Field enhancement in clusters and clustered plasmas in the Mie scattering regime

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Clusters and clustered plasmas in the Mie scattering regime are attractive targets in intense laser-cluster interaction experiments. In contrast with clusters in the Rayleigh regime, the internal field has a strong size dependence. In this paper, we investigate the electric field in dielectric clusters and clustered plasmas due to Mie scattering. In sub-wavelength sized dielectric clusters, the well-known diffractive focusing appears, increasing the maximum field. In fully ionized homogeneous clusters, while the center field decreases at large sizes due to skin effect, it may initially increase with the size at shorter wavelength. During intense laser interaction with large clusters, the skin effect prevents the field from penetrating, leaving clusters as an unionized core with a plasma shell. This core-shell clusters may greatly enhance the internal field at suitable sizes due to hybridization of plasmons. Control and optimization of large cluster sizes to enhance the internal field may have practical significance for laser-cluster experiments such as proton acceleration.

I. INTRODUCTION

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 strong laser-plasma coupling, opening new avenues for ion accelerators [4, 5], electron accelerators [6], X-ray sources [7], as well as neutron sources [8]. In addition to various practical applications, laser-excited clusters also provide a unique testbed to investigate the ultrafast nonequilibrium dynamics [9].

Many aspects of the interaction including multiple ionization [10], energy absorption [11], high harmonic generation [12], ion acceleration [13], and X-ray emission [14] exhibit a strong dependence on the cluster size. In these studies, clusters are often produced using high-pressure gas jets. To control the cluster size produced by gas jets, the backing pressure and reservoir temperature of the gas jet are often varied, or custom-designed nozzle geometries are used [15], and the average cluster size is estimated by scaling law [16] or characterized by Rayleigh scattering [16] or Mie scattering [17] depending on the size range.

Increasing the local field strength in the cluster is also important to optimize the intense laser interaction with clusters. Little attention has been paid to the interplay between the field and cluster size in high energy density physics. This is because the used clusters are often in the Rayleigh scattering regime. As a result, the internal field $E_c = 3E_0/\epsilon + 2$ is size-independent. Here $\epsilon$ is the dielectric constant of the bulk medium, and $E_0$ is the incident laser field. In simple models such as the nano-plasma model, the field is taken to be uniform. The assumption of field uniformity is often unsatisfied.

When $r \gtrsim 0.1\lambda$, where $r$ is the cluster radius and $\lambda$ is the laser wavelength, Mie theory is required to accurately calculate the electric field. In addition to dipole radiation, Mie scattering includes multipole radiation, and the pattern is more complicated than that in the Rayleigh scattering regime. The far-field measurement of the scattering has been used for particle size characterization [17], and recent near field studies have revealed a photonic nanojet effect behind the particle [18] with applications of super-resolution imaging [19]. Several studies also show an increased internal intensity within a large sphere, which leads to efficient supercontinuum generation in ultrashort laser interaction with 30-μm radius water droplets [20], and reduced breakdown threshold in air with water-cloud droplet [21].

![FIG. 1. Schematics of clusters and clustered plasmas in a laser field with filed strength $E_0$. (a) An unionized cluster with radius $r$. (b) An ionized clusters with radius $r$. (c) A cluster with unionized core and ionized skin layer. Here $r_1$ is unionized core radius, $r_2$ is total cluster radius, $\epsilon_1$ is the dielectric constant for core, and $\epsilon_2$ is the dielectric constant for the plasma.](image-url)

Sub-micron sized clusters are often used in experiments for electron acceleration [22] and ion acceleration [4, 23, 24]. Even with a moderate average size, some clusters are large since the size distribution is typically broad [25]. Extremely large clusters may also be present in pulsed gas jets under certain conditions even though the scaling law predicts the size is small [26, 27]. In this paper, we investigate the internal field enhancement due to Mie scattering in sub-micron sized unionized dielectric clusters, clustered plasmas, and clusters with a plasma shell, as depicted by Fig. 1. The fields...
inside and around a cluster are calculated using a well-benchmarked Mie code Scattlay v2.0 [28], 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. In Sec. II, we investigate the field distribution in dielectric clusters, and the well-known diffractive focusing appears within subwavelength sized clusters, which increases the maximum internal field. In Sec. III, we consider the internal field in a clustered plasma with a uniform density. We find that shorter wavelength the field can increase with the size. In Sec. IV, we investigate a cluster with an unionized core and an ionized skin layer. This is the typical scenario for large clusters in intense laser pulses [22, 29]. With appropriate core-shell ratio, this core-shell structure substantially enhances the field, allowing for a strong field beyond the skin depth. In the few-cycle regime, due to the large discrepancy between the laser frequency and Mie frequency, efficient energy coupling requires special attention and can be met by nonlinear resonant absorption [30, 31] or Mie oscillation [32]. Our study may offer an alternative approach to enhance the laser-cluster coupling by optimizing the cluster size, which may find applications in experiments such as proton acceleration.

