The interaction of intense ultrashort laser pulses with cryogenic He planar jets

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Abstract

We study the interaction of intense ultrashort laser pulses with cryogenic He planar jets, i.e., slabs, using 2D3V relativistic particle-in-cell simulations. Of particular interest are laser intensities $10^{15}$–$10^{16}$ W cm$^{-2}$, pulse lengths $\leq 100$ fs, and the wave length regime $\sim 800$ nm for which the slabs are initially transparent and subsequently inhomogeneously ionized. Pulses $\geq 10^{16}$ W cm$^{-2}$ are found to drive ionization along the slab and outside the laser spot, the ionization front propagates along the slab at a considerable fraction of the speed of light. Within the ionized region, there is a highly transient field which is a result of the charge-neutralizing disturbance at the slab-vacuum interface and which may be interpreted in terms of a two-surface-plasmon decay. The ionized region is predicted to reach solid-like densities and temperatures of few to hundreds of eV, i.e., it belongs to warm and hot dense matter regimes. Such extreme conditions are relevant for high-energy densities as found, e.g., in shock-wave experiments and inertial confinement fusion studies. The temporal evolution of the ionization is studied considering theoretically a pump–probe x-ray Thomson scattering scheme. We observe plasmon and non-collective modes that are generated in the slab, and their amplitude is proportional to the ionized volume. Our theoretical findings could be tested at free-electron laser facilities such as FLASH and the European XFEL (Hamburg) and the LCLS (Stanford).

Keywords: warm and hot dense matter, pump–probe x-ray Thomson scattering experiments, dense plasma jets, two-surface-plasmon decay, nonequilibrium x-ray Thomson scattering, cryogenic jets, particle-in-cell simulation

(Some figures may appear in colour only in the online journal)

1. Introduction

Material under extreme conditions—known as warm (WDM) and hot (HDM) dense matter—is located between conventional condensed matter states and ideal plasmas. Neither condensed matter nor plasma models are suited to describe this intermediate region correctly [1, 2]. Moreover, analytical models such as perturbation techniques become increasingly complicated due to partial ionization, electron degeneracy, strong correlations, bound-state level shift, and pressure ionization [3]. The rigorous determination of the equation of state, the radiative opacity, and the transport properties of WDM and HDM is mandatory for controlling the complex sequence of inertial confinement fusion (ICF) experiments [4–6], designing specific compression paths in shock-wave experiments, and developing interior and evolution models for, e.g., giant planets [7–9].

Warm and hot dense matter can be generated and investigated in the laboratory via intense ultrashort laser pulses provided by free electron laser (FEL) facilities such as FLASH and European XFEL in Hamburg and LCLS in Stanford. The state-of-the-art is to perform pump–probe experiments, where a pump pulse heats and compresses the target. To investigate the dynamic processes on such short time scales, the warm or hot dense matter is probed by a second pulse at well-defined time delays with respect to the pump pulse. The probe pulse operates at frequencies higher than the plasma frequency and thereby penetrates the target and scatters off electrons. Therefore, x-ray Thomson scattering (XRTS) [10] is a robust and flexible experimental
diagnostic tool for high-power laser-plasma interactions [11, 12], WDM studies [13–18], astrophysics [19–23], ICF plasmas and shock-wave experiments [24–26].

Different parameters control the energy deposited in the target and, consequently, determine the ionized volume, the electron temperature, the ion temperature, the density, and the degree of ionization. The wavelength of the pump pulse and the geometry of the target are examples of such parameters.

The interaction of optical lasers with H and He droplets has been studied theoretically using particle-in-cell simulations [27–29] which showed that compressed and inhomogeneous overdense plasmas are produced. Ionization inside the interior of the droplet was observed due to the penetration of the laser into the droplet. This could be interpreted in terms of an enhanced electric field at the droplet surface—as predicted by the Mie theory—and a plasma wave propagating from there into the target.

