Anharmonic resonance absorption of short laser pulses in clusters: A molecular dynamics simulation study

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Abstract

Linear resonance (LR) absorption of an intense 800 nm laser light in a nano-cluster requires a long laser pulse > 100 fs when Mie-plasma frequency (ω_M) of electrons in the expanding cluster matches the laser frequency (ω). For a short duration of the pulse the condition for LR is not satisfied. In this case, it was shown by a model and particle-in-cell (PIC) simulations [Phys. Rev. Lett. 96, 123401 (2006)] that electrons absorb laser energy by anharmonic resonance (AHR) when the position-dependent frequency ω(r(t)) of an electron in the self-consistent anharmonic potential of the cluster satisfies ω(r(t)) = ω. However, AHR remains to be a debate and still obscure in multi-particle plasma simulations. Here, we identify AHR mechanism in a laser driven cluster using molecular dynamics (MD) simulations. By analyzing the trajectory of each MD electron and extracting its ω(r(t)) in the self-generated anharmonic plasma potential it is found that electron is outer ionized only when AHR is met. An anharmonic oscillator model, introduced here, brings out most of the features of MD electrons while passing the AHR. Thus, we not only bridge the gap between PIC simulations, analytical models and MD calculations for the first time but also unequivocally prove that AHR processes is a universal dominant collisionless mechanism of absorption in the short pulse regime or in the early time of longer pulses in clusters.

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I. INTRODUCTION

Laser-driven atomic clusters absorb large fraction of laser energy compared to traditional solid and gas targets. Solid like overdense plasma density of a cluster and its smaller size (of a few nanometer) than the wavelength of 800 nm laser pulse (typically used in experiments) allow full penetration of laser field without its attenuation, contrary to micron-sized solids, forbidden by the laser field alone. Higher ionic charge states are liberated from the target or directed into the target. It can is discriminated here by restricting the laser intensity below 10^{17} W cm^{-2} where B field of the laser is negligible. One may surmise the “Brunel effect” or the “vacuum heating” as a probable collisionless process in the early time of interaction when plasma boundary is sharp [27]. Firstly, the “vacuum” as mentioned by Brunel may not be a real vacuum. Electrostatic field exists in the target vicinity (as we shall show here) that plays a crucial role for an electron’s dynamics before it is liberated from the target or directed into the target. It can not gain a net energy, unless there is any nonlinear interaction through the nonlinear space-charge field within the target and/or in the target vicinity. Therefore, the tautological name

The role of \( \mathbf{\nabla} \times \mathbf{B} \) heating as a collisionless mechanism is discriminated here by restricting the laser intensity below 10^{17} W cm^{-2} where B field of the laser is negligible. One may surmise the “Brunel effect” or the “vacuum heating” as a probable collisionless process in the early time of interaction when plasma boundary is sharp [27]. Firstly, the “vacuum” as mentioned by Brunel may not be a real vacuum. Electrostatic field exists in the target vicinity (as we shall show here) that plays a crucial role for an electron’s dynamics before it is liberated from the target or directed into the target. It can not gain a net energy, unless there is any nonlinear interaction through the nonlinear space-charge field within the target and/or in the target vicinity. Therefore, the tautological name
“vacuum heating” is improper. According to Brunel’s original proposition [27], when an intense laser pulse strikes a sharply bounded overdense plasma; electrons are dragged into the vacuum and then due to the laser field reversal, in the next half-cycle of the pulse, electrons are pushed back inside the target with a velocity on the order of the ponderomotive velocity \( v_0 = E_0/m_0 \). The crucial assumption in this model is that electrons experience no net field while they return to the target. As a consequence Brunel’s electron flow becomes laminar, meaning that their trajectories do not cross each other within a laser period irrespective of the laser intensity. We point out that, since different electrons originate from different parts of the target they experience different electrostatic fields and originate with different initial phases. When driven by a laser, their trajectory crossing is unavoidable at a later time. Detail analysis showing deficiency in Brunel’s “vacuum heating” is given in Ref. [28]. Brunel electrons, upon returning to the target, experience a field free region due to complete cancellation of induced electrostatic field by the laser field. Thus, the velocities acquired during their traversal in the vacuum are fully retained, they do not have chance to give energy back (even partly) to the electromagnetic field. In the context of laser-cluster interaction, induced electrostatic fields can not be fully compensated by the laser field (induced field may exceed the laser field) and cluster interior is rarely field free (as shown in this work) during the laser interaction. Otherwise, ionization ignition [21] can not happen and higher charge states [29,32] of ions can not be created. In this sense, Brunel effect is incomplete, warrant a re-look into the problem and search for an appropriate mechanism behind the laser absorption.