II. LARGE UNIONIZED CLUSTERS

Before cluster ionization starts, the field distribution can be modeled using the standard Mie theory of a dielectric sphere. The Kerr effect in clusters is small and not considered. Figure 2(a) and 2(b) show field distribution in argon clusters in a linearly polarized 800-nm plane wave. 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 cluster radius, the light is focused in a hotspot with a size of tens of nanometers at the rear of the cluster. Similar pattern is found in hydrogen clusters and xenon clusters as well, as shown in Fig. 2(c) and 2(d). While this focusing is similar to the diffractive focusing of a ball lens, the underlying physics is diffraction rather than refraction as diffractive focusing breaks down at approximately 10λ [33].

FIG. 2. 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 laser wavelength is 800 nm. The blue lines superimposed in each 2D images are the lineouts at x = 0.

FIG. 3. Maximum field as a function of cluster radius r in a hydrogen cluster (solid red line), an argon cluster (dashed blue line), a CO2 cluster (dash-dotted green line), and a xenon cluster (dotted magenta line).

The dependence of the field enhancement on cluster sizes for commonly used cluster species are shown in Fig. 3. The refractive indices of the bulk medium are nH2 = 1.11 [34], nAr = 1.28 [35], nCO2 = 1.40 [36], and nXe = 1.49 [35]. The maximum field almost increases linearly with the cluster size. A higher refractive index results in a stronger enhancement.

In a clustered gas jet, the size has a large spread. Thus the formation of a hotspot in larger clusters effectively lowers the minimum intensity of ionization, rendering a tighter requirement on the contrast ratio in ultra-intense laser-cluster interaction to eliminate the prepulse ionization [6]. The size distribution of clusters may also be modified by the prepulse. This diffractive focusing in laser-cluster interaction can also facilitate our understanding and control of ionization and heating phenomena in nanostructures and solids for the application of laser-based material processing [37].

III. LARGE UNIFORMLY IONIZED CLUSTERS

A cluster in intense laser pulse gets ionized rapidly and becomes a clustered plasma. During the ultrashort pulse, the ion motion is negligible. Therefore, after the clus-
ters are fully ionized, the rest of the pulse interacts with a sphere of uniform plasma density. Indeed, the model of preformed homogeneous plasma throughout the clusters is often used [30, 38, 39] to explain interesting phenomenon in laser-cluster interaction. In this section we investigate the field distribution in preformed clustered plasmas due to Mie scattering. Note that the formation of a large uniform plasma large sphere may require special pre-ionized scheme such as X-ray pulse ionization due to the limits of skin depth.

The dielectric constant of a plasma with uniform density \( n_e \) is given by

\[
\epsilon_c = 1 - \frac{2}{\omega_p^2 (1 + \nu_c / \omega)}. \tag{1}
\]

Here \( \omega_p = \sqrt{n_e e^2 / \epsilon_0 m_e} \) is the plasma frequency, \( e \) is the elementary charge, \( \epsilon_0 \) is the vacuum permittivity, \( m_e \) is the electron mass, \( \omega \) is angular frequency of the laser pulse, and \( \nu_c \) is the electron-ion collision frequency.

In our study we assume a fully ionized hydrogen cluster, which is often used in proton acceleration [23, 40] and has a lower intensity for fully ionization. The atomic density of the cluster is assumed to be that of the liquid hydrogen, which is \( 4.56 \times 10^{22} \text{ cm}^{-3} \) [40]. This corresponds to a plasma frequency \( \omega_p = 1.2 \times 10^{16} \text{ rad/s} \). The collision frequency is on the order of 1 \( \text{ fs}^{-1} \) [41], even when the surface collision is included [42], and we have used \( \nu_c = 1 \text{ fs}^{-1} \) for our calculation.

Figure 4 shows the field strength at the center (a) and maximum internal field (b) as a function of cluster radius \( r \) for three driving laser wavelength. Field strength at the center (c) and maximum internal strength (d) as a function of cluster radius with different collision frequency.