Flexible cryogenic liquid jets have been used in XRTS experiments [30, 31]. Understanding the ionization dynamics and kinetic instabilities during and after the pump pulse is necessary to interpret the XRTS spectrum. In general, the interaction of intense lasers with metals and overdense plasmas has widely been investigated for potential applications [32–39], e.g., plasmonics [40] and high harmonic generation [41]. In particular, the interaction of intense laser pulses with a metallic wire was investigated theoretically and experimentally [42]. A strong transient field propagating at the speed of light along the wire was observed, nevertheless, only a small number of fast electrons is propagating with velocities comparable to the speed of light. The transient field is confined at the surface and evanescent in the normal to the interface in a distance proportional to the skin depth. Transverse magnetic (TM) modes might be coupled to the surface electrons producing surface waves over long distances away from the interaction area.

In this work, we investigate a similar set-up but replace the metal wire with a neutral cryogenic He planar jet, i.e., we use a slab geometry. This allows to probe more complex features of the light–matter interaction. In the case of a di-electric planar jet, an inhomogeneous, possibly overdense, plasma is formed in the interaction area. Each overdense layer oscillates with the driving field and radiates a transient field with harmonics related to its density. When the field exceeds the ionization threshold, ionization takes place at the surface and within the interior of the jet, i.e., the field is not well trapped at the surface. We performed simulations for the interaction of s- and p-polarized laser light with He slabs. Both polarizations lead qualitatively to the same results. Pulses with intensities $\geq 10^{16}$ W cm$^{-2}$ generate overdense plasmas and drive ionization along the jet and outside the laser spot. The ionization front propagates at a considerable fraction of the speed of light.

The paper is organized as follows. Section 2 is devoted to the findings of the numerical simulations for the interaction of intense laser radiation with He slabs. Especially, it explains the plasma generation in He slabs of 0.5 and 5 $\mu$m width. Section 3 discusses possibilities to verify the theoretical findings experimentally by XRTS. We present synthetic spectra for the collective as well the non-collective scattering regimes. At the end we give conclusions.

2. Plasma generation in He slabs

The interaction of optical laser pulses with liquid He slabs is investigated utilizing the Xoopic code [43, 44] which is a 2D3V particle-in-cell code and fully electromagnetic. Xoopic is a multidisciplinary kinetic code for plasma simulation. It can be used to study microwave devices, plasma sources, beam optics, laser and beam plasma interaction, and accelerators; for more details concerning the packages and the simulation parameters please see [45].

Particles in the simulation domain interact with fields defined on the Yee mesh and with material boundaries. In this paper, the simulation domain is a rectangular area, one boundary is used to launch the laser pulse and the other three boundaries are open, thus electromagnetic waves can exit the grid with minimal reflection. The relativistic equations of motion are solved using an implicit-leapfrog scheme which is a second order accurate center difference scheme. The scheme requires few operations and minimal storage [46]. In addition, an explicit Boris scheme is employed, where the position update occurs in a rotated Cartesian frame to avoid the full cross product calculation and the non-physical heating of particles during the rotation around the magnetic field lines [46, 47]. The particle equations of motion are advanced using fields interpolated from the discrete grid to the continuous particle locations, then the field is updated according to the new particles distribution.

The laser energy is delivered to the target via tunneling ionization. Unless otherwise stated, impact ionization is switched off. The tunneling ionization is calculated based on an extension of the Keldysh theory, which provides a formula for the ionization probability of hydrogen-like atoms [48]. The ionization probabilities are extended to ionization from complex atoms and ions and for different polarizations [49]. The collisions of charged particles and neutral particles are considered utilizing a Monte-Carlo scheme [50], where different processes might occur; the interaction of electrons with
neutral particles may lead to impact ionization, excitation, or elastic scattering. Test runs showed that for the intensities considered here the effect of collisional ionization during the laser pulse is much smaller than that of field ionization [29, 51].

2.1. Pumping slabs with a width of 0.5 μm

As a first case, we consider a slab with a width of 0.5 μm along x-axis and a semi-infinite length along y-axis. The slab has an initial density of $2.2 \times 10^{22}$ cm$^{-3}$ and a temperature of 20 K.

As shown in figure 1, the laser pulse comes from the left and propagates along the x-axis in the vacuum until it reaches the slab. The laser wavelength is chosen to be 800 nm. The spatial profile is a Gaussian with 1.5 μm full width at half maximum along the y-axis. The temporal variation of the laser pulse in XOOPIIC can be either trapezoidal, Gaussian or a half-sine. We choose a Gaussian shape with a pulse length of $t_p = 20$ fs (at times $t_{\text{max}} \pm t_p/2$ the value of the envelope is $\text{exp}[-1]$, and zero beyond). The pulse is s-polarized. Different simulations have been carried out for laser intensities from $10^{15}$ to $10^{19}$ W cm$^{-2}$.