On the other hand, let us suppose that there is an anharmonic potential created at the target front (or in the target interior) due to the laser interaction. Such a potential is inevitably formed (for any finite size target) at the ion-vacuum boundary (where laser interacts first) due to \( \sim 1/r \) fall of the potential which may be asymmetric. Anharmonicity in the potential also appears due to local charge non-uniformity (via ionization, concentrated electron cloud etc.) in a laser driven plasma. The frequency \( \Omega \) of an electron in such a potential is dependent on its position \( r \). When driven by a laser field, its \( r \) changes with time which makes \( \Omega(r) \) time dependent, i.e., \( \Omega(r(t)) \). An initially bound electron, starting from some location in the overdense plasma potential, while becoming free must experience the \( \sim 1/r \) Coulomb tail of the potential and the corresponding \( \Omega(r(t)) \) of the electron must meet \( \omega \) while trying to come out of the potential. This dynamical resonance - the anharmonic resonance (AHR) - occurring in an anharmonic potential was studied before using a model and three dimensional PIC simulations of laser driven clusters [33]. However, collisionless processes and AHR phenomenon remain to be a debate [28,34,35]. In numerical simulations it is often obscured due to many body nature of interaction, since it needs clear examination of individual electron trajectory, identification of corresponding \( \Omega(r(t)) \) and a dynamical mapping of \( \Omega(r(t)) \) on to \( \omega \). To prove AHR for a laser driven cluster a three dimensional MD simulation code with soft-core Coulomb interactions among charge particles has been developed. By following the trajectory of each MD electron and identifying its time-dependent frequency \( \Omega[r(t)] \) in the self-generated anharmonic plasma potential it is found that electron leaves the potential and becomes free only when AHR condition \( \Omega[r(t)] = \omega \) is met. Thus, for the first time, our MD simulation clearly identifies AHR process in the laser cluster interaction. We further introduce a non-linear oscillator model that brings out most of the features of MD electrons while passing the AHR. Thus, we bridge the gap between PIC simulations, analytical models and MD calculations.

Atomic units (i.e., \( m_e = -e = 1, 4\pi\varepsilon_0 = 1, \hbar = 1 \)) are used throughout this work unless specified explicitly. We consider a single deuteronium cluster of radius \( R = 2.05 \text{ nm} \), Wigner-Seitz radius \( r_w = R/N^{1/3} \approx 0.17 \text{ nm} \) and number of atoms \( N = 1791 \). It is irradiated by 800 nm wavelength laser pulses of various intensity giving density \( \rho \approx 27.3 \rho_w \) and \( \omega_p^2/3\omega^2 = \omega_{\text{Seitz}}^2/\omega^2 \approx 9.24 \); where \( \rho_w = \omega_p^2/4\pi \) is the critical density at 800 nm and \( \omega_p \) is the plasma frequency. These parameters of the cluster are kept unchanged throughout this work.

Section [II] illustrates AHR by a simple model of a cluster while Sec.[III] proves the hypothesis of AHR by detailed MD simulations. Summary and conclusion are given in Sec.[IV].

### II. MODEL FOR ANHARMONIC RESONANCE ABSORPTION

Before studying the laser-cluster interaction by MD simulations, we show here various features of AHR by a model of a cluster which will provide an easy interpretation of MD results in Sec.[III]. In the model, cluster is assumed to be pre-ionized and consists of homogeneously charged spheres of massive ions and much lighter electrons of equal radii \( R_i = R_o = R \). When their centers coincide plasma becomes charge neutral. The motion of ions can be neglected for short laser pulses < 50 fs and non-relativistic laser intensities < \( 10^{15} \text{ Wcm}^{-2} \) as considered in this work. Thus ion sphere provides a sharp boundary with zero density gradient scale-length at the vacuum plasma boundary.

The equation of motion (EOM) of the electron sphere in a linearly polarized laser field along \( x \)-direction reads

\[
\frac{d^2 \vec{r}}{dt^2} + \frac{\vec{r}}{r} g(r) = \ddot{x}(q_e/m_e)E_i(t)
\]

where \( \vec{r} = \ddot{x}/R \) and \( r = |\vec{r}| \). The electrostatic restoring field

\[
g(r) = \omega_m^2 R \begin{cases} r & \text{if } 0 \leq r \leq 1 \\ 1/r^2 & \text{if } r \geq 1 \end{cases}
\]

can be derived by Gauss’s law. It shows that as long as the excursion \( r \) of the center of the electron sphere remains inside the ion sphere, it experience a harmonic oscillation with a constant eigen-frequency \( \omega_m \). Crossing the boundary of the ion sphere, it begins to experience the Coulomb force and its motion becomes anharmonic with gradual reduction in the eigen-frequency for increasing excursion from the center of the ion sphere. The nonlinear restoring field \( g(r) \) is simpler than used earlier [35,37]. Nevertheless, it exhibits all features
AHR phenomena elegantly, e.g., prompt generation of electrons within a time much shorter than a laser period, crossing of electron trajectories and subsequent non-laminar electron flow \cite{35}. It neglects the interaction of diffuse boundary of the electron sphere with the sharp boundary of the ion sphere, but allows us to calculate \( \Omega [r] \) of the electron sphere analytically for an arbitrary excursion which is not possible with the \( g (r) \) in Refs. \cite{36, 37}.

