Nonlinear resonance absorption in laser-cluster interaction

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Rare gas or metal clusters are known to absorb laser energy very efficiently. Upon cluster expansion the Mie plasma frequency may become equal to the laser frequency. This linear resonance has been well studied both experimentally and theoretically employing pump probe schemes. In this work we focus on the few-cycle regime or the early stage of the cluster dynamics where linear resonance is not met but nevertheless efficient absorption of laser energy persists. By retrieving time-dependent oscillator frequencies from particle-in-cell simulation results, we show that nonlinear resonance is the dominant mechanism behind outer ionization and energy absorption in near infrared laser-driven clusters.

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The construction of laser-based table-top sources of energetic particles into the cold bulk in the case of solids. In fact, almost 100% absorption of the laser light was observed in experiments with rare gas clusters [1].

The interaction scenario on which most researchers in the field agree upon is as follows: after removal of the first electrons from their “parent” ions (inner ionization) and the cluster as a whole (outer ionization) the cluster charges up. The total electric field (i.e., laser plus space charge field) inside the cluster leads to inner ionization up to high charge states not possible with the laser field alone (ionization ignition [2, 3]). However, the restoring force of the ions counteracts outer ionization so that ionization ignition stops at some point. Moreover, the cluster expands due to Coulomb explosion and thermal pressure, thus lowering the electric field due to the ions. The latter determines the dominant eigenfrequency of the cluster, i.e., the Mie frequency

$$\omega_{\text{Mie}}(t) = \frac{\omega_p(t)}{\sqrt{3}} = \sqrt{\frac{4\pi\rho(t)}{3}} = \sqrt{\frac{N_i(t)Z(t)}{R^3(t)}}$$

with $\rho$ the charge density, $N_i$ the number of ions of (mean) charge state $Z$, and $R$ the cluster radius (atomic units are used unless noted otherwise). For laser wavelengths $\approx 500$ nm, soon after the removal of the first electrons $\omega_{\text{Mie}}$ exceeds $\omega_l$. Hence linear resonance

$$\omega_{\text{Mie}}(t) = \omega_l$$

occurs not before the cluster has sufficiently expanded (typically after a few hundred femtoseconds). At linear resonance the electric field inside the cluster is enhanced instead of shielded [2] so that even higher charge states can be created and even more energy can be absorbed from the laser.

Absorption of laser energy is only possible through resonances (linear or nonlinear) or non-adiabaticities (collisions). Commonly used phrases to explain absorption like “laser dephasing heating” or “collisions with the cluster boundary” are correct but meaningless since dephasing is, according to Poynting’s theorem, a prerequisite for absorption while the electron collisions with the cluster boundary have to occur at the right frequency in order to yield efficient absorption.

The importance of the linear resonance has been demonstrated both in pump probe experiments and simulations [2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. The emission of third and higher harmonics of the incident laser light has been observed in computer simulations [13]. Collisional ionization and absorption are of minor importance at wavelengths $\approx 800$ nm or greater [14, 15, 16] whereas it is the dominant absorption mechanism at short wavelengths [16, 17] not studied in the present Letter.

One of the crucial points in the above mentioned scenario is the mechanism of outer ionization, which goes hand in hand with absorption [18] since the laser energy is transiently stored in the freed, energetic electrons. These electrons leave net positive charge behind, which finally Coulomb explodes. The latter converts electron energy into ion energy, which explains why experimentalists typically measure MeV ions but only keV electrons [19, 20, 21]. In order to separate outer ionization and absorption due to the linear resonance [2] from other mechanisms we consider only the first few tens of femtoseconds of the laser-cluster interaction where ion motion is negligible and linear resonance is therefore ruled out. Previous work pointed out already the possible importance of nonlinear resonance [22] or, equivalently, Landau damping in finite systems [23]. So far these mechanisms were clearly observed only in simplified model systems. The interpretation of results from molecular dynamics or particle-in-cell simulations, on the other hand, is of-
ten hampered by the complex dynamics of the individual particles that makes the clear distinction of absorption mechanisms difficult. In this Letter we bridge this gap by analyzing our particle-in-cell results in terms of nonlinear oscillators. This enables us to prove that essentially all electrons contributing to outer ionization pass through the nonlinear resonance, so that the latter is unequivocally identified as the collisionless absorption mechanism if linear resonance is impossible.