In order to minimize the numerical instabilities and to check the simulation convergence, we have chosen different grid sizes from $\Delta x = \Delta y = \lambda/200$ to $\Delta x = \Delta y = \lambda/50$, time steps from $\Delta t = 0.07\Delta x/c$ to $\Delta t = 0.6\Delta x/c$ where c is the speed of light, and the number of super-particles per cell from 60 to 120. All configurations fulfill the Courant–Friedrichs–Lewy criterion and, hence, produce similar results. Therefore, we show unless otherwise stated the results belonging to $\Delta x = \Delta y = \lambda/200$, $\Delta t = 0.07\Delta x/c$, and 120 super-particles per cell.

First, we show the electron density and the degree of ionization at the end of the laser pulse, i.e., at 20 fs for the intensity $10^{15}$ W cm$^{-2}$, see figures 2(a) and (b). Only a small area inside the laser spot (i.e., interaction area) is ionized. An underdense plasma with a degree of ionization close to 1 is formed. As clearly can be seen from figure 2(b), the laser energy is not sufficient to produce He$^{+2}$ ions and the deposited energy is restricted to a small area around the pulse peak.

For a higher laser intensity of $10^{18}$ W cm$^{-2}$, the electron density and the degree of ionization are shown in figures 3(a) and (b), respectively. An overdense plasma is formed: the electron density at the surface reaches 40 times the critical density. The generated plasma is spatially inhomogeneous. The electron density at the near side is greater than the electron density at the rear side. Therefore, figure 3(a) shows implicitly the localization of the energy at the near side of the slab. In figure 3(a), one can easily distinguish between two areas with different degrees of ionization. The laser spot area which is ionized by direct interaction with the laser pulse exhibits a degree of ionization close to 2, i.e., the concentration of He$^{+2}$ is dominant. Outside the laser spot area, a plasma with lower degree of ionization is formed. In addition, it is worth reporting that the Fourier analysis of the field inside the ionized area provides the existence of localized high harmonics with respect to the driving laser frequency. Moreover, all six components of the electromagnetic field, $E_n$, $E_x$, $B_n$, $B_x$, and $B_z$, exhibit a wave-like structure.

The laser pulse consists of a number of subcycles. If the intensity of the first few subcycles can produce overdense plasmas, the next subcycles in the laser pulse will excite a counterpropagating transient electric field along the slab in a process similar to the two-plasmon decay process discussed by Macchi et al [33, 34]. The transient field will ionize the slab outside the laser spot. Hence, the ionized area at the end of the laser pulse depends on the time at which the overdense plasma is generated. The average speed of the ionization front along the slab at the end of the laser pulse is shown in figure 4. In this context, the average speed is half of the length of the ionized area along the slab at the end of the laser pulse over the pulse length. There is an offset in the ionized area at the initial time due to the Gaussian of the laser pulse along y-axis. Therefore, the propagation speed of the ionization front would be smaller than the average speed shown in figure 4.
The average speed is a considerable fraction of the speed of light.

We want to emphasize the difference between dielectric, neutral slabs and metals or pre-ionized overdense plasmas interacting with high intense laser pulses ($10^{18}$ W cm$^{-2}$). In metals and homogeneously pre-ionized overdense plasmas with step-density profile, the laser pulse excites a strong transient field at the interface (the surface) instantaneously, the transient field propagates with the speed of light \[35, 42\]. This field is confined at the surface and evanescent normal to the interface in a distance proportional to the skin depth. TM modes might be coupled to the surface electrons producing surface waves over long distances away from the interaction area \[32–39\]. The field oscillations may decay parametrically into two counterpropagating electron surface waves labeled + and −, respectively, where the three-wave process holds:

\[ k_l = k_+ + k_-, \quad \omega_l = \omega_+ + \omega_- \] (1)