The cluster size being much smaller than the laser wavelength \( \lambda = 800 \) nm as considered in this work, the dipole approximation for the laser vector potential \( A (z, t) = A (t) \exp (-i 2 \pi \lambda/\lambda) \approx A (t) \) is assumed. Thus, the effect of propagation of light (directed in \( z \)) is disregarded. We take \( A (t) = (E_0 / \omega) \sin \left( \omega t / 2n \right) \cos (\omega t) \) for \( 0 < t < nT \); where \( n \) is the number of laser period \( T \), \( nT \) is the total pulse duration and \( E_0 \) is the field strength corresponding to an intensity \( I_0 = E_0^2 / 2 \). The driving field \( E_i (t) = -dA / dt \) reads

\[
E_i (t) = \frac{(E_0 / \omega)}{3} \sum_{i=1}^{3} c_i \omega_i \sin (\omega_i t) \quad \text{if} \quad 0 < t < nT \quad (3)
\]

\[
E_i (t) = 0 \quad \text{otherwise};
\]

where \( c_1 = 1/2, c_2 = c_3 = -1/4, \omega_1 = \omega, \omega_2 = (1 + 1/n) \omega, \) and \( \omega_3 = (1 - 1/n) \omega \). Eq. (3) leads to the correct dynamics of a free electron \cite{38} even for very short sub-cycle pulses in contrast to the often used \( \sin^2 \)-pulses \cite{39, 40, 41}.

### A. Effective frequency of the electron sphere

The electrostatic potential corresponding to Eq. (2) reads

\[
\phi (r) = \frac{\omega_M^2}{2} R^2 \times \left\{ \begin{array}{ll}
3/2 - r^2 / 2 & \text{if} \quad 0 \leq r \leq 1 \\
1 / r & \text{if} \quad r \geq 1.
\end{array} \right.
\]

In the absence of a driver, the eigen-period \( T \) of oscillation of the electron sphere in the potential \cite{42} can be calculated from

\[
T = \frac{4 R}{\sqrt{2}} \int_{0}^{r_m} \frac{dr}{\sqrt{\Phi (r_m) - \Phi (r)}}.
\]

Here \( \Phi (r_m) = q_e \phi (r_m) \) is the potential energy stored in the oscillator at an initial distance \( r = r_m \) from where it is left freely in the potential at a time \( t = 0 \). For \( 0 \leq r_m \leq 1 \), Eq. (5) yields a constant \( T = 2 \pi / \omega_M \) and the effective frequency of oscillation of the electron sphere as

\[
\Omega (r) = 2 \pi / T = \omega_M.
\]

When \( r_m > 1 \), we write \( T = T_1 + T_2 \) with \( T_1 / 4 \) as the time required for \( r = 1 \) to \( r = 0 \) and \( T_2 / 4 = \frac{4}{\omega_M} \int_{r_m}^{1} \frac{dr}{\sqrt{\Phi (r_m) - \Phi (r)}} \) is the time required for \( r = r_m \) to \( r = 1 \) which give

\[
T_1 = \frac{4}{\omega_M} \sin^{-1} \left( \sqrt{\frac{r_m}{3r_m - 2}} \right),
\]

\[
T_2 = \frac{2 (r_m)^{3/2}}{\omega_M} \left[ \sin^{-1} \left( \sqrt{\frac{r_m - 1}{r_m}} \right) + \sqrt{\frac{r_m - 1}{r_m}} \right].
\]

Figure 1. Normalized effective frequency \( \Omega / \omega \) of the electron sphere versus its excursion amplitude \( r_m \) for a deuterium cluster of radius \( R = 2.05 \) nm, Wigner-seitz radius \( r_0 \approx 0.17 \) nm and number of atoms \( N = 1791 \), density \( \rho \approx 27.3 \rho_c \) and \( \omega_M^2 / \omega^2 \approx 9.24 \); where \( \rho_c = \omega^2 / 4 \pi \) is the critical density at \( \lambda = 800 \) nm. Numerical approximation using Eq. (8) and the analytical result using Eqns. (6)-(7) are comparable. Vertical dashed lines indicate AHR is expected near \( r_m \approx 2 \) according to Eq. (8) and \( r_m \approx 2.5 \) according to Eq. (7).

\( T_1 \) is obtained from the harmonic solution \( r_m = \sqrt{R^2 + v_g^2 / \omega_M^2 \sin (\omega_M (t - T_2 / 4 - \omega_M R / v_R))} \) inside the cluster satisfied by the electron that enters the surface of the cluster with the velocity \( v_g = - \omega_M R \sqrt{2/3} (r_m - 1) / r_m \) (obtained from the energy conservation) at \( t = T_2 / 4 \). If the electron starts at the surface of the cluster, i.e., at \( r_m = 1 \), we get \( T_1 = 2 \pi / \omega_M \), \( T_2 = 0 \) and recover Eq. (4) with \( T = T_1 \). The effective frequency \( \Omega (r) = 2 \pi / (T_1 + T_2) \) now depends on the excursion amplitude \( r = r_m \), since the electron sphere interacts with the nonlinear part of the restoring field. In a laser field excursion changes with time. Thus \( \Omega (r) \) depends on \( t \) when \( r (t) > 1 \). In more realistic MD simulations of clusters, there is no pre-defined potential (as Eq. (4)) in which electrons oscillate. Therefore, finding \( \Omega (r) \) analytically is not possible in MD. From Eq. (1) we formally write (in analogy with a harmonic oscillator) the square of \( \Omega (r) \) as the ratio of restoring field to the excursion of the electron sphere \cite{33}.

\[
\Omega^2 (r) = \frac{g (r)}{r (r)}.
\]

Note that for a harmonic oscillator above relation yields the correct eigen frequency \( \Omega_d \) with \( g (r) = \Omega_d^2 r \).

