Collisionless absorption of short laser pulses in a deuterium cluster: dependence of redshift of resonance absorption peak on laser polarization, intensity and wavelength

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We study collisionless absorption of short laser pulses of various intensity, wavelength ($\lambda$) and polarization in a deuterium cluster using molecular dynamics (MD) simulation. For a given laser energy and a pulse duration $\approx 5$-fs (fwhm), it is found that maximum laser absorption does not happen at the well-known static Mie-resonance or linear resonance (LR) wavelength of $\Lambda_M \approx 263$ nm (for deuterium cluster) irrespective of linear polarization (LP) and circular polarization (CP) state of laser. As the laser intensity increases, the absorption peak is gradually red-shifted to a higher $\lambda$ in the marginally over-dense regime of $\approx (1-1.5)\lambda_M$ from the expected static $\lambda_M$ owing to gradual outer ionization and cluster expansion; and above an intensity the resonance absorption peak disappears (sometimes followed by even a growth of absorption) when outer ionization saturates at 100% for both LP and CP. This disappearance of the resonance absorption peak should not be misinterpreted as the negligible (or no) role of Mie-resonance. In fact, in this marginally over-dense band of $\approx (1-1.5)\lambda_M$, some electrons undergo dynamic Mie-resonance (dynamic LR) and others anharmonic resonance when they are freed. It is also found that before the absorption peak, laser absorption due to LP and CP lasers are almost equally efficient (CP case being inappreciably higher than LP) for all intensities and $\lambda$. However, after the absorption peak, at lower intensities, absorption due to LP appreciably dominates absorption due to CP with increasing $\lambda$ which gradually reverses at higher intensities. MD results are also supported by a naive rigid sphere model of cluster.

I. INTRODUCTION

The interaction of intense laser pulses with nanoscale targets, particularly with atomic clusters demonstrates enhanced absorption of laser [1] than expected from isolated atoms/molecules irradiated by same laser pulses. The localized solid-like density of a clustered target (deuterium cluster here) allows the laser field to penetrate fully with no attenuation, which helps in such an extraordinary amount of laser energy absorption. The rising edge of the laser pulse of intensity $> 10^{15}$ W/cm$^2$ ionizes the constituent atoms of the cluster (inner ionization) through optical field ionization (OFI) and forms a nano-plasma. These inner ionized electrons are governed by the laser field plus the transient local electrostatic field (due to charge separation) and escape from the cluster periphery (outer ionization) after absorbing energy from the remaining part of the laser pulse. Subsequent outer ionization of electrons leaves the cluster with a net positive charge which explodes due to inter-ionic Coulomb repulsion. The high energy absorption by escaping electrons [2-8], emission of x-rays [9-12] (typically in the KeV range) and finally the explosion of cluster resulting emission of ions with MeV energies [13-19] and MeV neutrals [20] in some conditions.

Several experimental, theoretical and particle-simulation works on laser-cluster interaction have reported the effect of various parameters of laser and cluster (i.e., peak intensity, wavelength, pulse duration, cluster size and type etc.) on average charge per atom, mean electron and ion energies and also total absorbed energy. However, Petrov et al [21-23] by MD simulations claimed that (i) there is “no enhancement of absorbed energy near the plasmon resonance” [21] while laser wavelength $\lambda$ is varied, (ii) absorbed energy depends linearly on $\lambda$ [23] and (iii) the Mie-resonance or linear resonance (LR) plays no role for the enhancement of absorbed energy, without giving any plausible justification. Note that Petrov et al [21] considered only three well-separated $\lambda = 100,248,800$ nm and did not resolve the wavelength-space meticulously while passing through the Mie-resonance, which possibly led to such misleading conclusions. On the contrary, we reported dependence of energy absorption on $\lambda$ for an argon cluster with linearly polarized (LP) laser by MD simulation [24]; and MD results were supported by a rigid sphere model (RSM). We showed that for a given pulse energy, maximum laser absorption in a cluster happens not at the well-known static Mie-resonance wavelength of $\Lambda_M$, but at a red-shifted $\lambda$ which lies in the marginally over-dense band of wavelength $\Lambda_d \approx (1-1.5)\lambda_M$. Note that linear Mie-theory (as in the case of nano-plasma model [3, 25]) is valid only at lower intensities where static Mie-resonance at $\lambda = \lambda_M$ is possible which is often identified as a sharp peak in the absorption curve. However, with increasing laser intensity, linear Mie-theory fails and resonance absorption peak is gradually red-shifted from $\lambda_M$ [24]. At this shifted $\Lambda_d \approx (1-1.5)\lambda_M$ all the possible resonances, i.e., dynamical linear resonance (LR) and anharmonic resonance (AHR) are unified [24] to yield maximum laser absorption. We termed this combined resonance as the unified dynamical linear resonance (UDLR) and strongly concluded that there is always a redshift of the absorption peak with respect to $\lambda_M$ in the marginally over-dense band of $\Lambda_d \approx (1-1.5)\lambda_M$ irrespective of the laser intensity, cluster size and laser pulse type [24].

However, for different types of clusters, it is still unknown how the redshift of the absorption peak changes with laser intensity. This is particularly very important to justify and validate the above mentioned UDLR [24] in the wavelength band of $\Lambda_d \approx (1-1.5)\lambda_M$ with a different cluster type (other than the argon cluster) for its universal acceptance. Further, the degree of polarization of the impinging laser pulse is also an
important parameter that changes the dynamics of the cluster electrons, hence it might effect the UDLR and the redshift of the absorption peak which remains to be explored. An electron under the influence of a circularly polarized (CP) laser moves spirally, whereas in LP laser it executes oscillatory motion perpendicular to the direction of the laser propagation. The spiral motion of electron in CP prevents the electron-scattering from the cluster boundary, whereas the probability of rescattering is more for LP. Particle-in-cell (PIC) simulations [26] performed for a xenon cluster at different intensities and cluster charge densities but at a fixed $\lambda = 1056$ nm (with immobile ions) concluded that energy absorption and outer ionization in CP and LP laser field are almost equally efficient. However, it is not known (i) how absorbed energies compare with LP and CP laser fields with the variation of $\lambda$ and (ii) how UDLR (in the marginally overdense regime) helps in the redshift of absorption peaks from the $\Lambda_M$ in LP and CP fields. Note that, even if laser intensity is kept fixed, ponderomotive energy $U_p = E_0^2\lambda^2/4$ of electron and its dynamics is affected by varying $\lambda$ which is expected to contribute to the redshift of the absorption peak. We thus present energy absorption and outer ionization for a deuterium cluster (as a different cluster type other than argon) with CP and LP lasers. Particularly, we show the effect of laser polarization on the redshift of the absorption peak in the collisionless regime. In passing, we also disprove some of the claims of Petrov et al [21] by performing additional MD simulations and discuss some deficits there corroborating to plasmon resonance.