Here, $\omega_l$ and $k_l$ are the frequency and the wavenumber of the driving field, respectively. $\omega_+$ and $k_+$ are the frequencies and the wavenumbers of the surface waves. Note that no pre-imposed target modulation or grating is required to satisfy the conditions of equation (1). In dielectric slabs, on the other hand, an inhomogeneous overdense plasma is formed in the interaction area, hence, the criterion of the two-surface-plasmon decay is matched. Each overdense layer oscillates with the driving field and radiates a transient field with harmonics related to its density. When the field strength exceeds the ionization threshold, ionization takes place. Moreover, the field can propagate also to the interior of the slab, i.e., it is not well trapped at the surface. Scattering of and interference between different fields in the plasma may occur. Therefore, the speed of the ionization front along the slab is always smaller than the speed of light.

2.2. Pumping slabs with a diameter of 5 μm

As a proposal for future experiments on inhomogeneous HDM and because the cryogenic sources of small jets (width $\approx 0.5$ μm) might not be stable, we repeated the simulation, however, assuming 100 fs pulse length and a slab width of 5 μm. Other parameters are kept the same. For a laser intensity $10^{15}$ W cm$^{-2}$, a small area within the interaction area is weakly ionized. Again, underdense plasmas with a degree of ionization of about one are formed. Increasing the laser intensity produces overdense plasmas, see figure 5(a). Consequently, part of the laser pulse energy is turned into transient fields. Therefore, a new energy absorption window opens. As clearly seen in figure 5(b), the transient field drives ionization along the slab. The average speed of the ionization front along the slab is always smaller than the speed of light.

2.3. Time evolution of the ionization after the laser pulse

After the laser pulse, the plasma waves in the interaction area sustain the transient field—this is due to the oscillation of...
critical surfaces and the charge-neutralizing disturbance at the slab edge. The ionization of the jet continues to hundreds of femtoseconds. Figure 7 shows the temporal variation of the ionized area length along He slabs as well as the temporal variation of a length corresponding to the speed of light (solid line). Here we consider the results of the two examples mentioned in section 2.1 (represented via blue stars) and in section 2.2 (represented via red stars) with a laser intensity of $10^{18}$ W cm$^{-2}$. To reveal the role of impact ionization, we display in figure 7 also the results of the same two examples (represented via blue and red diamonds), however, considering impact ionization in addition to tunnel ionization. In order to run the simulations for these two cases until 0.5 ps and due to the limitation of available computational resources, we performed the simulations within another configuration: $\Delta x = \Delta y = \lambda/50$, $\Delta t = 0.5\Delta x/c$, and 60 super-particles per cell. Moreover, we increased the width of the simulation domain, so that the number of hot electrons, which can escape from the simulation domain, is negligible. The coincidence between the results considering and neglecting impact ionization proves that this process is not important during the laser pulse.

The main phenomena found in our simulations are the following: (i) one can easily deduce that the ionized area increases with time and the ionization front propagates with a considerable fraction of the speed of light. (ii) The over-all number of free electrons is still increasing long after the laser pulse. (iii) The degree of ionization is, however, highly inhomogeneous. (iv) Approximately, local equilibrium conditions are fulfilled. The electron distribution function of the whole system, however, is a superposition of Maxwell distributions with different temperatures.

A promising experimental tool to track these time-dependent phenomena could be XRTS performed at FEL.

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**Figure 5.** The electron density in units of the critical density (a) and the degree of ionization (b) at 100 fs. The laser intensity is $10^{18}$ W cm$^{-2}$.

**Figure 6.** The average speed of the ionization front at 100 fs along the slab divided by the speed of light versus the laser intensity.

**Figure 7.** The temporal variation of the ionized area along the slabs for laser intensity of $10^{18}$ W cm$^{-2}$ with and without impact ionization (II), the field ionization is considered in all cases. Solid line represents the length corresponding to the speed of light.
facilities within pump–probe experiments. The x-ray pulses of about 100 fs duration have high intensities so that the evolution of the plasma on ultra-short time scales as well as non-equilibrium phenomena such as surface waves could be resolved. Therefore, we will revisit the XRTS theory and present new results in the next chapter.