Analytical result of the normalized effective frequency \( \Omega / \omega \) using (Eqns. (6)-(7)) as a function of excursion \( r_m \) of the electron sphere in the potential \cite{42} is plotted in Fig. 1. Numerical solution of Eq. (4) (without the laser field) gives \( r (t) \) and corresponding \( \Omega / \omega \) from Eq. (8). This numerical approximation is also plotted in Fig. 1 which matches reasonably well with the analytical \( \Omega / \omega \). As the electron sphere moves away from harmonic region of the potential, \( \Omega / \omega \) starts decreasing. At a distance of \( r_m \approx 2 \), the AHR condition \( \Omega / \omega \approx 1 \) is satisfied. If the laser field is strong enough to bring the elec-
tron sphere at a value of $r(t) = r_m \approx 2$, the electron sphere may gain significant energy from the laser field via such AHR.

**B. Dynamics of the electron sphere in the laser field**

The dynamical behaviour of the electron sphere (for the above cluster) irradiated by a $n = 5$-cycle pulse (3) of duration $nT = 13.5$ fs and peak intensity $5 \times 10^{15}$ W/cm$^2$ is now studied. Figure 2(a-d) depicts the normalized value of the square of the frequency $\Omega^2(r)$, excursion $x$, total energy $E_t$, and the laser field $E_l$ versus the normalized time $t = t/T$ for four electron spheres undergoing AHR at successive times $a) t = 2.2$, $b) t = 2.7$, $c) t = 3.2$, $d) t = 3.7$. The deuterium cluster of Fig.1 is irradiated by a $n = 5$-cycle pulse of peak intensity $5 \times 10^{15}$ W/cm$^2$.

The increasing laser field towards its peak value (i.e., $E_l \sim 1$) after $t/T \approx 2.0$ helps $|x|$ to exceed unity with a fast drop of $\Omega^2$ for $t/T > 2.1$. Around $t/T \approx 2.2$ (indicated by vertical dashed line), $\Omega^2$ meets the AHR condition with $|x| \approx 2$. The fact that AHR truly occurs at an excursion $|x| \approx 2$ is in agreement with Fig.1. It also justifies the robustness of the formal approximation (Eq.8) for retrieving the effective frequency from the numerical model. After the AHR (i.e., $t/T > 2.2$) particle becomes completely free with $\Omega^2[r(t)] \approx 0$ and final energy $E_t > 0$.

Figures 2(c-d) show electron spheres undergoing AHR at $t/T = 2.7, 3.2, 3.7$ respectively. Since initial positions are different, they have different initial phases and experience different restoring fields and total fields even though same laser field acts on them. As a result they are emitted at different times from the potential experiencing the AHR with different laser field strengths. Electron spheres experiencing AHR at a higher driving field strength generally acquire higher energies after the pulse as in Figs.2(a-c). Some electrons may also exhibit [as in Fig.2(d)] multiple AHR: they leave the cluster potential through AHR, return to the cluster interior by the laser field (or by the stronger restoring field than the laser field) and finally become free with a net positive energy via the AHR.

In this model electrons are frozen into a single sphere. Whereas, in reality, electrons in a cluster distinctly move even with a low intensity short laser pulse. Such a multi-electron system with all possible electrostatic interactions will be con-
sidered in detail by MD simulations in Sec III. To gain some more physical insight on the dynamics of electrons in a multi-electron cluster and the AHR through the above model, we consider \( N = 1791 \) non-interacting electron spheres (mimicking the multi-electron system) placed uniformly inside the ion sphere. Each electron sphere mimics a real point size electron.

Figures [3]a-d show snapshots of all non-interacting electron spheres (each dot represents a sphere) in the energy versus effective frequency plane at times \( t = 2.2, 2.7, 3.2, 3.7 \) corresponding to Figs [2]a-d. Colors indicate their normalized positions.

In an early time \( t/T = 2.2 \) [in Fig 3(a)] a large fraction of electrons are bound in the harmonic part of the potential with frequency \( \bar{\omega}^2 = \omega_0^2/\omega^2 = 9.24 \) and excursion \( r < 1 \) (dark blue to light blue). Some electrons first come out of the harmonic part and continue in the anharmonic part of the potential with a drop in \( \bar{\Omega}^2 \) (up to 4) and increasing excursion \( r \approx 2 \) and frequency \( \bar{\Omega} \approx 1.1 \). At a later time \( t/T = 2.7 \) [in Figs 3(b)], phase of the laser field is reversed with nearly the same strength. Beyond \( t/T = 2.5 \) [in Figs 3(b-d)] laser field strength becomes weaker than the restoring field on some quasi-free electrons. Those electrons (typically having low energies) are dragged inside the cluster (see their color changes from brown to yellow) even though they were made free via the AHR earlier.