In general, the eigenfrequency of a particle in a given potential depends on the excursion amplitude (or the energy) of the particle, \( \omega = \omega_1 \). In a (laser-) driven system the excursion amplitude is time-dependent so that it may dynamically pass through the resonance

\[
\omega[\tilde{r}(t)] = \omega_1, \tag{3}
\]

where \( \omega_1 \) is the driver frequency. Equation (3) defines nonlinear resonance (NLR). Refs. 23, 25 discuss at length the significant features of laser-driven systems undergoing NLR. Here we restrict ourselves to the prerequisites necessary to understand the analysis of our particle-in-cell results below.

In the rigid sphere-model (RSM) (see, e.g., 23, 26, 27) of a cluster both electrons and ions are modelled by homogeneously charged spheres which, in a linearly polarized laser field, oscillate along \( z \) around their common center of mass, which is in good approximation the center of the ion sphere. In the case of equal charge density and radii \( R \), the equation of motion for the center of the electron sphere can be written in dimensionless entities as

\[
\frac{d^2 \tilde{r}}{d\tau^2} + \left( \frac{\omega_{\text{Mie}}}{\omega_1} \right)^2 \text{sgn}(z) \left\{ r - \frac{9r^2}{16} + \frac{r^4}{64} \right\} = \frac{E(\tau)}{R\omega_1^2} \tag{4}
\]

Here, \( r = |z|/R, \tau = \omega_1 t \), and the upper line applies for \( 0 \leq r < 2 \), the lower line (Coulomb force) for \( r \geq 2 \). The amplitude of the dimensionless driver \( E(\tau)/R\omega_1^2 \) is the excursion amplitude of a free electron divided by the cluster radius. The only other independent dimensionless parameter in the RSM is the ratio of Mie frequency to laser frequency. As was observed in Ref. 23 the absorption of laser energy in the RSM is characterized by a threshold driver strength below which absorption is negligible (harmonic regime) and above which absorption is almost constant. Figure 1 shows this threshold behavior for \( \omega_{\text{Mie}}/\omega_1 = 2.7 \) and a \( n = 10 \)-cycle sin^2-pulse \( E(\tau) = -E_0 \sin^2(\tau/2n) \cos(\tau) \). The threshold driver strength in the RSM can actually be calculated, either employing perturbation theory (as long as \( \omega_{\text{Mie}} \) is only a few times the laser frequency) or by approximating it with the over-barrier field strength for the cluster potential at larger ratios \( \omega_{\text{Mie}}/\omega_1 \). Here we restrict ourselves to the method for the identification of NLR, which will be used for the more realistic particle-in-cell results below. Equation (4) can be formally rewritten as

\[
\frac{d^2 \tilde{r}}{d\tau^2} + \left( \frac{\omega_{\text{eff}}}{\omega_1} \right)^2 \text{sgn}(z) r = \frac{E(\tau)}{R\omega_1^2}. \tag{5}
\]

Although (5) is not useful for solving the equation of motion, it is helpful for analyzing the solution \( r(\tau) \) since

\[
\left( \frac{\omega_{\text{eff}}(\tau)}{\omega_1} \right)^2 = \frac{1}{\text{sgn}(z)(\tau)} \left( \frac{E(\tau)}{R\omega_1^2} - \tilde{r}(\tau) \right). \tag{6}
\]

yields the instantaneous, scaled effective frequency \( \omega_{\text{eff}}(\tau)/\omega_1 \), which passes through unity at the NLR 4. Figure 1 shows a typical example for the temporal behavior of \( \omega_{\text{eff}}(\tau)/\omega_1 \) above the threshold driver strength in Fig. 1. Since \( \omega_{\text{Mie}}(\omega_1)^2 = 7.29 \), \( \omega_{\text{eff}}(\tau)/\omega_1 \) starts at this value (dashed line in Fig. 1) and drops with increasing driver strength. It passes through unity at the time indicated by the vertical line, and it is exactly at that time where the electron sphere is set free, as it is clearly visible from the energy of the electron sphere, which passes through zero, and the excitation. We have checked that outer ionization and occurrence of NLR holds for all driver strengths above the threshold whereas the resonance is never met below threshold.