We consider a deuterium cluster of radius $R_0 \approx 2.05$ nm (charge state $Z = 1$) which is irradiated by LP and CP laser pulses of different peak intensities $I_0 = 5 \times 10^{15} - 5 \times 10^{17}$ W/cm$^2$ and $\lambda = 100 - 800$ nm. For a given intensity and polarization we vary $\lambda$. Similar to previous studies with argon clusters [23], here we find that, for both LP and CP, redshifts of the absorption peaks from the static Mie-resonance wavelength $\Lambda_M = 263$ nm (for deuterium cluster) still persist, which also lie in the marginally over-dense band of wavelength $\Lambda_d \approx (1 - 1.5)\Lambda_M$ as long as outer-ionization is below 100%. Redshift of the absorption peak monotonically increases with increasing $I_0$. Additionally, above a certain $I_0$, absorption peak is found to disappear with further increase in $I_0 \geq 10^{17}$ W/cm$^2$ as outer ionization saturates at 100%. It is also found that before the absorption peak in the band of $\Lambda_d \approx (1 - 1.5)\Lambda_M$, laser absorption due to LP and CP lasers are almost equally efficient (CP case being inappreciably higher than LP) for all intensities and $\lambda$. However, after the absorption peak, at lower intensities $\leq 10^{17}$ W/cm$^2$, absorption due to LP inapprica- bly dominates absorption due to CP with increasing $\lambda$ which gradually reverses at higher intensities $\geq 10^{17}$ W/cm$^2$.

Atomic units (i.e., $m_e = |e| = 1.44\pi e_0 = 1.\hbar = 1$) are used in this work unless mentioned explicitly. Section II discusses the form of the laser pulse. Section III illustrates laser absorption and the role of laser polarization on the redshift of the absorption peak in the deuterium cluster by a MD simulation. Section IV gives justification of MD results by RSM analysis. Summary and conclusion are given in Sec V.

II. THE LASER PULSE

As laser wavelengths ($\lambda = 100 - 800$ nm) are much longer than cluster sizes (2-4 nm), as considered here, the effect of propagation of laser (directed in $z$) is disregarded and the dipole approximation for laser vector potential $\vec{A}(\tau, t) = \vec{A}(t) \exp(-i2\pi\tau/\lambda) \approx \vec{A}(t)$ is assumed. In general, we write

$$\vec{A}(t) = \frac{E_0}{\omega} \sin^2 \left(\frac{\omega t}{2n}\right) \left[\delta \cos(\omega t) \hat{x} + \sqrt{1 - \delta^2} \sin(\omega t) \hat{y}\right],$$

for $0 \leq t \leq nT$. Here $\delta$ is the degree of ellipticity ($0 \leq \delta \leq 1$); $\delta = 1, 1/\sqrt{2}$ for LP and CP respectively; $n$ is the number of laser period $T$; $\tau = nT$ is the total pulse duration and $E_0 = \sqrt{8\pi I_0/c}$ is the field strength for the peak intensity $I_0$. The components of driving laser electric field $\vec{E}_i(t) = -d\vec{A}/dt$ along $x, y$ and $z$ directions are,

$$\vec{E}_x^x(t) = \frac{E_0}{\omega} \left[\sum_{i=1}^{3} c_i \omega_i \sin(\omega_i t) \right] \text{ if } 0 \leq t \leq nT$$
$$\text{otherwise};$$

$$\vec{E}_y^y(t) = \sqrt{1 - \delta^2} \frac{E_0}{\omega} \left[\sum_{i=1}^{3} c_i \omega_i \cos(\omega_i t) \right] \text{ if } 0 \leq t \leq nT$$
$$\text{otherwise};$$

$$\vec{E}_z^z(t) = 0.$$

Where $c_1 = 1/2, c_2 = -1/4, \omega_1 = \omega, \omega_2 = (1 + 1/n)\omega$, and $\omega_3 = (1 - 1/n)\omega$. Note that the ponderomotive energy $U_p = E_0^2/4\omega^2$ is the same for both LP and CP. For LP, only the $x$-component of laser electric field $\vec{E}_x^x(t)$ survives which may vanish at the completion of each laser cycle. Whereas, for CP, electric field components $\vec{E}_x^x(t), \vec{E}_y^y(t)$ in $x, y$ do not vanish simultaneously. Therefore, CP laser is expected to yield different results than LP laser while interacting with a cluster. Particularly, it is not known how resonance absorption maxima shifts w.r.t. polarization state of laser.

III. RESONANCE ABSORPTION BY MD SIMULATION

In a previous study [24] we reported MD simulation results for argon clusters of different sizes (2-4 nm) irradiated by LP laser fields with different peak intensities and wavelengths $\lambda = 100 - 800$ nm. We showed that in the short pulse regime (5 fs, fwhm) and at a given laser intensity, linear resonance (LR) and an-harmonic resonance (AHR) can be dynamically unified to yield maximum laser energy absorption at a particular $\lambda$, typically in the UV regime. The possible unification of all resonances comprises: (i) LR in the initial time of plasma creation, (ii) LR during Coulomb explosion in the later time and (iii) AHR for electrons in the intermediate time during the laser cluster interaction, leading to maximum energy absorption, maximum outer ionization and also maximum average charge states for the argon cluster. We found the wavelength $\Lambda_0$ of the absorption maxima is gradually red-shifted from the conventional static Mie-resonance wavelength $\Lambda_M$ in the band of $\Lambda_d \approx (1 - 1.5)\Lambda_M$ for increasing
$I_0 = 5 \times 10^{15} - 10^{17} \text{W/cm}^2$. We coined this marginally overdense regime $\Lambda_0 \approx (1 - 1.5)\lambda_M$ as the regime of unified dynamical linear resonance (UDLR), where dynamical LR and AHR are efficiently unified [24].