Thus a simple nonlinear oscillator model brings out most of the physics of AHR phenomena for a laser driven cluster in the temporal domain (Fig 2) as well as in the energy versus frequency domain (Fig 3). The identification of the effective dynamical frequency \( \Omega[r(t)] \) of the driven oscillator in the numerical model and the liberation of particles from the cluster potential only when \( \Omega \) matches the resonance condition \( \Omega \approx \omega \), clearly justifies the robustness of the formal approximation in Eq. (8) and permits its application in the self-consistent MD simulations in Sec III.

### III. ANHARMONIC RESONANCE ABSORPTION USING MOLECULAR DYNAMICS SIMULATION

It is mentioned that in the above model electrons are frozen in a sphere which moves in a predefined attractive potential due to the ion sphere. In reality, the potential of the ionized cluster varies with the time to time redistribution of charges. In the initial time, when cluster is charge neutral, potential must start from a zero value. Electrons may also face repulsive potential due to concentrated electron cloud in some part of the cluster.

In the model, the electron sphere either stays inside (0% outer ionization) the cluster or completely goes out of the cluster (100% outer ionization) for a given laser intensity. However, there is always a certain fraction of outer ionized electrons even at an intensity just above the inner ionization threshold. These shortcomings of the model can be addressed by MD simulation.

#### A. Details of molecular dynamics simulation

A three-dimensional MD simulation code is developed to study the interaction of laser light with cluster. Particular attention is given to the identification of AHR process using MD simulation. Cluster is assumed to be pre-ionized. This may be regarded as a situation where ionization has already taken place by a pump pulse and subsequent interaction of a probe pulse is studied. Electron-electron, ion-electron and ion-ion interactions through the Coulomb field are taken into account. Binary collisions among particles are neglected since we are interested in collisionless processes. The EOM of \( i \)-th particle in a laser field polarized in \( x \) and propagating in \( z \) (in the dipole approximation) reads

\[
m_i \frac{dv_i}{dt} = \vec{F}_i(r_i, v_i, t) + \vec{q}_iE_i(t) + q_i\vec{v}_i \times \vec{B}_i(t),
\]

where \( \vec{F}_i = \sum_{j \neq i} \vec{q}_j/r_{ij}^3 \) is the Coulomb force on \( i \)-th particle of charge \( q_i \) due to all \( j \)-th particles each of charge \( q_j \) in the system. \( E_i(t) \) and \( B_i(t) \) are the electric and magnetic part of the laser field. Usually, \( B_i(t) \approx E_i(t)/c \ll 1 \) for intensities \( < 10^{18} \text{ W cm}^{-2} \). To avoid steep increase in the Coulomb force \( \vec{F}_i \), for a small separation \( r_{ij} \rightarrow 0 \), a smoothing parameter \( r_0 \) is added with \( r_{ij} \). The modified Coulomb force on \( i \)-th particle and the corresponding potential at its location are

\[
\vec{F}_i = \sum_{j \neq i} \frac{\vec{q}_j r_{ij}}{(r_{ij}^2 + r_0^2)^{3/2}}, \quad \Phi_i = \sum_{j \neq i} \frac{q_j}{(r_{ij}^2 + r_0^2)^{1/2}}.
\]

This modification of the force allows a charge particle to pass through another charge particle in the same way as in the PIC simulation. Thus it helps to study collisionless energy absorption processes in plasmas, e.g., resonances.

Equation (9) is solved using the velocity verlet time integration scheme [42] with a uniform time step \( \Delta t = 0.1 \) a.u.. The code is validated by verifying the energy conservation of the system as well as identifying the electron plasma oscillation with desired Mie-plasma frequency \( \omega_0 \) for the spherical cluster plasma. Although there are plenty of MD simulations for clusters [2] [30] [41] [43] [58], verification of this natural oscillation through MD simulation is rarely reported which is extremely important, particularly to study frequency dependent phenomena, e.g., anharmonic and harmonic resonance absorption in laser driven plasmas. Otherwise, resonance physics may be missing in simulations and subsequent MD results could be misleading. In fact, MD codes in Refs. [47] [50] could not find the signature of resonances in laser cluster interaction and the reason was unknown; while
experiments [25] [59–61], theory and particle-in-cell simulation [33] [36] [39] [40] [42–64] studies clearly indicated its importance.

