Plasma Formation Dynamics in Intense Laser-Droplet Interaction

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We study the ionization dynamics in intense laser-droplet interaction using three-dimensional, relativistic particle-in-cell simulations. Of particular interest is the laser intensity and frequency regime for which initially transparent, wavelength-sized targets are not homogeneously ionized. Instead, the charge distribution changes both in space and in time on a sub-cycle scale. One may call this the extreme nonlinear Mie-optics regime. We find that—despite the fact that the plasma created at the droplet surface is overdense—oscillating electric fields may penetrate into the droplet under a certain angle, ionize, and propagate in the just generated plasma. This effect can be attributed to the local field enhancements at the droplet surface predicted by standard Mie theory. The penetration of the fields into the droplet leads to the formation of a highly inhomogeneous charge density distribution in the droplet interior, concentrated mostly in the polarization plane. We present a self-similar, exponential fit of the fractional ionization degree which depends only on a dimensionless combination of electric field amplitude, droplet radius, and plasma frequency with only a weak dependence on the laser frequency in the overdense regime.

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\textit{Introduction} — Spherical, wavelength-sized, homogeneous dielectric or metal objects in plane-wave electromagnetic radiation fall into the realm of Mie theory \cite{1} and are of fundamental importance in optics. Standard Mie theory provides the electromagnetic field configuration inside and outside a homogeneous sphere of a given dielectric constant, assuming an incoming plane wave. However, nowadays available short and intense laser pulses interacting with matter create plasmas on a sub-laser period time scale \cite{2,3}. These plasmas, in turn, modify the further propagation of the laser pulse. We call this the extreme nonlinear optics regime, and in the case of (initially) spherical targets nonlinear Mie optics.

As the laser field propagation is determined by the electron density distribution and the plasma is generated by ionization, the charge state and density distributions is expected to be sensitive to the ionization dynamics. In fact, even the strongest present-day lasers cannot directly fully ionize heavier elements so that the assumption of a preformed, throughout the target homogeneous plasma with a given dielectric constant may be inadequate. Furthermore, the skin-effect may prevent the laser from penetrating into targets that turn overdense in the course of ionization so that, in general, a richly structured space and time-dependent charge distribution develops \cite{4}. Such interactions of laser pulses with rapidly self-generated plasmas have found already applications, e.g., as “plasma mirrors,” which are routinely used to increase the pulse contrast for intense laser-matter experiments \cite{5,6}.

One expects that the part of the laser pulse that is scattered off an overdense target will be mainly determined by the ratio of laser to plasma frequency at the surface whereas possible inhomogeneities inside the target do not play a role. In fact, standard Mie scattering theory assuming a homogeneous, overdense plasma sphere was used to characterize rare gas clusters in recent experiments on a shot-to-shot basis \cite{7,8}. However, inside such a sphere, standard Mie theory would predict electric fields only within a narrow skin layer while, in this Letter, we will show that a highly inhomogeneous and temporally changing charge density distribution may be created in the droplet interior. In order to probe such inhomogeneous structures inside the target laser frequencies greater than the plasma frequency that corresponds to the maximum plasma density in the target should be used. Indeed, Thomson scattering of present-day short-wavelength free-electron laser radiation (from, e.g., DESY in Hamburg, LCLS in Stanford, or SACLA in Japan) is employed to probe overdense plasmas \cite{9,11}.

Molecular dynamics is a powerful tool that is widely used to describe the ionization dynamics in small laser-driven clusters \cite{12,14}. However, for wavelength-sized targets such as droplets the influence of the target on the propagation of the incident electromagnetic wave needs to be taken into account self-consistently. This requires the solution of Maxwell’s equations together with the equations of motion for the charged particles. In the case of weakly coupled plasmas the problem can be reduced to the solution of the Vlasov-Maxwell system of equations, which is efficiently achieved using particle-in-cell (PIC) codes \cite{2}.

Numerically, we study the non-linear Mie domain by means of a 3D relativistic PIC simulations with ionization included. The code UMKA originated from the study in \cite{15}. We show that in a certain laser intensity regime the droplet target is neither fully ionized nor are charges only created at the droplet surface. Instead, fields penetrate under a characteristic angle into the droplet, ionizing atoms in the polarization plane and triggering plasma waves which collide in a focal spot. We present results for the fractional ionization degree at various laser intensities, wavelengths, and densities, that turn out to follow an universal scaling law.

\textit{Simulations} — The ionization of an ion with charge state $Z - 1$ and ionization potential $I$ due to the electric field $E$ is
initially neutral He droplet. A spatial resolution of \( \Delta \) of the interaction of an intense, plane-wave laser pulse with an considered anymore during the current time step \[\text{[19]}\]. Energy conservation is accounted in a cell is insufficient for further ionization, this cell is not per cell were used. Absorbing boundary conditions for the fields and particles were employed in the propagation direction, periodic ones for the other directions. The size of the simulation box was always chosen big enough to rule out any boundary effects on the observables of interest due to reflections or particles leaving the box. A linearly (in \( y \)) boundary was located at \( x = 4 \lambda \) parallel to the electric field at the ion location. The value of \( j_{\text{ion}} \) is such that \( j_{\text{ion}} \) \( \cdot \mathbf{E} \) is the work spent on ionization per time step \[\text{[17, 18]}\]. Energy conservation is accounted for during the whole process; if the remaining field energy in a cell is insufficient for further ionization, this cell is not considered anymore during the current time step \[\text{[19]}\].

