Field localization in submicron-sized clusters and its effect on the ionization dynamics

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Submicron-sized clusters are attractive targets in intense laser-cluster interaction experiments. The internal field in the Mie scattering regime is inhomogeneous and size-dependent. Here we investigate the field localization and its effect on ionization in submicron-sized clusters. The diffractive focusing reduces the ionization threshold of these clusters and leads to an ionization hotspot at the rear of the clusters. At a higher intensity, a skin layer of plasma at the front surface shields the field, stopping further rear side ionization. Interestingly, an ionized core with a plasma shell at a suitable condition may excite hybridization of plasmons, allowing transiently enhanced field beyond the skin depth. Control and optimization of the field localization and the resulting ionization may have practical importance for relevant laser-cluster experiments.

Laser-cluster interaction has been the subject of active research for several decades [1–3]. The localized high density in connection with the isolated environment enables efficient laser-matter coupling, providing not only a unique platform for studying the ultrafast nonequilibrium dynamics [4] but also a multitude of applications ranging from ion accelerators [5, 6] and electron accelerators [7] to neutron sources [8] and nuclear isomers sources [9].

Ionization is the first step in the intense laser-cluster interaction. The surprisingly efficient ionization and heating observed in experiments has sparked a continued effort to unravel the underlying mechanism [2, 10]. One of the reasons is the field enhancement in the cluster, which may originate from field resonance at the critical density layer [11] or field amplification due to cluster polarization [12]. The geometry effect of the cluster can also lead to beam focusing, increasing the local field strength. For particles with size of tens of microns or more, the refractive focusing of the dielectric sphere increases the internal field, giving rise to reduced ionization threshold [13] and efficient supercontinuum generation [14]. While refractive focusing breaks down at approximately $10\lambda$ [15], where $\lambda$ is the laser wavelength, diffractive focusing emerges at a smaller size, leading to the internal or external field enhancement commonly referred as a photonic nanojet [16]. The sub-wavelength field localization behind the particle have found applications in super-resolution imaging [17] and nanopatterning [18], and the field localization at the inner rear surface has been exploited for controlled ablation that enables backward jet propulsion of particles [19]. As the size decreases to a fraction of the laser wavelength, the scattering enters the Rayleigh regime, and the internal field becomes uniform and field enhancement occurs when the laser frequency approaches the resonant frequency.

A through understanding of the ionization dynamics of submicron-sized clusters has practical significance for laser-cluster applications. While many experiments use nanometer-sized clusters in the Rayleigh regime, submicron-sized clusters are also used for electron acceleration [20] and ion acceleration [5, 21, 22]. Even with a moderate average size, some clusters are large since the size distribution is typically broad [23]. Extremely large clusters may also be present in pulsed gas jets under certain conditions even though the scaling law predicts the size is small [24, 25]. In particular, the ionization threshold is relevant in proton acceleration using intense laser irradiated clusters or foils, where the contrast ratio is essential [7, 26] and a precisely controlled pre-pulse plasma is desirable [26]. In addition, ionization of particles in air such as dusts and waterdrops plays an important role in mid-infrared filamentation [27–29].

In this paper, we numerically investigate the field localization in submicron-sized clusters and the effect of the enhanced field on the cluster ionization dynamics. Near the ionization threshold, the diffractive focusing of submicron-sized clusters increases the local field at the rear side of the cluster and creates an ionization hotspot. At a higher intensity, the leading part of the pulse ionizes the front surface of the cluster. The resulting overdense plasma layer shields the field, shutting off diffractive focusing effect. While such a plasma shell typically shields the laser field, a transient field enhancement is observed in some simulations, which may be attributed to the coupling of sphere plasmon and cavity plasmon.

Before significant ionization, the fields inside and around a cluster are calculated using a Mie code SCAT-TNLAY v2.0 [30], which numerically evaluates an analytical solution in terms of infinite series with scattering coefficients. These coefficients are obtained from the Maxwell equations in together with the boundary conditions at the interfaces. These calculations are simple and allows rapid parameters scanning. The standard Mie theory of a dielectric sphere is a linear theory which is appropriate when the nonlinear effect such as ionization and Kerr effect can be neglected. In our case, the cluster size is much smaller than the self-focusing distance and Kerr effect is negligible.