3. XRTS spectrum

3.1. Theoretical basics

The cross section for a photon with certain wave vector and frequency, \( \mathbf{k}_0, \omega_0 \), into a photon state with \( \mathbf{k}, \omega \), is related to the dynamic structure factor (DSF) of all electrons according to:

\[
\frac{d^2\sigma}{d\omega d\Omega} = \sigma_T S_{ee}^{\text{tot}}(\mathbf{k}, \omega)
\]

with the Thomson scattering cross section \( \sigma_T \) and the dynamical structure factor of the electrons, \( S_{ee}^{\text{tot}}(\mathbf{k}, \omega) \), where \( \mathbf{k} = \mathbf{k}_0 - \mathbf{k}_0 \) and \( \omega = \omega_0 - \omega_0 \).

According to Chihara’s approach [52], the DSF can be expressed by a sum of independent contributions

\[
S_{ee}^{\text{tot}}(\mathbf{k}, \omega) = Z_e S_{ee}(\mathbf{k}, \omega) + \left[ f(k) + q(k) \right]^2 S_k(k, \omega)
+ Z_b \int d\omega' S_k(k, \omega) S_k(k, \omega - \omega').
\]

The first term denotes the DSF of free electrons with \( Z_e \) being the number of free electrons per nucleus. The second term characterizes the scattering from tightly and weakly bound electrons. Its amplitude is determined by the sum of the form factor \( f(k) \) of bound electrons and the screening cloud \( q(k) \) of free electrons; the ion–ion structure factor, \( S_k(k, \omega) \), which represents the thermal motion of the ions. The last term is due to the contribution of the bound-free transitions \( S_k \). This term describes Raman-type transitions of inner shell electrons to the continuum which are modulated by the ion motion contained in \( S_k(k, \omega) \) and multiplied by the number of core electrons.

Here we will concentrate on the free electrons’ contribution. In thermodynamic equilibrium, the DSF of free electrons can be expressed via the fluctuation-dissipation theorem by the inverse dielectric function \( \epsilon^{-1}(k, \omega) \):

\[
S_{ee} = -\frac{\epsilon_0 \hbar k^2}{\pi e^2 n_e} \operatorname{Im} \epsilon^{-1}(k, \omega) = \frac{\epsilon_0 \hbar k^2}{\pi e^2 n_e} \operatorname{Im} \frac{-\hbar\omega}{\hbar^2 k^2}.
\]

The dielectric function is usually calculated in random phase approximation or generalizations like the Born–Merrin approximation [27]. Relation (4) is used also for inhomogeneous situations with only local thermodynamic equilibrium. In this case, the Thomson scattering signals of different volume elements with local temperature and densities are summed up.

In non-equilibrium situations with stronger deviations of the electron distribution from an equilibrium form, equation (4) is not applicable. Instead, one can use the non-equilibrium random phase approximation expression [3, 53–55] for the
Figure 9. Collective (a) and non-collective (b) Thomson scattering spectrum of free electrons of the 5 μm jet considering impact ionization beside field ionization, a laser intensity of $10^{18}$ W cm$^{-2}$, and a pulse length of 100 fs at 354 and 504 fs. The x-axis is the energy shift, $h\omega$, of the photons.

For a Maxwell–Boltzmann distribution function, i.e. in equilibrium, the integral can be performed analytically to give

$$S_{ee}^0(k, \omega) = \frac{m_e}{\hbar k \sqrt{2\pi m_e k_B T}} \exp \left[ -\frac{m_e (\omega / c + \frac{\hbar k}{2m_e})^2}{2k_B T} \right]. \quad (9)$$

The asymmetry of this (equilibrium) function is connected with the detailed balance condition

$$\frac{S_{ee}^0(k, -\omega)}{S_{ee}^0(k, \omega)} = \exp \left( \frac{\hbar \omega}{k_B T} \right). \quad (10)$$

Because $|\varepsilon(k, -\omega)| = |\varepsilon(k, \omega)|$, the detailed balance equation is also valid for $S_{ee}(k, \omega)$.