We note that the artificial free parameter $r_0$ in most of the earlier works has been chosen by considering only the energy conservation point of view in the simulation. In Refs. [43–45], $r_0 = 0.02$ nm for electron-electron interaction and $r_0 = 0.1$ nm for electron-ion interaction have been taken which do not violate the energy conservation in the case of xenon cluster. In Ref. [46], $r_0 = 0.15$ nm for argon and $r_0 = 0.12$ nm for xenon cluster were chosen such that the minimum of the electron-ion interaction potential agrees with the ionization potential of the neutral atom. MD codes in Refs. [47–53] have reported similar kind of $r_0$ values as in Refs. [43–45]. Some of the authors have also chosen large $r_0$ on the order of cluster radius [2] [54] and energy conservation is still obeyed.

In our simulation, frequency of oscillation ($\omega_M$) of electrons is found to be sensitive on the value of the $r_0$ while conservation of energy is obeyed even for larger values of $r_0$. But energy conservation alone can not grant correctness of particle dynamics in simulations. To get correct oscillation frequency ($\omega_M$) one can not choose $r_0$ arbitrarily and a law has to be enforced. For a very small separation $r_{ij} \ll r_0$, the space charge field on the $i$-th particle $\vec{E}_{i}^{sc} = \vec{F}_{i}/q_i$ has to be linear in $r_{ij}$ and its slope has to be $\omega^2_M$; i.e., $\vec{E}_{i}^{sc} = \vec{F}_{i}/q_i = \omega^2_M \vec{r}_{ij}$ in order to get correct plasma oscillations. From Eq. [10], we find that $\vec{E}_{i}^{sc} = \vec{F}_{i}/q_i \approx \sum_{j} (q_j/r_{ij}) \vec{r}_{ij} \approx (Q_0/r_0^3) \vec{r}_{ij}$ for $r_{ij} \ll r_0$; assuming $Q_0$ is the total uniformly distributed charge of type $j$ inside the sphere of radius $r_0$ where all $r_{ij}$ are nearly same for collective plasma oscillations. From the above two expressions of space-charge field $\vec{E}_{i}^{sc} = \omega^2_M \vec{r}_{ij}$ and $\vec{E}_{i}^{sc} \approx (Q_0/r_0^3) \vec{r}_{ij}$ we get $\omega^2_M = Q_0/r_0^3$. For uniform ion charge density $\rho$ we write $\rho = Q/(4\pi/3)^3 = Q_0/(4\pi/3)r_0^3$ which gives $Q_0/r_0^3 = Q_0/r_0^3 = 4\pi\rho/3 = \omega^2_M$. Thus we get $Q_0/r_0^3 = N_0Z/r_0^3 = Q/r_0^3 = NZ/R_0^3$ and $r_0 = R(N_0/N)^{1/3}$; where $Z$ is the uniform charge state of ions in the cluster, $N_0$, $N$ are the number of ions inside the sphere of radius $r_0$ and $R$, $Q_0 = N_0Z, Q = NZ$ are the total ionic charge in the sphere of radius $r_0$ and in the cluster respectively. At this point $N_0$ remains arbitrary. We note that $r_0$ should be as small as possible, but non-zero. For a non-zero ionic charge density there should be at least one ion (to provide the restoring force to an electron) in the sphere of radius $r_0$. Therefore, setting $N_0 = 1$, we find that $r_0 = R/N^{1/3}$ is the most legitimate choice [55] which is equal to the well known Wigner-Seitz radius $r_w = R/N^{1/3}$ for a given cluster that leads to correct Mie-plasma oscillation if the law of force is of Coulombic in nature.

To prove that our MD code is capable of producing oscillation of electrons at the Mie-plasma frequency in the absence of a laser field, the deuterium cluster of radius $R = 2.05$ nm, and number of atoms $N = 1791$ (as in Sec II) is considered. To make homogeneously charged positive background in which electrons will oscillate, all $N_0$ ions ($N_0 = N = 1791$) are uniformly distributed initially. It gives ionic charge density $\rho_0 = 0.007$ a.u. and $\omega_M = \sqrt{4\pi\rho_0/3} = 0.1735$ a.u.. A fewer number of $N_e$ electrons (forming a homogeneous sphere of radius $R/2$) is uniformly and symmetrically distributed about the center of the spherical ion background and the whole system is at rest.

This may represent a situation when most of the electrons are removed from the cluster by a laser field, and the remaining cold electrons occupying the central region collectively oscillate with the frequency $\omega_M$. Electrons are now uniformly shifted (small perturbation) from the ionic background along $x$. The space charge field due to the local charge imbalance acts like a restoring field. Whether these electrons will oscillate at $\omega_M$ is determined by the homogeneity of the charge density $\rho$, and the linearity of the restoring field decided by the amount of perturbation. These are ensured by making the ion background homogeneous and keeping perturbation small so that electrons do not cross the cluster boundary. Under this condition one may write EOM of the center of mass of the electron cloud as $\ddot{x} = \omega^2_M x$, giving $x = x_0 \cos(\omega_M t)$ with initial conditions $x(0) = x_0$, $\dot{x}(0) = 0$; and verify the MD simulation results.