Figure 1(a) and 1(b) show the field distribution of argon clusters of two different sizes subject to a 800-nm
continuous plane wave linearly polarized in y direction. With a 10-nm radius, only the near field outside the cluster is redistributed and the internal field is uniform, as it is in the Rayleigh scattering regime. With a 400-nm radius, the light is focused in a hotspot with a size of tens of nanometers at the rear of the cluster. This complicated field distribution can be considered as the superposition of multiple radiation. Similar pattern is found in hydrogen clusters and xenon clusters as well, as shown in Fig. 1(c) and 1(d).

FIG. 1. Field distribution in (a) a 10-nm-radius argon cluster, (b) a 400-nm argon cluster, (c) a 400-nm hydrogen cluster, (d) a 400-nm xenon cluster. The blue lines superimposed in the images are the lineout at y = 0.

The dependence of the internal field enhancement on cluster sizes for commonly used gas species are shown in Fig. 2. The refractive indices of the bulk medium are $n_{H_2} = 1.11$ [31], $n_{Ar} = 1.28$ [32], $n_{CO_2} = 1.40$ [33], and $n_{Xe} = 1.49$ [32]. The maximum internal field reaches the incident value $E_0$ at around $r = 130$ nm. After that, it almost increases linearly with the size. A higher refractive index results in a stronger enhancement. We do not calculate for clusters larger than 0.6-μm due to limits of numerical precision. As the size increases, the number of digits needs to be increased to calculate of spherical harmonics.

When the laser pulse is intense enough to ionization the clusters, the action and reaction of the laser field and plasma give rise to a highly dynamic and nonlinear process, which is not captured by static Mie calculations. To investigate this ionization dynamics, we performed three-dimensional particle-in-cell (3D PIC) simulations using an open-source code SMILEI [34]. This code includes various physics modules such as field ionization, binary collision with collisional ionization (CI).

The simulation set-up is presented in Fig. 3. The simulation box was $6.4 \times 2.4 \times 2.4 \mu$m$^3$. The spatial step $\Delta x = \Delta y = \Delta z = \lambda/64$, and the time step $\Delta t = 0.023$ fs, corresponding to a Courant number of 0.956. A laser pulse linearly polarized in the y direction enters the simulation box from the left. The laser wavelength is centered at 800 nm, and the temporal profile is Gaussian. The full width at half maximum of the field is two cycles. We assume a plane wave with constant profile. A submicron-sized hydrogen cluster is located at the center of the simulation box. The time zero $t = 0$ marks the moment that the peak of the pulse reaches $x = 0$. The number density of hydrogen atoms in the cluster is assumed to be that of the liquid hydrogen, which is $4.56 \times 10^{22}$ cm$^{-3}$ or $26.8 n_c$ [35], where $n_c$ is the critical density of the plasma at $\lambda = 800$ nm. Each cell in the clusters has 125 macro-particles. Silver-muller boundary conditions are adopted for the electromagnetic fields in the longitudinal direction, and periodic boundary conditions are used for the transverse directions. Quasi-static rates for tunnel ionization are used with Monte-Carlo procedure. Electron-electron collision and electron-ion collision with collisional ionization are included using the binary colli-
The code does not account for recombination and lowering of ionization potential. As these two effects contribute in the opposite way to the average charge [36], they may get partially canceled.

Since PIC codes typically consider scenarios where permittivity is dominant by that of free electrons, the index of the neutral atoms is assumed to be unity. Thus in normal configuration, the diffractive focusing effect is absent in the code. To imitate the index contrast between the clusters and the environment, we assume that a plasma with 0.2\(n_{cr}\) density is present for \(-r < x < r\), as indicated by the light green region in Fig. 3. Note that these plasmas do not participate in the collisional process.