3.2. Calculation of synthetic XRTS spectra for the simulation case

The PIC simulation results of the cryogenic He slab pumped by the intense ultrashort laser pulse show highly inhomogeneous conditions for the ionization degree and free electron density. Concerning the use of local temperatures, one has to be aware of the limitations: PIC simulations require fine spatiotemporal grids; moreover, the number of super-particles in each cell is finite. Therefore, the electron distribution cannot be fitted in each cell with Maxwell–Boltzmann distributions. Instead, we look for larger, semi-homogeneous regions (i.e., group of cells in the PIC simulation) and approximate the electron distribution by a Maxwell–Boltzmann distribution with an effective temperature $k_B T_{eff} = 2/3E$. The distributions functions in some regions have a noisy, but very hot tail. Thus, to avoid the over-estimation of the electron temperature, we excluded in the calculation of the mean energy all electrons with energies greater than 1 keV. We found that the cut-off at 1 keV is appropriate from the statistical point of view for most of the cells, as shown in figure 8.

We would like to emphasize that changing the arbitrary cut-off will change the estimated temperature, but the results in figure 9 will be qualitatively the same. Especially, hot electrons will not contribute to collective plasma excitations (plasmons). Therefore, for inhomogeneous systems with local thermodynamic equilibrium conditions, the scattering signal can be evaluated theoretically by a sum of the different cells. This applies especially for the collective scattering regime, i.e. a probe laser wavelength $\lambda_0 = 13.5$ nm and for a scattering angle of 60°. We use the plasma parameter profiles from the PIC simulations of the 5 μm slab, laser intensity $10^{18}$ W cm$^{-2}$, and considering impact ionization and field ionization at 345 and 504 fs.

The overall Thomson scattering due to free electrons is displayed in figure 9(a). The collective signal has been calculated using equation (4). As shown, the amplitude of the plasmons increases by time due to the propagation of the transient field and the increase in the ionized volume. We want to note that in inhomogeneous systems, despite that each cell or group of cells obeys approximately local thermodynamic equilibrium, the total system might be far away from equilibrium. Hence, the full system is not characterized just by one Maxwell–Boltzmann distribution with an effective temperature.

In the non-collective scattering regime, no assumption on local thermodynamic equilibrium is necessary provided the velocity distribution function $f_e(p)$ of the whole scattering volume is known from the PIC simulations. In this regime we have $|\varepsilon_R(k, \omega)|^2 \approx 1$ and $S_{ee} \approx S_{ee}^0$ determined by equation (8). In order to calculate the non-collective scattering off the jet, we consider a probe pulse with a wavelength of 2 nm and a scattering angle of 160°. Figure 9(b) displays the non-collective scattering of free electrons at the same delay times as in figure 9(a). Again, the intensity of the spectrum at 504 fs is larger than that at 354 fs due to the increase in the ionized volume. Moreover, the qualitative behavior of the results in
figure 9(b) depends only weakly on $\xi$ provided the criteria of non-collective scattering are fulfilled.

4. Conclusion

We studied the interaction of ultrashort and intense laser pulses with cryogenic He slabs. For intensities $\geq 10^{16}$ W cm$^{-2}$, the laser pulse has been found to produce overdense plasmas and strong transient fields sufficient to ionize the slab outside the interaction area. The ionization front propagates along the jet with a considerable fraction of the speed of light. An appropriate experimental method to infer this ionization dynamics in the planar jets after excitation by an intense laser pulse could be XRTS. Of course we are aware of the difficulties to resolve the highly transient nature of these evolving plasmas. To illustrate the principal prospects of the method, synthetic XRTS spectra have been calculated for different delay times after the laser pulse and different scattering conditions. The calculated amplitude of the plasmons in the collective scattering regime has been found to increase as a function of delay time whereas the position does not change by more than some percent. The intensity of the non-collective scattering signal increases as well with time, connected with the increase of the ionized volume and, therefore, the number of scatterers. Beside the variation of the time delay between pump and probe pulse, it might be useful to vary the focus of the probe beam in order to get information on the spatial extension of the ionized area.

We want to remark that the transient fields inside the plasma which cause ionization outside the interaction area are a result of kinetic instabilities as two-plasmon decay, oscillation of critical surfaces, and the charge disturbance at the slab edge (i.e., the jet surface in 3D) due to the escape of hot electrons. All these phenomena might be interrupted with the ion motion on ps time scales. Therefore, we expect the decay of the transient field and the stop of the ionization wave along the slab after few to tens ps. Because the kinetic simulation is demanding, we postpone the radiation-hydrodynamic investigation of the decay of the transient field to future studies.

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