Harmonic generation in clusters

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(Dated: March 18, 2004)

Abstract

A model is presented for the nonlinear response of a small cluster, with a size much smaller than the wavelength, at the third harmonic of the laser frequency. The model involves collective modes of a cold electron core confined within a positively charged ion background. The response of the electron core to the laser field is similar to that of a weakly nonlinear oscillator driven by an external force. In particular, there is a resonant enhancement of the third harmonic when the frequency of the applied field is close to one third of the core eigenfrequency. It is shown that density nonuniformity or nonspherical shape of the ion background is necessary for harmonic generation. Particle-in-cell simulations have been performed to model the time evolution of the third harmonic response as the ion density profile changes due to cluster expansion. The simulation results are consistent with the predictions of the cold electron core model. In addition, the code quantifies the role of stochastic electron heating, an alternative harmonic generation mechanism.
I. INTRODUCTION

Recent experiments on laser interaction with small clusters reveal an interesting phenomenon of harmonic generation [1, 2]. This effect can serve as a diagnostic tool in cluster experiments. It may also be useful for producing coherent short pulses of soft X-rays. The experiments typically deal with clusters that are smaller in size than both the incident laser wavelength and the radiated wavelength. It is therefore appropriate to treat the harmonic emission as dipole radiation of the cluster. This approach reduces the problem to finding Fourier harmonics of the cluster dipole moment, which is the main technical goal of our paper.

It is apparent that the cluster response to the laser field has to be nonlinear to produce harmonics. The physics origin of this nonlinearity is the nonlinearity of electron oscillations in the potential well created by the ion background. There are several simplifications that facilitate the corresponding analysis. First, if the laser pulse is not too strong (which we assume to be the case in this paper) then the electron motion in the cluster is nonrelativistic. It is noteworthy, that the applicability condition for this assumption to a high density cluster is considerably weaker than the corresponding condition in vacuum (see Section II). Second, it is allowable to use electrostatic approximation to calculate the electron response. Third, the problem becomes axisymmetric when the laser is linearly polarized.

Yet another important aspect of the problem is that the electron response can be strongly enhanced by collective effects when the harmonic frequency resonates with a linear eigenmode in the cluster. The mode of primary interest is the Mie dipole mode [3, 4]. The corresponding eigenfrequency for a nearly uniform cluster is $\omega_p/\sqrt{3}$, where $\omega_p$ is the electron plasma frequency.

Making use of the features described above, we develop an analytical model and a numerical code to calculate nonlinear electron response induced in the cluster by the laser pulse. The code allows us to go beyond the technical limitations of the analytical theory. In particular, the theory is restricted to the case of only slightly nonuniform cluster with a fixed ion density profile, whereas the code is free from this constraint, which enables it to simulate self-consistently the resonant enhancement of harmonic generation during cluster expansion. In addition, the code takes account of hot electrons that undergo “vacuum heating” at the cluster boundary. This will enable us to compare the relative roles of the hot
electron population and the cold electron core.

Nonlinear excitation of the Mie resonance has recently been discussed in Refs. [5–7] in relation to harmonic generation in clusters. Although the model developed in these references has the same basic ingredients as our theory (collective eigenmode and nonlinear coupling), the actual cause of the nonlinear response and the rigorousness of its analysis are different. Reference [6] deals with a single-fluid model for heated electrons in a spatially uniform spherical ion background. The authors assume that the electron cloud oscillates as a rigid body in step with the applied field and that certain part of the electron cloud crosses the ion boundary. Within this model, the effect of the sharp edge in ion density is crucial. The resulting nonlinear force is proportional to the gradient of the electron density at the edge of the ion sphere. The problem with this model is that the rigid displacement approximation fails for the edge electrons, which invalidates the perturbative approach used in Refs. [5–7].

As shown in Ref. [8], there are two electron populations in the cluster, the cold core and the heated halo, and they respond differently to the applied laser field. In an equilibrium state, the radius of the cold electron core is smaller than that of the ion-cluster. As long as the core is inside the ion sphere, it moves coherently in response to the laser-field. In Sec. III we will show that the core motion can be described by rigid displacement as long as the cluster non-uniformity is small. Although this may resemble the rigid displacement approximation from Ref. [5], the fact that we consider only those core electrons that do not touch the edge makes a big difference and allows us to derive an accurate analytical expression for their nonlinear response. In contrast with the core electrons, the halo electrons move chaotically. The chaotic electron halo that crosses the ion boundary can contribute to harmonic generation. However, this contribution cannot be described in terms of rigid coherent displacement.

Based on the two-component electron picture, we will demonstrate that the cold electron core plays the dominant role in harmonic generation, provided that the ion density profile inside the cluster is nonuniform. A natural reason for this nonuniformity is the ion motion during cluster expansion.