Let us now turn to the particle-in-cell (PIC) 29 results. We consider pre-ionized clusters of fixed radius \( R = 3.2 \text{ nm} \) but of various charge densities (i.e., different degree of inner ionization). The ratio of charge density to critical density \( \rho/\rho_c = \omega_{\text{Mie}}^2/\omega_1^2 = 3\omega_{\text{Mie}}^2/\omega_1^2 \) varies from 20 to 100. The clusters are exposed to 8-cycle sin^2-pulses of wavelength \( \lambda = 1056 \text{ nm} \). Since ion motion does not play an important role during the simulation time the ions are fixed, which ensures a well defined, constant Mie frequency \( \omega_{\text{Mie}} \). Figure 2 shows the absorbed energy per electron in units of the ponderomotive potential
because of depletion of electrons. The threshold intensity increasing charge density the maxima of the absorbed
maximum in the corresponding PIC absorption curve. With
is indicated in Fig. 3. It agrees very well with the max-
ities while the absorbed energy per electron decreases.

FIG. 2: (color online). Typical behavior of \((\omega_{\text{eff}}(\tau)/\omega_l)^2\)
(drawn red) vs. time above the threshold driver strength in
Fig. 1 (actual value was \(E_0/R\omega_l^2 = 3.3\)). Excursion \(\varphi/\rho\)
(black) and energy of the electron sphere \(E_{\text{tot}}/R^2\omega_l^2\) (green)
are included in the plot. Outer ionization (i.e., \(E_{\text{tot}}/R^2\omega_l^2 \geq 0\))
and occurrence of nonlinear resonance \((\omega_{\text{eff}}(\tau)/\omega_l)^2 = 1\)
dashed-dotted line always coincide (vertical line).

\[ U_p = E_0^2/4\omega_l^2, \text{ i.e., the time-averaged quiver energy of} \]
a free electron in the laser field. One sees that the ab-
sorbed energy per electron is always on the order of \(U_p\).
However, the absorbed energy is nonlinear in \(U_p\) and dis-
plays a maximum around an intensity \(I_{th}\) before it drops
because of depletion of electrons. The threshold intensity
\(I_{th}\) for the case \(\rho/\rho_c = 40\), as it is predicted by the RSM,
is indicated in Fig. 2. It agrees very well with the max-
imum in the corresponding PIC absorption curve. With
increasing charge density the maxima of the absorbed
energy (divided by \(U_p\)) move towards higher laser inten-
sities while the absorbed energy per electron decreases.

The motion of the PIC particles can be analyzed in the
same way as it was done with the motion of the electron

FIG. 3: (color online). Total absorbed energy per electron in
units of \(U_p\) vs. laser intensity for charge densities between 20
and 100 times the critical density \((N = 2749\) to 13694 elec-
trons). \(I_{th}\) is the threshold intensity for \(\rho/\rho_c = 40\) predicted by
the rigid sphere-model.


\[ \omega_{\text{eff},i}^2(t) = \frac{[E(t) + \vec{r}_i(t)] \cdot \vec{r}_i(t)}{r_i^2(t)} = \frac{E_{\text{sc}}(\vec{r}_i,t) \cdot \vec{r}_i(t)}{r_i^2(t)}. \]  

The equation for the effective, time-dependent oscillator
frequency analogous to (6) then reads

Clearly, \(E_{\text{sc}}(\vec{r}_i,t)\) depends on the position of all other
particles \(\neq i\) as well. The PIC simulation starts with
the neutral cluster configuration where the electrons sit
on top of the ions so that \(E_{\text{sc}}(\vec{r},0) = 0\). Hence, a PIC
electron “sees” initially an effective frequency \(\omega_{\text{eff},i}(0) = 0\).
The laser field disturbs the charge equilibrium and
\(\omega_{\text{eff},i}(t)\) becomes different from zero. \(\omega_{\text{eff},i}(t)\) may be even
negative in regions of accumulated electron density (re-
pulsive potential). As the cluster charges up, \((\omega_{\text{eff}}/\omega_l)^2\)
quickly increases beyond unity (where the RSM starts in
the first place). The starting from \(\omega_{\text{eff},i}(0) = 0\), the possi-
bility of negative \(\omega_{\text{eff},i}(t)\), and the three-dimensionality
are the main differences to the RSM analysis above.