We start by presenting typical results from PIC simulations of the interaction of an intense, plane-wave laser pulse with an initially neutral He droplet \[\text{[20]}\]. A spatial resolution of \( \Delta x = \Delta y = \Delta z = \lambda / 100 \), 125 macro-ions and 250 macro-electrons per cell were used. Absorbing boundary conditions for the fields and particles were employed in the propagation direction, periodic ones for the other directions. The size of the simulation box was always chosen big enough to rule out any boundary effects on the observables of interest due to reflections or particles leaving the box. A linearly (in \( y \)-direction) polarized 10-cycle sin\(^2\)-laser pulse of carrier frequency \( \omega_0 \) enters the numerical box through the boundary \( x = 0 \) and propagates into the region \( x > 0 \). The dimensionless vector potential amplitude \( a = |eA/mc| = |eE/m\omega_0c| \) was 0.5, corresponding to a laser intensity \( I \simeq 5.2 \times 10^{17} \text{ W/cm}^2 \), the wavelength \( \lambda = 2\pi c/\omega_0 \) was 800 nm (i.e. for a laser period \( T_L = 2.66 \text{ fs} \)). The density of the 2R = 4\( \lambda \) = 3.2\( \mu \text{m} \) diameter He-droplet was \( \rho = 0.14 \text{ g/cm}^3 \). If the droplet was completely preionized such a density would correspond to an electron density \( n_{e0} = 24 n_{cr} \), where \( n_{cr} = 1.8 \times 10^{21} \text{ cm}^{-3} \) is the critical density for 800-nm wavelength light. The droplet center was located at \( x = 4\lambda \), \( y = z = 10\lambda \). In the simulations implemented using the tunneling ionization rate formula \[\text{[16]}\]

\[
w(E) = \left( \frac{2E_{ch}}{|E|} \right)^{2n^*} \frac{k^2 h}{m} \frac{|E|}{E_{ch}} \exp \left( -\frac{2E_{ch}}{3|E|} \right) \]

(1)

with \( k = \sqrt{2mI/\hbar^2} \), \( E_{ch} = \hbar^2 k^2/2m \), \( n^* = Z\sqrt{|E|} \). Here, \( m \) is the electron mass and \( I_H \) is the ionization potential of atomic hydrogen. When an ionization event takes place a free electron at rest is created at the position of the ion. The energy needed for ionization is taken out of the field via an “ionization current” \( j_{\text{ion}} \) parallel to the electric field at the ion location. The vector potential \( \mathbf{A} \) is \( \mathbf{E} \) is the work spent on ionization per time step \[\text{[17, 18]}\]. Energy conservation is accounted for during the whole process; if the remaining field energy in a cell is insufficient for further ionization, this cell is not considered anymore during the current time step \[\text{[19]}\].

Results — Figure 1 shows snapshots of the volume distribution of electron and He\(^{2+} \) densities. In the beginning the droplet is non-ionized and thus transparent for the leading part of the laser pulse. Later, as the field strength of the laser pulse increases in magnitude, ionization becomes more efficient, and an overdense plasma is generated rapidly on the droplet surface as the pulse propagates over it, leading finally to almost full ionization of a thin surface layer. Moreover,

FIG. 1: (color online). Electron density in the beginning (a) and at the end (b) of the interaction with the laser pulse. He\(^{2+} \) density (c) at the end of the interaction. For better visualization of the droplet interior a quarter of it was cut-out. Laser and droplet parameters are given in the text. The laser propagation direction is indicated by an arrow in each panel.

FIG. 2: (color online). Electron (bottom), He\(^{1+} \) (middle) and He\(^{2+} \) (top) density in two perpendicular planes (\( \hat{k}, \mathbf{E} \)) (left) and (\( \hat{k}, \mathbf{B} \)) (right) at \( t = 7T_L \) (a) and \( t = 12T_L \) (b). Laser and droplet parameters are given in the text.
we observe that a highly inhomogeneous density distribution inside the droplet is formed, concentrated mostly in the polarization plane. In particular, there seems to be a focal spot (blue area in the polarization plane in Fig. 1b and 1c). The fractional ionization degree \( I \) (denoted by the blue area in the polarization plane in Fig. 1b and 1c) of the droplet at the end of the interaction is \( \approx 35\% \). The field enhancement predicted by Mie theory is in excellent agreement with the PIC results (\( \approx 1.9 \) times the incident field).