In this work, we will limit our analysis to the case of a single cluster. By doing so, we will demonstrate the basic mechanism of harmonic generation with particular attention to the third harmonic. However, our results will not be quite ready for quantitative comparison with experimental data since the experiments typically deal with many clusters. One would
therefore need to take into account that the measured signal involves averaging over the cluster distribution and the laser beam profile. Also, interference effects need to be considered to properly interpret the data. These two aspects go beyond the scope of the present paper.

The paper is organized as follows. In Sec. II, we present a qualitative analysis of electron dynamics and introduce parameters relevant to the problem. In Sec. III, we develop an analytical description of harmonic generation by a cold electron core within a slightly non-uniform ion background. We show that coherent response of the electron core satisfies the equation for a driven nonlinear oscillator. In Sec. IV, we describe our particle-in-cell numerical model that employs electrostatic approximation and axial symmetry. The model treats electron and ion dynamics self-consistently. The laser electric field is included as a spatially uniform time-dependent field. In Sec. V, we study the harmonic generation numerically and we demonstrate that the cold electron core, if present in the cluster, plays the dominant role in the effect. We also verify the analytical conclusion that non-uniformity of the ion background is essential for harmonic generation. In Sec. VI, we study the role of hot electrons in more detail. In Sec. VII, we summarize the results.

II. PRELIMINARY REMARKS ON ELECTRON DYNAMICS

The typical laser intensity in cluster experiments [2] is in the range of $10^{15} - 10^{16}$ W/cm$^2$, which is significantly larger than the ionization threshold ($10^{14}$ W/cm$^2$ for hydrogen). Therefore, the cluster becomes ionized at the very beginning of the laser pulse. For light atoms like hydrogen or helium the ionization is complete; for heavy atoms partial ionization may lead to additional effects that will not be discussed here.

Inside the cluster the plasma frequency $\omega_p$ is typically larger than the laser frequency $\omega_0$. In response to the laser electric field $E_0 \cos(\omega_0 t)$ the inner electrons move adiabatically in order to partially compensate the applied field. These electrons represent a cold electron core (see Fig. 1). Collisional heating of the core is usually insignificant on the time scale of the laser pulse [8].

In the case of uniform spherical cluster, the electron core remains uniform and spheri-
The displacement of the core is
\[ d = \frac{1}{\omega_p^2/3 - \omega_0^2} \frac{eE_0}{m_e} \cos(\omega_0 t), \] (1)
where \( e \) is the electron charge, \( m_e \) is the electron mass. The radius of the electron core is \( R_e = R - d_{\text{max}} \), where \( d_{\text{max}} \) is the maximum value of the core displacement during the interaction and \( R \) is the radius of the cluster. The core oscillates at the frequency \( \omega_0 \), so that its characteristic velocity is \( v_{\text{core}} = \omega_0 d \). The motion of the core is nonrelativistic even for relatively large amplitude pulses with \( eE_0/m_e c \omega_0 \sim 1 \) as long as \( \omega_0 \ll \omega_p \):
\[ v_{\text{core}} \approx \frac{3c \omega_0^2}{\omega_p^2 m_e c \omega_0}. \] (2)

There are two factors that lead to the nonlinearity of the electron core response. First, relativistic motion is nonlinear. The small dimensionless parameter describing this nonlinearity is \( v_{\text{core}}/c \). Second, nonuniformity of the cluster density makes the oscillations nonlinear. The corresponding small dimensionless parameter is \( \delta n/n_0 \cdot d/R \), where \( d/R \) is the ratio of the core displacement to the cluster radius and \( \delta n/n_0 \) is the relative nonuniformity of the ion density.

In order to understand the relative role of the two factors leading to the nonlinearity, we estimate the ratio of the small parameters involved:
\[ \frac{v_{\text{core}}/c}{\delta n/n_0 \cdot d/R} = 2\pi R n_0 / \lambda \delta n. \] (3)

Since the laser wavelength is much larger than the cluster radius (the ratio \( \lambda/R \) is about \( 10^2 \) in experiments described in Ref. [2]), we conclude that a small density perturbation in the cluster can easily be the main reason for nonlinearity of the core response.

In addition to the core electrons, there are some electrons which are extracted from the cluster and accelerated by the external field. The behavior of these electrons depends on the amplitude of the laser field. In the strong field limit, when the characteristic excursion of an extracted electron \( \zeta = eE_0/m_e \omega_0^2 \) is much larger than the cluster radius \( R \), these electrons are not trapped in the cluster potential and they leave the cluster. In the opposite case of \( \zeta \ll R \), the extracted electrons return to the cluster as the external field changes sign. As these electrons bounce back and forth, they continuously gain energy due to so-called
vacuum heating [9] or surface heating [4]. As a result, a two-component electron distribution arises in the cluster [8]. Of these two, it is the cold component that produces the most of the coherent nonlinear response.