By following the dynamics of the electrons in the effec-
tive frequency vs. energy-plane one can identify the
main pathway to outer ionization and efficient absorp-
tion. In Fig. 4 a–d the scaled effective frequencies squared
\((\omega_{\text{eff}}/\omega_l)^2\) of the individual PIC electrons are plotted vs.
their energies \(E_{\text{tot}}(t) = \frac{\vec{r}_i^2(t)}{2R\omega_l^2}\) they would have
if the driver is switched off instantaneously at \(t = 2.5, 3, 3.5,\) and 4 laser cycles, respectively. We define the
time when, for a particular electron, \(E_{\text{tot}}\) becomes
0 as the ionization time of that electron. The laser
intensity is \(2.5 \times 10^{16}\) Wcm\(^{-2}\), and the preionized cluster
is 40 times overcritical so that \((\omega_{\text{Mie}}/\omega_l)^2 = 40/3\). As
is clearly visible in Fig. 4 each electron reaches posi-
tive energy close to the point \((\omega_{\text{Mie}}/\omega_l^2, E_{\text{tot}}(U_p)) = (1,0)\).
The radial position of each electron is color coded, indi-
cating that outer ionization occurs at radii around \(2R\).
Data points at radii \(> 3R\) (orange and yellow colors in
Fig. 4 a,b) with positive but very small \(E_{\text{tot}}\) and \(\omega_{\text{eff}}^2 \approx 0\)
represent low energetic electrons removed earlier during
the pulse. Electrons with positive energy but small radii
(visible in (a) and (b)) are those driven back to the clus-
ter by the laser field. For the electrons inside the cluster
potential (negative energies and \(R < R\), color coded
blue and black in Fig. 4) \((\omega_{\text{eff}}/\omega_l)^2\) spreads over a wide
range, starting from the maximum value \((\omega_{\text{Mie}}/\omega_l)^2\) down
to negative values due to the repulsive force exerted by
the compressed electron cloud. Note that negative values
occur mainly at early times where most of the electrons

sphere in the RSM above. A PIC particle has the same
charge to mass ratio as a “real” electron, that is, \(e/m = -1\) in atomic units. Each PIC particle moves under the
influence of the external laser field and the space charge
field \(E_{\text{sc}} = -\nabla \Phi(\vec{r},t)\) due to the potential \(\Phi(\vec{r},t)\) that
is created by all charges (mapped to a numerical grid).
Hence the equation of motion of the ith PIC particle is

\[ \ddot{r}_i + E_{\text{sc}}(\vec{r}_i,t) = -E(t). \]  

The radial position of each electron is color coded, indi-
cating that outer ionization occurs at radii around \(2R\).
Data points at radii \(> 3R\) (orange and yellow colors in
Fig. 4 a,b) with positive but very small \(E_{\text{tot}}\) and \(\omega_{\text{eff}}^2 \approx 0\)
represent low energetic electrons removed earlier during
the pulse. Electrons with positive energy but small radii
(visible in (a) and (b)) are those driven back to the clus-
ter by the laser field. For the electrons inside the cluster
potential (negative energies and \(R < R\), color coded
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range, starting from the maximum value \((\omega_{\text{Mie}}/\omega_l)^2\) down
to negative values due to the repulsive force exerted by
the compressed electron cloud. Note that negative values
occur mainly at early times where most of the electrons
are still inside the cluster. The occurrence of NLR is less clear for the few electrons leaving the cluster even earlier than \( t = 2.5 \) laser cycles. As mentioned above, these electrons move in a shallow effective potential with \( (\omega_{\text{eff}}/\omega)^2 < 1 \) when they leave the cluster with ease and with rather low kinetic energy because the laser intensity is still low at the time of their emission.

One may object that, since the denominator in (8) necessarily increases while the nominator decreases for \( t \) when they leave the cluster with ease and the nominator decreases for \( \omega_{\text{eff}} < \omega_{\text{min}} \), the occurrence of NLR is indeed the responsibility of outer ionization accompanied by efficient absorption of laser energy.

In summary, we have shown that cluster electrons contributing to efficient absorption and outer ionization in near infrared laser fields undergo nonlinear resonance, meaning that the instantaneous frequency of their motion in a time-dependent, anharmonic, effective potential meets the laser frequency. Nonlinear resonance is the only possible absorption mechanism if the laser pulse is too short for the linear resonance to occur (or during the early cluster dynamics in longer pulses) and if electron-ion collisions (inverse bremsstrahlung) are negligible, as it is the case at near infrared or greater wavelengths. In order to prove the occurrence of nonlinear resonance we introduced a method to analyze the results obtained from particle-in-cell simulations, namely the mapping of the system of electrons and ions that interact through their mean field onto a system of nonlinear oscillators whose time-dependent frequencies unequivocally revealed the coincidence of electron removal and nonlinear resonance.

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