The Fourier transform (FT) of the center of mass position $X_m(t) = \sum_{N} m_n x_n(t)/N_e$ of MD electrons gives the collective oscillation frequency of the electron cloud which is plotted with the FT of the analytical solution $x = x_0 \cos(\omega_M t)$ in Fig. 4 after the normalization by $\omega_M$. An excellent match between the numerical (dashed-circle) and analytical (solid line) results confirms the collective oscillation of electrons at the Mie-plasma frequency by MD simulations. Extensive simulations have been performed for other values of $r_0 = 0.5r_w, 2r_w, 3r_w$, $4r_w$ to check the effect of $r_0$ on the plasma oscillation dynamics. As time progresses, we find $5-20\%$ reduction in the amplitude of plasma oscillation with $5-10\%$ elongation in the plasma period compared to the case of $r_0 = r_w$ and the desired analytical solution shown in Fig. 4.

B. Laser energy absorption and outer ionization

The MD code is now used to study interaction of $n = 5$-cycle laser pulse of duration $nT = 13.5$ fs with the deuterium
cluster as in Sec [I]. Initially, the cluster has equal number of uniformly distributed ions and electrons (i.e., $N_i = N_e = N$) so that it is macroscopically charge neutral.

Figure 5 shows total absorbed energy per electron normalized by $U_p$ and corresponding degree of outer ionization at the end of the laser pulse as the peak intensity is varied. Normalized absorption per electron [in Fig 5(a)] attains a maximum between intensities $5 \times 10^{15} - 10^{16} \text{Wcm}^{-2}$. This non-linear variation of absorbed energy with intensity is similar to that reported earlier using PIC simulations [33] of xenon clusters. The outer ionized fraction [in Fig 5(b)] of electrons, on the other hand, increases gradually with the peak intensity and saturates at unity ($\%$100 outer ionization) at some higher intensity even for this short 5-cycle pulse. At an intensity $5 \times 10^{15} \text{Wcm}^{-2}$, it is inferred that almost 60% electrons are outer ionized ($N \approx 0.6$) which contribute to the total absorbed energy of $\approx 2000 U_p$.

C. Analysis of electron trajectory and finding the AHR

The high level of absorption and outer ionization shown in Fig 5 with a short 13.5 fs laser pulse is certainly not due to the linear resonance process. Figures 6(a-b) show normalized space charge field $E^sc_x = E^sc_x(t)/E_0$ and the total field $E_x = E_x(t)/E_0$ versus excursions $|x|$ of a few selected outer ionized electrons (only 29 electrons are plotted) at the peak intensity $5 \times 10^{15} \text{Wcm}^{-2}$ of Fig 5[1]. Corresponding $|x|$ versus $t$ are shown in Fig 5[c]. The crossing of trajectories of MD electrons [in Fig 5(c)] emitted from the cluster at different times, their non-laminar motion in time, the uncompensated laser field by the space charge field [in Fig 6(a)] and the corresponding non-zero total field [in Fig 6(b)] inside the cluster (-1 $\leq x \leq 1$) clearly suggest that absorption is not due to the celebrated Brunel effect [27]. The underlying mechanism can be understood by analyzing trajectories of those MD electrons and finding the corresponding effective frequency as shown by the model in Sec [I-B]. We write the time dependent frequency of the i-th MD electron (in analogy with Eq. (8)) as

$$\Omega^2[\tau(t)] = \frac{E^sc_x(t) \cdot q_i}{r^2} = \frac{\text{restoring field}}{\text{excursion}}$$

where $E^sc_x = F_i/q_i$ is the electrostatic field on the i-th MD electron obtained from Eq. (10).

Figures 6[a-b] show different normalized quantities, i.e., frequency squared $\Omega^2$, excursion $|x|$, total energy $\Omega^2|\Omega|$, laser field $E_l$ versus normalized time $\tau$ for selected MD electrons which are outer ionized at times: (a) $\tau = 1.1$ and (b) $\tau = 2.2$ from the cluster irradiated by the same 5-cycle laser pulse of peak intensity $5 \times 10^{15} \text{Wcm}^{-2}$. Initially, the cluster is charge neutral, electrostatic field is zero inside the cluster and all particles are at rest. As a result, the effective frequency $\Omega^2[|x(t)|]$ of each electron is zero. As the laser field is switched on, the charge separation potential and the corresponding field are dynamically created due to the movement of more mobile electrons than the slow moving ions. The MD electron in Fig 6(a) is first attracted inside such potential by the restoring force due to ions (see its excursion $|x|$ decreases towards the center of the cluster and total energy starts be-
coming negative) where its effective frequency $\Omega(r(t))$ after increasing from zero exceeds the laser frequency $\omega$ and goes to a maximum value when its total energy reaches a minimum negative value (i.e., it becomes more bound in the potential). From this point onwards the dynamics of the MD electron is very similar to the electron sphere in the model. As the laser field changes further, electron is pulled towards the negative value (i.e., it becomes more bound in the potential). From this point onwards the dynamics of the MD electron is very similar to the electron sphere in the model. As the laser field changes further, electron is pulled towards the negative value (i.e., it becomes more bound in the potential).

The cluster is irradiated by a $n = 5$-cycle pulse of peak intensity $5 \times 10^{15}$ W/cm$^2$. These results resemble with the results of model analysis in Fig. 2.