Figure 4(a) and 4(b) show the field at the cluster center and the maximum field in the cluster as a function of the size for laser of 400 nm, 533 nm, and 800 nm wavelength. Representative 2D field profiles are shown in Fig. 5. For 400-nm wavelength, the center field and maximum internal field increase with an increasing cluster size up to approximately 40 nm, and the internal field is greater than the incident field. For greater sizes, the center field decays with the size and the maximum internal field oscillates with a decreasing oscillation amplitude. For 800-nm wavelength, the center field decreases monotonically with the size. The oscillation is due to the interference of the incident field and scattered field, and the decay of center field is due to skin effect. The pattern for the center field and maximum field preserves for different collision frequency as shown in Fig. 4(c) and 4(d). The profiles in Fig. 5 show that the maximum internal field is located in the periphery of the cluster and the minimum is near the center.

Figure 6 shows the center field and maximum internal field as a function of incident wavelength for different sizes. In the Rayleigh regime at 5-nm size, the field enhancement exhibits a resonance at 0.27 \( \mu \text{m} \), which is exactly the Mie frequency \( \omega_p / \sqrt{3} \). The full width at half maximum of the resonance is more than 50-nm. The field enhancement gradually decreases as the frequency moves away from the Mie frequency. As the cluster size increases, the peak of the center field is red-shifted. This is because the separation between the charges at two poles increases, which reduces the restoring force [43]. The maximum internal field develops minor peaks due to excitation of higher-order modes. Note that the real part of the refractive index increases for longer wavelength for \( \lambda > 2.7 \mu \text{m} \). The complex refractive index is \( 0.28 + 2.3 j \) at 400 nm, \( 0.98 + 4.7 j \) at 800 nm, and \( 3.1 + 8.3 j \) at 1.6 \( \mu \text{m} \). Despite of the large real refractive index at longer wavelength, no diffractive focusing is observed due to strong absorption coefficient.
IV. LARGE CLUSTERS WITH AN IONIZED SKIN LAYER

For submicron-sized clusters, the intense laser pulse does not normally produce a homogeneous overdense plasma. This is because the skin effect prevents the field from further penetrating. After the ionization at the skin layer is complete, the laser interacts with a target consisting of an ionized core and a plasma shell of skin-layer thickness. This scenario is supported in several simulations and experiments. Particle-in-cell simulations of the femtosecond laser interaction with micron-sized target shows that the ionization and heating is limited to the skin depth in helium droplets [29] and hydrogen droplets [44]. Experiments have shown that the average radius of CO$_2$ clusters in the wake of the prepulse is reduced from 0.18 $\mu$m to 0.15 $\mu$m [45], indicating the the ionization occurs at peripheral region of the clusters just above the ionization threshold.

The skin depth for collisionless plasmas is $l_{skin} = c / \sqrt{\omega_p^2 - \omega^2}$, where $c$ is the speed of light in vacuum. If the collision frequency is comparable to the laser frequency and $\omega \ll \omega_p$, the skin depth can be modified as [46]

$$l_{skin} = \frac{c}{\omega_p} \sqrt{\frac{2[1 + (\nu_c/\omega)^2]}{1 + 1 + (\nu_c/\omega)^2}}.$$ \hspace{1cm} (2)

For $\lambda = 800$ nm and $\nu_c = 1$ fs, the skin depth of hydrogen clustered plasmas is roughly 25 nm. Since most intense laser studies use Ti:sapphire laser systems, where the wavelength is 800-nm or 400 nm with frequency doubling, we focus on the wavelength range between 400-nm and 800-nm. For this range of laser frequency and collision frequency, the skin depth does not vary significantly.

In the following calculations, we keep the shell thickness fixed, and vary the radius of unionized cluster core.

The dielectric-core/plasma-shell system allows field enhancement at a wavelength substantially longer than that of Mie resonance at $\omega_p/\sqrt{3}$. Figure 7(a) shows the electric field at the center of the cluster as a function of the core size $r_1$ with a 2.5-nm shell thickness. The angular frequency of driving laser is $\omega_0$ (red), $1.2\omega_0$ (blue), $1.4\omega_0$ (green), $1.6\omega_0$ (magenta), $1.8\omega_0$ (cyan), and $2\omega_0$ (yellow).

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The field enhancement is observed for all of these frequencies. The two peaks at $\lambda = 400$ nm appears at 19 nm and 117 nm while at $\lambda = 800$ nm, the resonance occurs at 80 nm and 275 nm, respectively. The corresponding 2D field profiles are given in Fig. 8, which shows that the field penetrates beyond the skin depth, and field is relatively uniform in the central part and periphery part of the core.