Here, we report a comparative study of laser absorption and outer ionization and the dependence of the red-shift of absorption maxima in the UDLR regime with a deuterium cluster for different polarization states of laser pulses.

A. Details of MD simulation

The main workhorse here is the MD simulation code. More details are given in Refs. [23, 27]. For conciseness, we only mention necessary points here. A single deuterium cluster of radius $R_0 \approx 2.05 \text{ nm}$ and $N = 1791$ number of neutral atoms is considered, unless mentioned explicitly. Initially, atoms are placed according to the Wigner-Seitz radius $r_w \approx 0.17 \text{ nm}$, giving $R_0 = r_w N^{1/3} \approx 2.05 \text{ nm}$. When all atoms are ionized initially, it gives a charge density $\rho_i \approx 7 \times 10^{-3} \text{ a.u.}$ and $\omega_0 \approx \sqrt{4\pi\rho_0/3} = 0.1735 \text{ a.u.}$. For 800 nm, it represents an over-dense plasma with $\rho_i/\rho_0 \approx 27.87$ and $\omega_0/\omega \approx 3.05$, where $\rho_0 \approx 1.75 \times 10^{27} \text{ m}^{-3}$ is the critical density. Ionization of deuterium atoms is treated by “over the barrier” ionization (OBI) model [5] of optical field ionization (OFI) [28, 30],

$$E_c = I_p^2(Z)/4Z,$$

which is valid at higher intensities $> 10^{14} \text{ W/cm}^2$. Here $I_p(Z)$ is the ionization potential for the charge state $Z$ and $E_c$ is the corresponding critical field. The position and velocity of a newly born electron are assumed same as the parent ion conserving the momentum and energy. The equation of motion (EOM) of $i$-th charge particle in the laser field with the electric field $E_i(t)$ and the magnetic field $B_i(t)$ reads,

$$\frac{d\vec{p}_i}{dt} = F_i(r_i, v_i, t) + q_i \left[ E_i(t) + \vec{v}_i \times \vec{B}_i \right],$$

where $F_i = \sum_{j \neq i} q_j q_i r_{ij} / r_{ij}^3$ is the Coulomb force on $i$-th particle of charge $q_i$ due to all other $N_p - 1$ particles each of charge $q_j$, where $N_p$ is the total number of particles including ions and electrons. Components of laser electric fields are chosen from Eqs. [2, 3, 4] depending upon LP and CP cases. Accordingly, components of laser magnetic fields are oriented. For $I_0 < 10^{15} \text{ W/cm}^2$ usually $B_1(t) \ll 1$. To mitigate Coulomb singularity of $F_i$ for $r_{ij} \to 0$, an artificial smoothing parameter $r_0$ is added with $r_{ij}$. For a given cluster we choose $r_0 = r_w$, which produces accurate Mie-plasma frequency $\omega_{M}$ (see Ref. [27]). The modified Coulomb force on $i$-th particle and the corresponding potential at its location are,

$$F_i = \sum_{j=1, i \neq j} q_j q_i r_{ij} / (r_{ij}^3 + r_0^3)^{1/2}, \quad \phi_i = \sum_{j=1, i \neq j} q_j / (r_{ij}^2 + r_0^2)^{1/2}.$$

Eq. (5) is solved using the velocity verlet method with a time step $\Delta t = 0.01 \text{ a.u.}$ to resolve the $\omega_M$.

Figure 1. (Color online) MD results showing average total absorbed energy $\overline{E}_r$ per atom (a) and corresponding fractional outer ionization $N$ versus $\lambda$ for deuterium cluster ($R_0 = 2.05 \text{ nm}, N = 1791$) at different peak intensities $I_0 = 5 \times 10^{15} \text{ W/cm}^2 - 5 \times 10^{17} \text{ W/cm}^2$. At a particular $I_0$, pulse energy for all $\lambda$ is kept constant with constant pulse duration $\tau = 13.5 \text{ fs}$ (fwhm $\approx 5$-fs). Vertical dashed line indicates $\lambda_{M}$ that corresponds to LR wavelength for $Z = 1$. The shaded bar highlights that absorption maxima are redshifted in the marginally overdense regime of $\lambda = (1 - 1.5)\lambda_M$.

The deuterium cluster is irradiated by short laser pulses of duration 13.5 fs (FWHM of 5-fs) at different $I_0 = 5 \times 10^{15} \text{ W/cm}^2 - 5 \times 10^{17} \text{ W/cm}^2$. The pulse energy is kept fixed for a particular $I_0$ and wavelength is varied in the range of $\lambda = 100 - 800 \text{ nm}$ for both LP and CP laser light.