**Mie field enhancement** — We attribute the fact that the field and ionization front dynamics originate from a surface region under a certain angle \( \theta \geq \pi/2 \) with respect to \( \hat{\mathbf{k}} \) to a local, time-dependent field enhancement on the droplet surface. In order to corroborate this statement, we show in Fig. 4 the radial electric field along the droplet surface in the polarization plane vs time and \( \theta \) as obtained from the PIC simulation (a) and according to Mie theory (b). Standard Mie theory is formulated for plane incident waves. As Mie theory is linear we synthesized our pulse via spectral decomposition and added the fields coherently. In the Mie simulation the droplet is assumed to be homogeneous and conducting, with a dielectric constant \( \epsilon = 1 - n_e n_\infty \). Under such conditions Mie theory predicts in the strongly overdense regime (where the skin depth is \( \delta_e \approx c/\omega_p \ll R \) with \( \omega_p = \sqrt{4\pi n_e n_\infty / m_e} \) the electron plasma frequency) that the electric field on the droplet surface is perpendicular to it. In Fig. 4 the time axis has been shifted such that \( t = 0 \) corresponds to the moment when the maximum of the incident laser pulse arrived at the droplet center. Both PIC and Mie result predict maxima of the electric field on the droplet surface for angles \( \theta/\pi \in [0.4, 0.7] \). The slight disagreement in the field distributions in forward direction (small \( \theta \)) is due to the fact that in the Mie calculation the droplet is assumed conducting (i.e., completely ionized) from the very beginning whereas in the PIC simulation there is not yet plasma at the rear side of the droplet (see Fig. 2). The fractional ionization degree — Figure 6 collects all our results on the ionization dynamics (small \( \theta \)). Fractional ionization degree — Figure 6 collects all our results on the ionization dynamics (small \( \theta \)).

**Focused plasma waves** — The propagation direction \( \chi \) of the field structures inside the droplet seen in Fig. 3 is tilted with respect to \( \hat{\mathbf{k}} \), leading to the observed focusing effect. In order to interpret correctly these structures, we project the field structures inside the droplet onto the polarization plane and as predicted by Mie theory (b). The horizontal black lines indicate the angle at which the electric field at the droplet surface is highest.

**Fractional ionization degree** — Figure 6 collects all our results on the ionization dynamics (small \( \theta \)).
I complete ionization face gets ionized, one expects big droplets, when only the thin skin layer on the droplet surface tunneling ionization the electric field amplitude matters, not collisionless skin depth \( \delta \). Inserting the expression for the ionization rate formula (1). We find

\[
\gamma = \frac{a}{(R \lambda / \delta^2) - 2 \pi m R (\omega_p^2 - \omega_0^2)}
\]

in the tunneling ionization rate formula. Introducing the dimensionless parameter \( \eta = a/(R \lambda / \delta^2) \), it turns out that for all the various cluster sizes \( R > \delta \), densities \( \omega_p^2 \gg \omega_0^2 \), laser intensities and wavelengths simulated, \( I_r \) is well described by \( I_r \sim 1 - \exp(-\eta \gamma) \). In our case of He we find \( \gamma = 1560 \). Note that the species-dependence only enters via the ionization potentials \( I \) in the tunneling ionization rate formula (1). Inserting the expression for the collisionless skin depth \( \delta_c = c/\sqrt{\omega_p^2 - \omega_0^2} \) we obtain

\[
\eta = \frac{e E}{(2 \pi m R (\omega_p^2 - \omega_0^2))} \approx \frac{e E}{(2 \pi m R \omega_p^2)}
\]

showing that there is only a weak dependence on the laser frequency. Indeed, for tunneling ionization the electric field amplitude matters, not the laser frequency. For small laser intensity and sufficiently big droplets, when only the thin skin layer on the droplet surface gets ionized, one expects

\[
I_r = \frac{4 \pi R^2 \omega_p^2}{2 \pi m R \omega_p^2} \sim R^{-1}
\]

In the opposite limit of very high laser intensity or small droplets complete ionization \( I_r = 1 \) is expected. Both limiting cases are contained in our formula. The chosen exponential interpolation between those two limiting cases matches the simulation results for the fractional ionization degree very well.

**Summary** — A strong near-infrared or optical laser pulse interacting with an initially neutral, wavelength-sized He droplet may generate a charge density distribution that neither is homogeneous throughout the droplet nor created only within a thin skin layer at the surface. Instead, electric fields may penetrate into the droplet interior for certain angles of incidence predicted by standard Mie theory. However, the time-dependent field and density distributions inside the target get are not accessible to standard Mie theory but fall into the realm of extreme nonlinear optics. The field penetration causes ionization inside the droplet, mainly confined to the polarization plane. The resulting inhomogeneous charge distribution may be probed via scattering of short-wavelength radiation and should be taken into account when studying typical laser-plasma interaction applications such as ion acceleration or x-ray radiation from recombination in ionized droplets. A particularly high abundance of He\(^{2+}\) is observed where the ionization fronts and the trailing plasma waves collide. The fractional ionization degrees for various droplet and laser parameters are found to be in good agreement with a self-similar exponential fit. At higher laser intensities a qualitatively similar ionization dynamics is expected for higher-Z materials as well.

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