III. ANALYTICAL TREATMENT OF THIRD HARMONIC GENERATION BY COLD ELECTRONS

The aim of this section is to describe analytically how the laser field produces a strong response of the cluster at the third harmonic of the laser frequency. This effect is very similar to the response of a weakly nonlinear oscillator that is driven by a sinusoidal force, \( f \sin \omega_0 t \), where \( \omega_0 \) is close to \( \frac{1}{3} \) of the oscillator eigenfrequency \( \Omega \) [10].

In order to link the cluster problem to the oscillator problem, we use a Lagrangian for the cluster electrons in the presence of immobile ions,

\[
L = \frac{m_e}{2} \int d^3 r n(r) \left( \frac{\partial \xi_i}{\partial t} \right)^2 - \frac{e^2}{2} \int d^3 r d^3 r' n(r) n(r') \left( \frac{1}{|r + \xi(r,t) - r' - \xi(r',t)|} - \frac{1}{|r - r'|} \right) \\
+ e^2 \int d^3 r d^3 r' n(r) N(r') \left( \frac{1}{|r + \xi(r,t) - r'|} - \frac{1}{|r - r'|} \right) + e \int d^3 r n(r) E_i(t) \xi_i(r,t).
\]

The dynamical variable in this Lagrangian is the electron displacement vector \( \xi_i(r,t) \), where \( r \) is the particle equilibrium position, and the subscript \( i \) for \( \xi \) denotes the corresponding Cartesian component. The functions \( n(r) \) and \( N(r) \) are the unperturbed densities of electrons and ions in the absence of the laser electric field \( E_i(t) \). The individual terms in the Lagrangian represent the electron kinetic energy, the electron-electron Coulomb energy, the electron-ion Coulomb energy, and the electron interaction with the laser electric field (this field is time-dependent, but spatially uniform within the cluster). It is apparent that \( n(r) \) and \( N(r) \) are equal within the cold electron core. We will assume that the ion density profile extends beyond the electron core, so that there is a positive ion shell around the neutralized core (see Fig. 1). We will also assume that the electron displacement is smaller than the shell thickness \( \Delta \), so that all electrons remain inside the potential well in their oscillatory motion. This requires the laser electric field to be smaller than: \( E_0 \ll (\omega_0^2 / 3 - \omega_0^2) m_e \Delta / e \) [see Eq. (1)].

In order to make the problem tractable analytically, we will introduce an additional
assumption that \( N(r) \) is nearly flat within a spherical boundary. Yet, we will retain a small deviation from a purely flat ion profile, which turns out to be essential for harmonic generation.

We now expand the Lagrangian in powers of \( \xi_i(r,t) \) up to fourth-order terms. For brevity we suppress \( r \) and \( t \) in the arguments of displacement, but we keep \( r_1 \) in the argument. This expansion starts from quadratic terms since the linear term vanishes due to the requirement that \( \xi_i(r,t) = 0 \) represents electron equilibrium. We then find that

\[
L = \frac{m_e}{2} \int d^3r n(r) \left( \frac{\partial \xi_i}{\partial t} \right)^2 + \frac{e^2}{2} \int d^3r d^3r_1 n(r) n(r_1) \xi_i \xi_k(r_1) \frac{\partial^2}{\partial x_i \partial x_k} \frac{1}{|r - r_1|} \\
- \frac{e^2}{12} \int d^3r d^3r_1 n(r) n(r_1) [\xi_i - \xi_i(r)] [\xi_k - \xi_k(r_1)] [\xi_i - \xi_i(r_1)] \frac{\partial^3}{\partial x_i \partial x_k \partial x_i} \frac{1}{|r - r_1|} \\
- \frac{e^2}{48} \int d^3r d^3r_1 n(r) n(r_1) [\xi_i - \xi_i(r_1)] [\xi_k - \xi_k(r_1)] \\
\times [\xi_i - \xi_i(r_1)] [\xi_m - \xi_m(r_1)] \frac{\partial^4}{\partial x_i \partial x_k \partial x_i \partial x_m} \frac{1}{|r - r_1|} \\
+ \frac{e^2}{6} \int d^3r d^3r_1 n(r) N(r_1) \xi_i \xi_k \xi_l \frac{\partial^3}{\partial x_i \partial x_k \partial x_l} \frac{1}{|r - r_1|} \\
+ \frac{e^2}{24} \int d^3r d^3r_1 n(r) N(r_1) \xi_i \xi_k \xi_l \xi_m \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|} + e \int d^3r n(r) E_i(t) \xi_i. \tag{5}
\]

In this expression, the quadratic parts of the electron-electron and electron-ion Coulomb energies are combined into a single term. In doing so, we take into account that the quantity

\[
\int [n(r_1) - N(r_1)] \frac{d^3r_1}{|r - r_1|} \tag{6}
\]

is constant within the electron core, because this quantity represents the equilibrium electrostatic potential created by the ion shell in the core.