B. Absorption and outer ionization with LP laser

Figures 1(a)-(b) depicts average total absorbed energy $\overline{E}_r = \sum_{i=1}^{N_e} (\phi_i^2 / 2 + \phi_i) / N$ per atom and corresponding fractional outer ionization $N = N_{out} / N$ of electrons at the end of 13.5 fs LP laser pulses versus $\lambda$ for different $I_0$, where $N_{out}$ is the number of electrons outside the initial radius $R_0$. At lower $I_0 \leq 5 \times 10^{16} \text{ W/cm}^2$, $\overline{E}_r$ and $N$ initially increase with increasing $\lambda$, attain different maximum values at different $\lambda$ between 263 - 400 nm, i.e., in the band of $\lambda = \Lambda_0 \approx 330 \pm 67 \text{ nm}$ which is equivalent to $\Lambda_d \approx (1 - 1.5)\lambda_M$, then drop as $\lambda$ is increased further. For the deuterium cluster, the static Mie-resonance (or the static LR) with a sharp absorption maximum is often conventionally expected at $\lambda = \lambda_M \approx 263 \text{ nm}$ (marked
by vertical dashed line) according to the nano-plasma model. In stead, it is seen that $\delta_E$ and $\overline{N}$ attain maximum values in the band of $\Lambda_d \approx (1 - 1.5)\lambda_M$ which are red-shifted from the expected $\lambda_M \approx 263$ nm, and the redshift increases with increasing values of $\delta_E$ and $\overline{N}$ for increasing $I_0 \lesssim 5 \times 10^{16}$ W/cm$^2$. For higher $I_0 > 5 \times 10^{16}$ W/cm$^2$, absorption peaks gradually disappear due to faster saturation of outer ionization of electrons to 100%. Approaching towards $\lambda_M \approx 263$ nm from 100 nm, higher intensity pulse expels more electrons from the cluster at a much faster rate than at a lower intensity [see Fig.1(a)-(b)]. As electrons move far from the cluster, the laser field dominates over the restoring field of background ions acting on those free/quasi-free electrons. Therefore, after the 100% outer ionization for an intensity, the average $\overline{E}_i$ may also grow [see Fig.1(a) for $I_0 \geq 10^{17}$ W/cm$^2$] with increasing $\lambda$ since the average energy of a laser-driven free electron scales as $\approx U_p \propto E_0^2 \lambda^2/4$. Because electrons (free or bound) do not have same energy and liberated at different times, the growth of $\overline{E}_i$ is slower than the scaling of $\approx U_p$. In this regime of 100% outer ionization, absorption maxima does not show up and the absorption curve does not bend down with increasing $\lambda$. So, the survival of the absorption peak (and its redshift from the static LR wavelength $\lambda_M \approx 263$ nm) depends on the population of bound and free electrons during the interaction. As the free population of electrons increases with increasing $I_0$, absorption peak gradually shifts towards higher $\lambda$ in the band of $\Lambda_d \approx (1 - 1.5)\lambda_M$ from $\lambda_M$, and finally it disappears with the free population eventually reaching to 100%.

To understand the growth of average energy with increasing $I_0$ and dis-appearance of the absorption peak when outer ionization is 100% saturated for some higher $I_0$, we look into the kinetic energy scaling of free/quasi-free electrons which are far from the cluster. We extract the average total kinetic energy of the electrons $\overline{E}_k = \sum m_i v_i^2/2N$ for the highest intensity of $5 \times 10^{17}$ W/cm$^2$ corresponding to Fig.1 and compare with the ponderomotive energy scaling $U_p \propto E_0^2 \lambda^2/4$. Fig. 2 shows $\overline{E}_k$ for those electrons which are beyond different radii $3R_0$, $5R_0$, $10R_0$, $20R_0$ for $I_0 = 5 \times 10^{17}$ W/cm$^2$. The solid (blue) line with circle represents $U_p/5$ for different $\lambda$. It is found that the growth of $\overline{E}_k$ is $\propto U_p$, but it is slower than $U_p$ scaling as different electrons are liberated from the cluster potential at different times by experiencing different laser fields and restoring forces of background ions. Moreover, the scaling $U_p \propto E_0^2 \lambda^2/4$ is an over-estimation for a short pulsed light, i.e., shorter the pulses more is the over-estimation. Thus Fig. 2 justifies that growth of absorption beyond some intensity and wavelength (when outer ionization reaches 100% in Fig.1) is due to increasing population of free electrons.

The redshifts of absorption peaks in the case of deuteron cluster for lower intensities in the band of $\Lambda_d \approx (1 - 1.5)\lambda_M$ also resembles redshifts in the case of argon cluster in the previous work [24], which justifies red-shifting of the absorption maxima irrespective of the atom type of a cluster. However, for an argon cluster, absorption peak does not disappear due to supply of electrons via inner ionization and unsaturated outer ionization < 100% for the same laser intensity $\lesssim 5 \times 10^{17}$ W/cm$^2$ (see Ref. [24]). For deuterium cluster, in Fig.1 the red-shifted absorption peaks for $I_0 = 5 \times 10^{15}$ W/cm$^2$ and $5 \times 10^{16}$ W/cm$^2$ are located at $\Lambda_d = 320, 360$ nm respectively and the corresponding free electron’s population is 88.8%, 98.6%. With increasing $I_0$, for the deuteron cluster, average charge state is quickly saturated at $Z = 1$ in the early time of the laser pulse and remaining pulse energy helps in increasing the absorption, outer ionization of electrons (also early Coulomb explosion of cluster) and gradual increase of redshift of the absorption peak from the expected $\lambda_M \approx 263$ nm. Note that Coulomb explosion of background ions also contributes to the redshift of the absorption peak in the band of $\Lambda_d \approx (1 - 1.5)\lambda_M$ from $\lambda_M$. Therefore larger the peak intensity, larger will be the redshift in the absorption peak from the expected $\lambda_M = 263$ nm when $Z = 1$ is saturated as compared to argon cluster [24].

The occurrence of distinct maxima in the absorption and outer ionization in the red-shifted band of $\Lambda_d \approx (1 - 1.5)\lambda_M$ for lower intensities and also dis-appearance of absorption maxima after the 100% saturation of outer ionization for some higher intensities clearly shows the effect of $\lambda$ variation. Our results in Figs.1 contradicts earlier MD simulation works by Petrov et al. [21, 23] where they considered only three wavelengths $\lambda = 100, 248, 800$ nm at a fixed intensity of $I_0 = 5 \times 10^{16}$ W/cm$^2$. They could not find any maxima in the absorption while passing through Mie-resonance and nullified any role of Mie-resonance for laser absorption. They further argued that “non-uniform electron density” inside the cluster is responsible for the “null effect” of Mie-resonance [21, 23]. Firstly, we point out that, electron density is always non-uniform within the cluster and its surroundings (during the laser pulse driving; also by birth due to the random dis-