Figure 4(a) and 4(b) show the electron density in two perpendicular planes (y,x) and (z,x) at \(t = 16\) fs when a 500-nm cluster is irradiated with a pulse of \(7 \times 10^{13}\) W/cm\(^2\) intensity. The colorbar indicates the electron density normalized by the critical density \(n_c\). The transverse field \(E_y\) map (c) and longitudinal field \(E_z\) map (d) in the y-x plane at \(t = 1.0\) fs. The fields are normalized by the incident amplitude \(E_0\). (e) The electron density at \(t = 16\) fs with no index difference between the cluster and the surrounding. (f) The electron density for a 200-nm cluster at \(t = 16\) fs.

The ionization maximum occurs at the rear of the cluster only for a limited range of intensity near the threshold. At a higher intensity, the front surface becomes heavily ionized. The electron density and field distribution for a 500-nm cluster subject to \(I = 2 \times 10^{14}\) W/cm\(^2\) are presented in Fig. 5 (a) and 5(b), respectively. At this intensity, the leading part of the pulse is intense enough to create sufficient ionization at the front surface. Due to the skin effect, this ionization is limited to a thin layer, and the internal field is shielded as demonstrated in Fig. 5(b). Figure 5(c) shows the electron density without the diffractive focusing. The overall ionization pattern is similar, except for the minor feature at the rear of the cluster. Figure 5(d) shows the case of a 200-nm cluster, where a higher plasma density is observed.
FIG. 6. (a) Time evolution of the average charge in the cluster for $I = 7 \times 10^{13}$ W/cm$^2$ with a 500-nm cluster (solid blue line), a 500-nm cluster with no index contrast (dashed orange line), and a 200-nm cluster (dotted green line) (b) The cases for $I = 2 \times 10^{14}$ W/cm$^2$. The dashdotted gray line represents the case for a 500-nm cluster without collisional ionization, and the dashdotdotted purple line represents the case for a 200-nm cluster without collisional ionization.

respectively. Near the ionization threshold, a larger size causes stronger diffractive focusing and is beneficial for ionization. At a higher intensity, however, a smaller size leads to a higher average charge. This is because more atoms are in the skin layer. In addition, the charge state at the front surface is higher as shown in Fig. 5. Note that the ionization continues after the pulse is gone. This rise is mainly due to collisional ionization. As we turn off the collisional ionization, the charge state for the 500-nm cluster (dashdotted gray line) levels off. However, a significant increase remains after the pulse for the 200-nm cluster (dashdotdotted purple line).

To clarify this delayed ionization, the electron density map and field distribution at $t = 0$ and $t = 2.4$ fs are shown in Fig. 7. Figure 7(b) shows that the internal field is shielded at the peak of the pulse. However, at $t = 2.4$ fs, the internal field is enhanced, as shown in Fig. 7(d). This difference can be caused by the different densities of the plasma layer shown in Fig. 7(a) and 7(c). It is known that a dielectric-core/plasma-shell system allows field enhancement at a wavelength substantially longer than that of Mie resonance at $\omega_p/\sqrt{3}$ because the symmetric coupling of the sphere plasmon and cavity plasmon reduces the resonance frequency [40, 41]. In the Mie regime, the inhomogeneous polarization and retardation effects can excite higher order modes and have more resonances [42]. Indeed, quadrupole pattern is shown in Fig. 7(d).

In summary, we have shown the diffractive focusing effect in an unionized dielectric cluster and our PIC simulations have confirmed the ionization hotspot at the rear of the submicron-sized cluster. At a higher intensity, however, the diffractive focusing only causes a minor difference in the ionization due to the shielding of the plasma layer at the front surface. As the laser turn the cluster into an unionized core with an ionized thin layer, the hybridization of plasmon in cluster-core/plasma-shell may allow resonant enhancement of field, which is uncommon for near-infrared laser pulse at solid density target. Control and optimization of the field localization and the ionization dynamics of clusters in the Mie regime may find applications in laser-cluster experiments. It can also facilitate our understanding and control of ionization and heating phenomena in nanostructures and solids for the application of laser-based material processing [18].

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