For a shell-core system with field enhancement, in general, the temporal evolution is necessary to be considered,
as the enhanced field will further ionize the interior region and destroy the shell structure. Our model is suitable for a two-color scheme. An ionizing pulse at an off-resonant wavelength creates a cluster with a plasma shell and a neutral core. When this layered cluster interacts with the main pulse at the resonant wavelength, the field beyond the skin depth can be enhanced rather than shielded due to excitation of plasmons, ionizing the interior of the cluster and enhancing the laser energy absorption. Recent simulations show that a hollow Au nanocluster inside a hollow nanosphere enhances the proton acceleration [47]. Our shell-core clusters, which are practical to produce, bear some similarities with such a system. Thus, these clusters are likely to find applications in proton acceleration.

The behavior of the first resonance is the dipole resonance, which is qualitatively similar to that of resonance in the Rayleigh scattering, as shown in Fig. 7 for \( \delta = 2.5 \) nm. As the frequency increases, the resonance occurs at a smaller core size, and the amplitude of the first resonance increases. The symmetric coupling of the sphere plasmon and cavity plasmon reduces the resonance frequency, which is given by at [48, 49]

\[
\frac{\omega^2}{\omega_p^2} = \frac{1}{2} \left( 1 - \frac{1}{3} \left( 1 + \frac{8 \delta r_1}{r_2^3} \right)^3 \right)
\]

This formula is for a hollow plasma shell, and if we assume a core index of 1.0, the location of the peak match with the theory exactly, which is the case for the dashed curves in Fig. 7(b). The dielectric core slightly shifts the peak to the larger size. The resonant characteristic was analyzed in the thin and thick shell limit in the Mie scattering regime [50]. In the Mie regime, the inhomogeneous polarization and retardation effects excites higher order modes. As shown in Fig. 7(a), there is a prominent peak at 275-nm. This is a result of quadrupole resonance [43]. This resonance is relatively insensitive to the shell thickness, which is shown in Fig. 9. As the shell thickness increases, the optimal size for the prominent peak increases very little, but the width of peak decreases substantially.

V. SUMMARY

We have calculated the laser field in an unionized dielectric cluster, clustered plasmas with uniform density, and clusters with a plasma shell. The diffractive focusing in unionized submicron-sized clusters increases the internal field. In fully ionized homogeneous clusters, field enhancement can occur at shorter wavelength. While the real refractive index is larger at longer wavelength, the strong absorption inhibits diffractive focusing. The hybridization of plasmon in cluster-core/plasma-shell allows strong resonant enhancement of field, which is uncommon for near-infrared laser pulse at solid density target. Our model is a reasonable approximation for laser-cluster interaction prior to significant ionization and right after the ionization is complete. While particle-in-cell simulation provides accurate results, it requires intense computational resources and the interpretation is often elusive due to interplay of various effects. Our simple model allows rapid parameters scanning and reveals underlying mechanism for field enhancement that may occur during complicated laser-cluster interaction. Control and optimization of this field enhancement by selecting appropriate cluster size has practical significance for laser-cluster experiments such as proton acceleration.

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[43] A. M. Ahmed, A. Mehaney, M. Shaban, and A. H. Aly, Materials Research Express 6, 085073 (2019).

[44] U. Zastrau, P. Sperling, C. Fortmann-Grote, A. Becker, T. Bornath, R. Bredow, T. Döppner, T. Fennel, L. B. Fletcher, E. Förster, S. Göde, G. Gregori, M. Harmand, V. Hilbert, T. Laarmann, H. J. Lee, T. Ma, K. H. Meiws-Broer, J. P. Mithen, C. D. Murphy, M. Nakatsutsumi, P. Neumayer, A. Przystawik, S. Skruszewicz, J. Tiggesbäumker, S. Toleikis, T. G. White, S. H. Glenzer, R. Redmer, and T. Tschentscher, J. Phys. B: At., Mol. Opt. Phys. 48, 224004 (2015).

[45] A. Y. Faenov, I. Y. Skobelev, T. A. Pikuz, S. A. Pikuz, V. E. Fortov, Y. Fukuda, Y. Hayashi, A. Pirozhkov, H. Kotaki, T. Shimomura, H. Kiriyama, S. Kanazawa, Y. Kato, J. Colgan, J. Abdallah, and M. Kando, Laser Part. Beams 30, 481 (2012).

[46] F. F. Chen, Phys. Plasmas 8, 3008 (2001).

[47] M. Mehrangiz, Plasma Phys. Controlled Fusion (2021), doi:10.1088/1361-6587/ac4312.

[48] G. Mukhopadhyay and S. Lundqvist, J. Phys. B: At. Mol. Phys. 12, 1297 (1979).

[49] E. Prodan and P. Nordlander, J. Chem. Phys. 120, 5444 (2004).

[50] D. C. Tzarouchis and A. Sihvola, IEEE Trans. Antennas Propag. 66, 323 (2018).