Figure 2. (Color online) MD results showing average kinetic energy $\overline{E}_k$ per electron versus $\lambda$ at $I_0 = 5 \times 10^{17}$ W/cm$^2$, corresponding to Fig. 1 (orange curve). Red, green, black, and magenta curves are the average $\overline{E}_k$ of those electrons which are beyond $3R_0$, $5R_0$, $10R_0$ and $20R_0$ respectively for $\lambda \geq 400$ nm. Blue curve represents $U_p/5$ for different $\lambda$. Thus growth of $\overline{E}_k$ and $\overline{E}_i$ (in Fig.1) are due to free electrons.
tribution of parent atoms in the position and velocity space) that does not explain the absence of Mie-resonance absorption peak. Secondly, only three well-separated wavelengths \( \lambda = 100, 248, 800 \text{ nm} \) chosen by Petrov et al. \cite{21, 22} can not resolve the absorption maxima which is evident from Fig. [1]. Thirdly, due to high intensity, resonance absorption maxima is actually red-shifted in the band of \( \Lambda_d \approx (1 - 1.5) \lambda M \) from the commonly expected \( \lambda M \) of Mie-resonance as shown in Fig. [1] and linear Mie-resonance theory is invalid here. It is delusive to look for the absorption maxima exactly at the \( \lambda M \) in presence of non-zero outer ionization at higher intensities. However, the absence of absorption maxima at the expected \( \lambda M \) does not mean that Mie-resonance has “null effect” in the absorption and outer ionization \cite{21, 23}. In steady, dynamic LR (dynamic Mie-resonance) and AHR work in unison (i.e., UDLR works) in the band of \( \Lambda_d \approx (1 - 1.5) \lambda M \) very efficiently and the near-the-LR effective field \( E_{eff} \) [as understood from the simple estimate of \( E_{eff} = E_0/(\Omega_2 - \Omega_1^2) \)] inside the cluster is so much enhanced that it forces almost 90% electrons to be outer ionized even at the near-resonance (under-dense) values of \( \lambda \approx 250 - 260 \text{ nm} \) before the \( \lambda M \approx 263 \text{ nm} \) and responsible for the resonance peak shift towards a higher \( \lambda > \lambda M \) as seen in Fig. [1]. For elaborate discussion on UDLR see Ref. \cite{24} and Sec. III C 1.

### 1. Resonance peak shift with radius variation of cluster

For a cluster, the static Mie-resonance can be met by a transition from over-dense to under-dense plasma regime (artificially) by increasing the cluster radius (mimicking cluster expansion) for a fixed number of ions/electrons at a fixed \( \lambda \), e.g., 800 nm as performed by Petrov et al. \cite{21} by a MD test simulation where “no enhancement of energy absorption” was found “near the plasmon resonance”. Contrarily, we show by our MD simulation that LR indeed plays a major role for pronounced laser absorption during the variation of cluster radius, but red-shift of absorption peak occurs depending upon laser intensity due to the UDLR in presence of outer ionization.

The deuterium cluster \( (N = 1791 \text{ atoms}) \) is irradiated by LP laser pulses of fixed \( \lambda = 800 \text{ nm} \) and \( \tau = 13.5 \text{ fs} \). At a given \( I_0 \), cluster radius \( R \) is varied from \( R = R_0 = 2.05 \text{ nm} \) to \( R = 15R_0 \) with fixed \( N \) and immobile ions. Thus plasma density is successively reduced. For \( R = 2.05 \text{ nm} \), the density is \( \rho_0 = 27.87 \rho_c \), where \( \rho_c \approx 1.75 \times 10^{21} \text{ cm}^{-3} \) and \( \omega M/\omega \approx 3.05 \). Since \( \omega M^2 \approx 1/R^3 \), the static LR condition \( \lambda M = \lambda \) (or \( \omega M = \omega \)) is achieved at \( R \approx 2.1R_0 \).

Test MD results with above parameters in Figs. 3(a)-(b) show the absorbed energy \( \mathcal{E}_t \) per atom and fractional outer ionization \( \mathcal{N} \) of electrons versus \( R/R_0 \) (top row) for different \( I_0 \) at the end of LP laser pulses. Same results are shown in Figs. 3(c)-(d) with the corresponding \( \lambda M/\lambda \) (bottom row). Our results are different than Petrov et al. \cite{21}. It is seen that maximum absorption and outer ionization occur for all three intensities but at red-shifted wavelength ratios of \( \lambda M/\lambda \) before meeting the static LR condition at \( \lambda M/\lambda \approx 1 \) (or \( R/R_0 \approx 2.1 \), vertical dashed lines). The gradual red-shifting of absorption and outer ionization maxima with increasing intensity in Fig. 3 in the marginally over-dense regime of \( 0.7 \lesssim \lambda M/\lambda \lesssim 1 \) (or \( 1.5 \lesssim R/R_0 \lesssim 2.1 \)) is very much consistent with the findings in Figs. 1(a)-(b) with varying \( \lambda \) and mobile ions.

It is known that for a over-dense cluster AHR happens \cite{27, 31} for different electrons at different times when dynamical frequency \( \Omega_i(r) \) of \( i \)-th electron at the position \( r(t) \) satisfies \( \Omega_i(r(t)) = \omega \). If plasma is too much over-dense w.r.t. \( \omega \), the absorption via AHR can not be collective and absorption peak is not expected. As we reach to the marginally over-dense regime of \( \Lambda_d \approx (1 - 1.5) \lambda M \), where \( \omega M \) and \( \omega \) become very close, many electrons may pass through AHR at the same time at ease, being excited by the near-the-LR enhanced effective field \( E_{eff} = E_0/(\omega M^2 - \omega^2) \), depending upon the laser intensity. In this case, the dynamical LR and AHR are often indistinguishable where they work together (UDLR happens) to maximize the laser absorption \cite{24}. The absorption peak in Figs. 1(a) at various intensities is the clear manifestation of collective effect of UDLR in the marginally over-dense band of \( \Lambda_d \approx (1 - 1.5) \lambda M \) dominated by the near-LR (near Mie-resonance) enhanced field effects.