As the laser field increases further, electron is pulled towards the negative $x$-direction, $|x|$ increases beyond unity, $\Omega^2$ drops from its maximum and crosses the line of AHR (horizontal dashed line where $\Omega^2 = 1$) near $t/T \approx 0.95$ with the corresponding increase in $E_t$ from negative to positive value (bound to free motion) similar to that shown in Fig. 2 using the model. After the AHR, electron leaves the cluster forever with a total energy of $0.8 U_p$ in the end of the pulse. In this early time of interaction, the laser field being very weak, only the loosely bound outermost electrons as compared to the core electrons leave the cluster. Such early leaving electrons which experience AHR in a shallower potential with a low laser field strength generally carry low kinetic energies.

1. **AHR in the frequency vs energy plane**

To prove that all MD electrons essentially pass through AHR during their outer ionization, Figures 8(a-d) show snapshots of all electrons in the ($E_t$, $\Omega^2$) plane at different times $\tilde{t} = 1.1, 1.7, 2.2, 3.2$ respectively. Colors indicate normalized positions $(r)$ of those electrons as in the Fig. 2. From Fig. 8, it is clear that each electron leaves the cluster ($r > 1$, green to dark red) and its energy becomes positive only when it crosses the line of AHR (dashed horizontal line at $\Omega^2 = 1$). After becoming free, electrons have zero effective frequency as they are beyond the influence of the electrostatic field.

In the early time $\tilde{t} = 1.1$, in Fig. 8(a), only few electrons are outer ionized from the cluster and the resulting potential is shallow. As a result energies ($E_t$) of the bound electrons are very close to zero but negative. Some of the bound electrons have negative $\Omega^2$ due to the repulsion of the compressed electron cloud in their vicinity at this early time.

At later times $\tilde{t} = 1.7, 2.2$, in Figs. 8(b)-(c), as the laser field approaches its peak value, an increasing number of electrons are outer ionized via the AHR channel. As a result the potential depth gradually increases, remaining bound electrons move to a deeper potential due to the gradually stronger attractive force of the uncompensated bare ionic background, the population of negative $\Omega^2$ valued electrons moves gradually to the attractive potential (repulsion vanishes with increasing potential depth) and becomes almost negligible in Fig. 8(c) where all bound electrons are aligned to pass the AHR in the next time interval.

![Figure 7](image-url)

Figure 7. (color online) Normalized value of the square of the effective frequency $\Omega^2(r)$, excursion $\pi$, total energy $E_t$ and the laser field $E_l$ versus the normalized time $\tilde{t}$ for different MD electrons undergoing AHR and outer ionization at times (a) $\tilde{t} = 1.1, (b) \tilde{t} = 2.2$. The cluster is irradiated by a $n = 5$-cycle pulse of peak intensity $5 \times 10^{15}$ W/cm$^2$. These results resemble with the results of model analysis in Fig. 2.

![Figure 8](image-url)

Figure 8. (color online) Snapshots all MD electrons in the ($E_t$, $\Omega^2$) plane at times (a) $\tilde{t} = 1.1$, (b) $\tilde{t} = 1.7$, (c) $\tilde{t} = 2.2$, (d) $\tilde{t} = 3.2$. As the laser field strength increases with time, more and more electrons are drawn towards the line of AHR, i.e., dashed line at $\Omega^2 \approx 1$. The parameters of laser and cluster are same as in Fig. 2.

The occurrence of AHR for MD electrons in Fig. 7 resemble with Fig. 2 in the model in Sec. 11 except that frequency and potential start from zero and self-consistently generated in the case of MD while those are predefined in the model.
After the peak of the laser pulse, e.g., at $\tilde{t} = 3.2$ in Fig.8, since outer ionization is mostly saturated and the potential has already reached to its near maximum depth at the pulse peak before (i.e., near $\tilde{t} = 2.5$), many bound electrons are dragged into the potential (they have more negative energies) due to the weakening of the laser field compared to the attractive force due to ions. Some of the quasi-free electrons (electrons with positive $\Omega^+$ and positive $\delta_{1}$) near the cluster boundary also return inside due to such attraction.

The feature of AHR shown in Fig.8 in the frequency versus energy plane resembles Fig.3 obtained by the model, except that some MD electrons in Fig.8 experience negative frequencies due to repulsion of the neighbouring electrons.

Above analysis of trajectories of MD electrons in the self-generated, time-varying potential clearly indicates that the passage of AHR is must during their outer ionization. The fact that large amount of energy absorption by an electron and weakening of the laser field compared to the attractive force due to ions. Some of the quasi-free electrons (electrons with positive $\Omega^+$ and positive $\delta_{1}$) near the cluster boundary also return inside due to such attraction.

The prompt generation of electrons via AHR within a time much shorter than a laser period, the crossing of electron trajectories [in Fig.6(c)] demonstrated by MD simulations and the breaking of laminar flow of electrons may lead to plasma wave-breaking and subsequent mixing of wave-phases even at sub-relativistic laser intensities in an extended plasma.

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