different delay times between pump and probe pulse as derived from the HELIOS simulations. The black curve is the result for the plasma conditions 0.5 ps after the pump pulse without irradiation by a probe pulse. Both FLASH laser pulses (pump: 40.5 nm, probe: 13.5 nm, 150 fs pulse length each) come from the left.

Figure 3: Temperature profile of free electrons $T_e(r)$ as derived from the HELIOS simulations, see Fig. 2.

a SESAME-like equation of state [37]. We show results for several delay times between pump and probe beam. The black curve with $t = 0.5$ ps is the result for a target without irradiation by a probe pulse. With increasing delay times a progressing expansion of the plasma occurs and a shock front penetrates into the target. The maximum of the temperature curves is broadened with increasing delay time because of thermal flux.

In Fig. 2 and Fig. 3 a higher absorption is seen due to the higher wavelength of 40.5 nm in comparison with previous results of Fortmann *et al.* [22] where a wavelength of 13.5 nm was considered for self-Thomson scattering. Therefore, the first few μm of the target are well ionized and a subsequent hydrodynamic expansion is caused by the laser heating. In the remaining part of the target, the plasma temperature and density is very low due to the low penetration depth of the pump laser.

Figure 4: Free-electron Thomson scattering spectrum from liquid hydrogen droplets as function of the photon energy shift ΔE . The "ideal" probe (black curve) is compared with the full pump-probe scenario considering delays of 1 ps (dotted red line), 5.0 ps (broken blue line), and 10 ps (broken dotted dark green line).

4. Results for the Thomson scattering spectrum

Using the electron density and temperature profiles through the hydrogen droplet as derived from the HELIOS simulations as input in Eq. (1), we calculate the Thomson scattering spectrum of the probe pulse for several delay times. Non-convoluted Thomson scattering spectra are shown in Fig. 4. Please note that the curves in Fig. 4 show only the contribution of free electrons. The HELIOS one-dimensional free-electron densities and temperatures are divided in 450 equidistant sampling points. For each cell we have calculated the dynamic structure factor $S_{ee}^0(k, \omega) \omega_{pe}^2$ with the electronic plasma frequency $\omega_{pe}^2 = n_e e^2 / (\epsilon_0 m_e)$ for a scattering angle of $\theta_S = 90^\circ$. For a full scattering spectrum, all cells are weighted by the factor $\exp(-r/l_{abs})$, with the absorption length of $l_{abs} = 11 \mu\text{m}$ as derived from Fig. 1 for $\lambda_0 = 13.5$ nm. Therefore, we consider the weakening of the incoming 13.5 nm FLASH probe pulse with increasing target depth.

The high plasma density in the outer shell of the droplets yields a strong plasmon peak in the spectrum at an energy shift of about $\Delta E = 6.25$ eV for a 1 ps delay time. Later, the densities decrease and the plasmon peak is shifted to lower energies. For the maximum delay time of 10 ps considered, the plasmon peaks have already disappeared. The central peak in the spectrum can be related to collective as well as non-collective scattering from the lower density region in the droplet behind the shock front.

An important question for such Thomson scattering experiments is whether or not the plasmon peaks shown in Fig. 4 can be distinguished from the central Rayleigh peak which measures the elastic scattering from bound electrons. Therefore, and in extension of previous theoretical work [23], we have also calculated the respective second term in Eq. (2). For simplicity we apply the Debye-Hückel approximation which is sufficient for our purposes here. Improved treatments, e.g., by performing hypernetted chain calculations (HNC) or *ab initio* molecular dy-

Figure 5: Convolved full Thomson scattering spectrum from liquid hydrogen droplets as function of the photon energy shift ΔE for a delay time of 1 ps and various atomic temperatures T_a . The electron temperatures T_e are obtained from the HELIOS simulations. The Rayleigh peak increases with atomic temperature. For the convolution, a normalized Gaussian distribution with a FWHM of $\Delta\omega = 1$ eV is employed.

namics simulations for the static ion-ion structure factor $S_{ii}(k)$, are straightforward [38, 39, 40].

As an example we show in Fig. 5 the total Thomson scattering spectrum including the ion feature for a delay time of 1 ps and different temperatures of the atoms T_a . The ion feature represents the contribution of bound electrons which in the case of hydrogen refer to atoms. We assume that the atom temperature is equal to the temperature of the ions $T_i = T_a$. For a more detailed treatment of a three-component, partially ionized system with different temperatures T_e , T_i , and T_a , see Ref. [40, 41]. The spectra are convoluted with a FWHM $\Delta\omega = 1$ eV. For a fully ionized plasma, the Rayleigh peak would be zero. In case of partial ionization, especially for equilibrium with $T_a = T_e$, the Rayleigh peak stemming from the weakly- and tightly-bound electrons is a factor of about 30 times higher than the plasmon peak. Furthermore, the calculations show that the Rayleigh peak increases with the temperature of the atoms. Since a non-equilibrium state with $T_a \ll T_e$ is relevant for short times of a few ps after the pump pulse, the equilibration rate between the free electrons and the heavy particles (ions, atoms) can be derived by measuring the heights of the plasmon and Rayleigh peaks simultaneously. This is in principle possible via the proposed two-color pump-probe experiments at the FLASH facility.

5. Conclusion

We have shown with detailed calculations and modeling that electron-ion temperature equilibration in warm dense hydrogen can be measured with two-color Thomson scattering experiments at the FLASH facility. The proposed setup would use a fundamental wavelength of 40.5 nm to pump liquid hydrogen droplets and the third

harmonic at 13.5 nm to probe the inhomogeneous plasma. Radiation- hydrodynamics simulations using the HELIOS code determined the expected electron density and temperature profiles in the droplet. The pump pulse induces strong gradients in the free electron density and temperature. The Thomson scattering spectrum is calculated via the Born-Mermin approximation for the electron feature and the Debye-Hückel structure factor for the ion feature. The high- density plasma in a thin surface layer of the droplet generates a plasmon signal which could be observed for short delay times of less than 5 ps. The Rayleigh signal of tightly-bound electrons is very large compared to the plasmon signal of free electrons for equilibrium conditions, i.e., $T_a = T_e$. For short times of few ps, we expect a non-equilibrium plasma with substantially lower temperatures for the heavy particles (ions, atoms), i.e., $T_a \ll T_e$, and thus a much smaller Rayleigh signal. Therefore, the equilibration rate between the free electron system and the heavy particles could be derived by measuring the heights of the plasmon and Rayleigh peaks simultaneously via the proposed two-color pump-probe experiments at FLASH. The evaluation of the scattering signal would also require more realistic ion-ion structure factors for the ion feature than the simple Debye-Hückel expression used here. The performance of HNC or ab initio MD simulations can be checked via $\mathbf{k}$ -dependent measurements of the Rayleigh signal using different scattering angles. Since multi-angle spectrometers are currently being designed, see e.g., [42], this task seems to be feasible.

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