### C. Absorption and outer ionization with CP vs LP light

To see the impact of laser polarization on the absorption and outer ionization; and to know how the resonance absorption maxima shifts due to it, we simulate the same deuterium
cluster with CP light (Eqn. 2.3) with similar laser parameters as in Fig. 1 of the LP case. Laser electric field components being different for LP and CP at a given instant, overall electron dynamics becomes different for a given intensity, and which may impact differently (i.e., CP may yield different results than LP) on energy absorption, outer ionization and the redshift of the resonance absorption peak.

Figure 4 shows the comparison of average total absorbed energy $\overline{E}_i$ per atom and corresponding fractional outer ionization $N$ versus $\lambda$ for the deuterium cluster ($K = 2.05$, $N = 1791$) as in Fig. 1 with the same parameters of LP (solid line) and CP (dashed line) laser pulses.

By analyzing the results in time domain, we now justify the shifting of absorption maxima from the expected static LR wavelength of $\lambda_M = 263$ nm in Fig. 4 at some intensity of $10^{16}$ W/cm$^2$ (red, dashed square line of Fig. 4) and various $\lambda$ with LP and CP light. Similar analysis for the redshift of absorption maxima was previously reported in Ref. [24] in detail for a more complicated argon cluster illuminated by LP light. Figures 5(a), 5(b) present comparison of scaled Mie-frequency $\omega_M(t)/\omega$, total absorbed energy $\overline{E}_i(t)$ per atom and fractional outer ionization $N(t) = N_{\text{out}}(t)/N(t)$ of electrons respectively with LP (solid line) and CP (dashed line) versus normalized time in units of the laser period $T$ of $\lambda = 800$ nm. The horizontal dashed line in Fig. 5(a) indicates the line of static LR condition $\omega_M = \omega$. The dynamical $\omega_M(t)$ is calculated from the relation $\omega_M(t) = Q_0(t)/R_0$, where $Q_0(t)$ is the instantaneous total positive charge defined by $Q_0(t) = N(t)Z(t)$ inside the initial cluster radius $R_0$ having $N(t)$ number of ions with average charge $Z(t)$. At $I_0 = 10^{16}$ W/cm$^2$, the only charge state $Z = 1$ of the deuterium ions for all $\lambda$ is created during $t/T = 0.9 - 1.1$ by the OFI and the corresponding dynamical $\omega_M(t)/\omega$ jumps form zero to its maximum for both LP and CP. After the respective maximum val-
The longer time spent by the dynamical \( \omega_M(t)/\omega \) near the line of LR (\( \omega_M/\omega = 1 \)) and in the marginally over-dense band of \( \Lambda_d \approx (1 - 1.5)\lambda_M \), the higher is the absorption and outer ionization due to the combined effect of AHR and dynamical LR.\(^{[24]}\), coined as unified dynamical LR (UDLR)\(^{[24]}\). In this marginally over-dense band of \( \Lambda_d = (1 - 1.5)\lambda_M \), some electrons undergo dynamical LR (due to dynamical \( \omega_M(t) \)) with great enhancement of the near-LR effective field \( E_{\text{eff}} \approx E_0/(\omega_M^2(t) - \omega^2) \)\(^{[24]}\) and some electrons undergo AHR by meeting their time dependent frequencies \( \Omega_s(t) = \omega \) in the anharmonic part of the potential at that time at ease for the close proximity of \( \Omega_s(t) \) to \( \omega_M(t) \) and the effect of near-LR enhanced field \( E_{\text{eff}} \approx E_0/(\omega_M^2(t) - \omega^2) \)\(^{[24]}\).

For the deuteron cluster also (in this work) the absorbed energy and outer ionization are enhanced (see Figs. 5(b),(c)) for those wavelengths which lie in the marginally over-dense band of \( \lambda \approx 330 \pm 67 \text{ nm} \) (e.g., 260 nm, 320 nm and 360 nm), equivalent to \( \Lambda_d \approx (1 - 1.5)\lambda_M \) as compared to the other wavelengths which lie outside this band (e.g., 200 nm and 800 nm). Among the three intermediate \( \lambda \), dynamical \( \omega_M(t)/\omega \) for \( \lambda = 260 \text{ nm} \) (magenta line) is very close to the static LR, \( \omega_M/\omega = 1 \) (where \( \lambda_M = 263 \text{ nm} \)), but the absorption and outer ionization are not still at their expected maximum. For 260 nm, the dynamical \( \omega_M(t)/\omega \) just meet the line of static LR only for a very short time during \( t/T = 0.9 - 1.2 \) and rest of the time it remains in the under-dense regime below the line of static LR (where AHR does not work), so cannot absorb energy efficiently. Instead, for \( \lambda = 320, 360 \text{ nm} \) (green, red lines), the respective dynamical \( \omega_M(t)/\omega \) continues to remain in the marginally over-dense band of \( \Lambda_d \approx (1 - 1.5)\lambda_M \) for a prolonged period up to \( t/T \approx 4, 1, 5.0 \), where both AHR and dynamical LR with near-LR enhanced effective field \( E_{\text{eff}} \approx E_0/(\omega_M^2(t) - \omega^2) \) contribute uniedly (i.e., UDLR happens) to maximize absorption and outer ionization for both LP and CP. Thus UDLR explains the red-shift of absorption maxima from the static LR wavelength of \( \lambda_M = 263 \text{ nm} \) in Fig.4 for LP and CP light. It is observed that the maximum of \( \bar{\mathcal{N}}(t) \) is little enhanced for CP in comparison to LP for those \( \lambda \) which lie in the UDLR regime of \( \Lambda_d \approx (1 - 1.5)\lambda_M \). This enhanced energy is partly due to non-zero rotating electric field vector of the CP laser, although \( \bar{\mathcal{N}}(t) \) for CP light is either less (or nearly equal) compared to LP light in Fig. 5(b). The oscillatory nature of \( \bar{\mathcal{N}}(t) \) and \( \mathcal{F}(t) \) for LP are due to the oscillating electric field vector while for CP they smoothly increase due to non-zero rotating electric field vector.

IV. RIGID SPHERE MODEL: ABSORPTION AND OUTER IONIZATION WITH CP VS LP LASER

We further justify MD results (qualitatively) by a simple rigid sphere model (RSM)\(^{[24]}\).\(^{[27]}\). Here, cluster is assumed to be pre-ionized and consists of uniformly charged spheres of ions (e.g., deuterons) and electrons of equal radii \( R_i = R_e = R_0 \). The ionic sphere is considered immobile (ions are frozen)
for the short laser pulse duration < 14-fs and laser magnetic field is also neglected for \( I_0 < 10^{14} \text{ W/cm}^2 \). The same deuterium cluster with \( N = 1791, R_0 = 2.05 \text{ nm} \) and charge density \( \rho_1 \approx 7 \times 10^{-3} \text{ a.u.} \) is considered. For \( \lambda = 800 \text{ nm} \), it represents an over-dense plasma of \( \omega_M/\omega \approx 3.05. \) For simulating multi-electron system by the RSM, we assume \( N = 1791 \) non-interacting electron spheres with their centers uniformly positioned inside the ionic sphere at the initial time. Center of each electron sphere mimics a real point size electron. We simulate the system with LP and CP laser pulses with same parameters as in Sec. III. The dynamics of each electron sphere is governed by the laser field pulse the electrostatic restoring field in the background potential of positively charged ion sphere. In the case of LP light (polarized in \( x \) ), the equation of motion (EOM) of an electron sphere can be written as \( \hat{\mathbf{r}} + \hat{\mathbf{r}}_g(r)/r = \hat{\mathbf{q}} (e/m_e) E_0(t)/R_0, \) (8) where \( \hat{\mathbf{r}} = \mathbf{x}/R_0 \) and \( r = |\mathbf{r}|. \) The potential \( \phi(r) \) and the corresponding electrostatic restoring field \( g(r) \) are respectively, \( \phi(r) = \omega_M^2 R_0^2 \left\{ \begin{array}{ll} \frac{3}{2} - r^2 / 2 & \text{if } 0 \leq r < 1 \\ 1/r & \text{if } r \geq 1 \end{array} \right. \) (9) \[ g(r) = \omega_M^2 R_0^2 \left\{ \begin{array}{ll} r & \text{if } 0 \leq r < 1 \\ 1/r^2 & \text{if } r \geq 1. \end{array} \right. \] (10) The electric field component polarized in \( x \)-direction for LP is given by Eq. 2 with \( \delta = 1. \) Each electron sphere will oscillate in the potential \( \phi(r) \) with a position dependent frequency \( \Omega[r(t)] \) \( \left( \frac{27}{23} \right). \) To study the dynamics of electron spheres in the potential \( \phi(r) \) with CP light we write EOM in two dimension as \( \left( \frac{23}{27} \right). \) \[ \hat{\mathbf{r}} + \hat{\mathbf{r}}_g(r)/r = \hat{\mathbf{q}} (e/m_e) E_0^2(t)/R_0, \] (11) \[ \hat{\mathbf{r}} + \hat{\mathbf{r}}_g(r)/r = \hat{\mathbf{q}} (e/m_e) E_0^2(t)/R_0 \] (12) Here \( \hat{\mathbf{r}}_g = \mathbf{x}/R_0, \hat{\mathbf{r}}_g = \mathbf{y}/R_0, r = \sqrt{r_x^2 + r_y^2}. \) The electric field components for CP light are given by Eqns. 2 and 3 with \( \delta = 1/\sqrt{2}. \) Similar to MD study in sec. III C here also we have same \( U_p = E_0^2/4\omega^2 \) as in the LP case. Each electron sphere moves spirally in the potential \( \phi(r) \) driven by CP light.

Since initial positions of electron spheres are different, they experience different restoring forces. As laser drives those electron spheres, they will climb up in the potential and some of them will leave the potential at different instant of time experiencing different total fields. Inside the potential their motion is harmonic with a constant frequency \( \Omega[r(t)] = \omega_M. \) As soon as they cross the cluster boundary \( (r > 1) \) their motion becomes anharmonic with a gradual decrease of \( \Omega[r(t)] \) \( \left( \frac{27}{31} \right). \) When \( \Omega[r(t)] = \omega \) is met for an electron, AHR happens, and the electron leaves the cluster \( \left( \frac{27}{31} \right) \) by absorbing energy. Thus, RSM can be used to understand both LR and/or AHR processes depending upon the laser frequency and intensity.

Figures 6(a)-(b) show comparison of average total absorbed energy \( \bar{\mathcal{E}} = \sum N (m_i v_i^2 + q_i \Phi_0)/N \) per electron sphere for LP and CP laser pulses (after \( \tau = 13.5 \text{ fs} \)) versus \( \lambda \) for different \( I_0. \) Figure 6(c)-(d) are the corresponding fraction of outer ionized electrons \( \bar{\mathcal{N}} = N_{out}/N. \) Results are separated in two regimes of intensities for more clarity. Figures 6(a),(c) are for \( I_0 = 10^{17} \text{ W/cm}^2 \) and Figs. 6(b),(d) are for \( I_0 = 5 \times 10^{17} \text{ W/cm}^2. \) It is noted that RSM results (for both LP and CP) resemble the MD results in Fig. 4(i) growth of absorption and outer ionization with increasing \( \lambda \) for \( I_0 \lesssim 10^{17} \text{ W/cm}^2 \) with distinct maxima located in the band of \( \Lambda_M \approx (1 - 1.5) \lambda_M, \) i.e., in between 263 - 400 nm in Figs. 6(a),(c); (ii) increasing redshift of the absorption and outer ionization maxima from the static LR wavelength of \( \lambda_M \approx 263 \text{ nm} \) (vertical dashed line) with increasing \( I_0 \lesssim 10^{17} \text{ W/cm}^2 \) due to increasing outer ionization; and (iii) gradual disappearance of absorption maxima (followed by even a growth of \( \bar{\mathcal{E}} \), for \( \lambda > 400 \text{ nm} \)) due to 100% saturation of \( \bar{\mathcal{N}} \) when \( I_0 \) exceeds \( 10^{17} \text{ W/cm}^2 \) in Figs. 6(b),(d).

Similar to the MD results in Fig. 4 the magnitude of the maximum value of \( \bar{\mathcal{E}} \) and \( \bar{\mathcal{N}} \) are more for CP than LP in the marginally overdense band of \( \Lambda_M \approx (1 - 1.5) \lambda_M. \) The maxima of \( \bar{\mathcal{E}} \) for LP (CP) are 19.1, 31.9, 54.9, and 64.5 a.u. \((40.3, 49.4, 62.4, \text{ and } 72.8 \text{ a.u})\) for \( I_0 = 5 \times 10^{15}, 10^{16}, 5 \times 10^{16}, 10^{17} \text{ W/cm}^2 \) respectively and corresponding \( \bar{\mathcal{N}} \) are 33%, 57%, 89%, and 94% \((74, 90, 98, \text{ and } 99%)\) for respective \( I_0 \) in Figs. 6(a),(c). Also, after the maxima, for lower intensities, both \( \bar{\mathcal{E}} \) and \( \bar{\mathcal{N}} \) with CP light drop below the values with LP light which, on the other hand, gradually reverses at higher intensities \((i.e., \text{CP dominates LP as in Figs. 6(b),(d))}, \) as \( \lambda \) is increased. At the higher intensity \( 5 \times 10^{17} \text{ W/cm}^2, \) after \( \lambda \approx 320 \text{ nm} \), \( \bar{\mathcal{N}} \) is saturated at 100% for CP and almost 98.5% for LP but \( \bar{\mathcal{E}} \) gradually grows with increasing \( \lambda > 320 \text{ nm} \) for both CP and LP. For higher intensities \( \approx 5 \times 10^{17} \text{ W/cm}^2, \) when 100% outer ionization is reached (almost all electrons are freed) in the early cycles of a laser pulse due to efficient UDLR, the average absorbed energy \( \bar{\mathcal{E}} \) per electron sphere may grow \((\text{as for MD in Fig. 4 at the intensity } 5 \times 10^{17} \text{ W/cm}^2)\) with increasing \( \lambda \) due to the dominant variation of free electron kinetic energy as \( U_p \approx E_0^2 \lambda^2/4. \) To support this again for the RSM, we look at the free electrons kinetic energy scaling similar to the MD case presented in Fig. 2. Inset Fig. 6(c) show the comparison of the average kinetic energy \( \bar{\mathcal{E}} \) per electron sphere versus higher \( \lambda \geq 500 \text{ nm} \) for both LP (solid-squared orange line) and CP (dashed-squared orange line) with \( U_p \approx E_0^2 \lambda^2/4 \) (dashed-dotted blue line) at \( 5 \times 10^{17} \text{ W/cm}^2 \) corresponding to Fig. 6(b). One can see the growth of \( \bar{\mathcal{E}} \approx U_p, \) but it is slower than the scaling of \( U_p \) for both LP and CP since the electrons (free or bound) do not have same kinetic energy and liberated at different times of the laser cycle. Also, growth \( \bar{\mathcal{E}} \) is more close to \( U_p \) for CP than LP due to more number outer electrons with higher kinetic energies at different \( \lambda \) for CP than LP. This justifies the growth of \( \bar{\mathcal{E}} \) with \( \lambda \) at a higher \( I_0 = 5 \times 10^{17} \text{ W/cm}^2 \) for CP and LP, and also explains why \( \bar{\mathcal{E}} \) for CP dominates LP as in Fig. 4 for \( \lambda \) beyond the band of \( \Lambda_M \approx (1 - 1.5) \lambda_M. \)

Thus RSM results (in Fig. 6) bring out most of the features of MD results (in Fig. 4) which justify the dependence of redshift of absorption maxima on laser polarization and intensity and also disappearance of absorption maxima followed by a growth of absorption at higher intensity of \( 5 \times 10^{17} \text{ W/cm}^2 \) for CP and LP, with increasing \( \lambda \).
V. SUMMARY AND CONCLUSION

We demonstrate the effect of laser polarization on the redshift of the resonance absorption peak for a deuterium cluster irradiated by short 5-fs (fwhm) laser pulses using MD simulation and supported by RSM analysis. For both the polarization cases (LP and CP), we show that for a given intensity $<10^{17}$ W/cm$^2$ the optimized wavelength for maximum laser absorption in deuterium cluster lie in the band of wavelengths $\lambda \approx 330 \pm 67$ nm in stead of the commonly expected static LR (Mie-resonance) wavelength of $\lambda_M = 263$ nm. MD simulation and the RSM show gradual red-shift of the absorption maxima towards higher wavelengths in the marginally over-dense band of $\Lambda_d \approx (1 - 1.5)\lambda_M$ from $\lambda_M$ of static LR with increasing laser intensity; and for higher intensities $>10^{17}$ W/cm$^2$ absorption peak disappears as outer ionization saturates at 100% for both LP and CP. This disappearance of the resonance absorption peak should not be misinterpreted as the negligible (or no) role of Mie-resonance. In fact, in this marginally over-dense band of $\Lambda_d \approx (1 - 1.5)\lambda_M$, both AHR and dynamical LR with near-LR enhanced effective field $E_{eff} = E_0/\left(\omega_M^2(t) - \omega^2\right)$ contribute in unison very efficiently – UDLR happens – to maximize absorption and outer ionization for both LP and CP. It is also found that before the absorption peak, laser absorption due to LP and CP lasers are almost equally efficient (CP case being inapprecia-
of the states of laser polarization for intensities absorption and outer ionization are almost same irrespective we conclude that laser marginal differences for LP and CP cases, which may not inappreciably dominates absorption due to CP with increasing λ which gradually reverses at higher intensities. Neglecting marginal differences for LP and CP cases, which may not

\[ \lambda \approx (1-1.5)\lambda_M \]  

in presence of non-zero outer ionization at higher intensities since linear Mie-resonance theory is invalid here. Our future work will report the pulse length dependence of the red-shift of the absorption peak.

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