It is noteworthy that the electron-ion terms in Eq. (5) vanish in the case of a uniform spherical ion background, which can be verified in a straightforward way. We also observe that the third- and the fourth order electron terms in the Lagrangian vanish if we consider a rigid displacement of the electron core. The underlying physics is that rigid displacement of all electrons does not affect their mutual potential energy. As a result, the rigid displacement is an exact solution of the equations of motion for a uniform spherical cluster in the presence of a sinusoidal driving field. These observations suggest that a displacement \( \xi_i(r,t) \) with a
weak spatial dependence is a relevant candidate solution for a slightly nonuniform and/or non-spherical cluster. The spatial variation in such a solution would scale linearly with the small nonuniform part of \( N(r) \). This allows us to neglect the third- and fourth order electron-electron interaction terms in Eq. (5) compared to the corresponding electron-ion terms and to obtain the following Euler-Lagrange equation:

\[
m_e \frac{\partial^2 \xi_i(r; t)}{\partial t^2} - e^2 \int d^3r_1 n(r_1) \frac{\partial^2}{\partial x_i \partial x_k} \frac{1}{|r - r_1|} - \frac{e^2}{2} \xi_k(r; t) \xi_t(r; t) \int d^3r_1 N(r_1) \frac{\partial^3}{\partial x_i \partial x_k \partial x_l} \frac{1}{|r - r_1|} - \frac{e^2}{6} \xi_k(r; t) \xi_t(r; t) \xi_m(r; t) \int d^3r_1 N(r_1) \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|} - eE_i(t) = 0.
\] (7)

Here the space-time dependance of \( \xi \) is now explicitly displayed.

Assuming that the laser electric field is monochromatic and linearly polarized, we set

\[
E_i(t) = \frac{\epsilon_i}{2} [E_0 \exp(-i\omega_0 t) + \text{c.c.}],
\] (8)

where \( \epsilon_i \) is the unit polarization vector and \( E_0 \) is the field amplitude, and we seek \( \xi_i \) in the form:

\[
\xi_i = \xi_{(0)i} + \frac{1}{2} [\xi_{(1)i} \exp(-i\omega_0 t) + \xi_{(2)i} \exp(-2i\omega_0 t) + \xi_{(3)i} \exp(-3i\omega_0 t) + \text{c.c.}] + \cdots .
\] (9)

This representation leads to a truncated set of equations for the Fourier amplitudes of the displacement. In particular, to the leading order the third harmonic equation of the displacement has the form:

\[
9m_e \omega_0^2 \xi_{(3)i}(r) + e^2 \int d^3r_1 n(r_1) \xi_{(3)k}(r_1) \frac{\partial^2}{\partial x_i \partial x_k} \frac{1}{|r - r_1|} + \frac{e^2}{4} [\xi_{(2)k}(r) \xi_{(1)i}(r) + \xi_{(1)k}(r) \xi_{(2)i}(r)] \int d^3r_1 N(r_1) \frac{\partial^3}{\partial x_i \partial x_k \partial x_l} \frac{1}{|r - r_1|} + \frac{e^2}{24} \xi_{(1)k}(r) \xi_{(1)i}(r) \xi_{(1)m}(r) \int d^3r_1 N(r_1) \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|} = 0.
\] (10)

Calculation of the nonlinear force in Eq. (10) involves equations for the second and first
harmonics:

\[ 4 m_e \omega_0^2 \xi_{(2)i}(r) + e^2 \int d^3 r_1 n(r_1) \xi_{(2)k}(r_1) \frac{\partial^2}{\partial x_i \partial x_k} \frac{1}{|r - r_1|} 
+ \frac{e^2}{4} \xi_{(1)k}(r) \xi_{(1)i}(r) \int d^3 r_1 N(r_1) \frac{\partial^3}{\partial x_i \partial x_k \partial x_l} \frac{1}{|r - r_1|} = 0, \]  
(11)

\[ m_e \omega_0^2 \xi_{(1)i}(r) + e^2 \int d^3 r_1 n(r_1) \xi_{(1)k}(r_1) \frac{\partial^2}{\partial x_i \partial x_k} \frac{1}{|r - r_1|} + e E_0 \xi_i = 0. \]  
(12)

It is apparent that \(\xi_{(1)i}\) scales linearly with the laser field amplitude \(E_0\) and that \(\xi_{(2)i}\) scales as \(E_0^3\). Therefore, both nonlinear terms in Eq. (10) scale as \(E_0^3\). Yet, in the case of nearly flat ion density profile, the first of the two nonlinear terms in Eq. (10) is much smaller than the second one. The reason is that \(\int d^3 r_1 N(r_1) \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|}\) vanishes for a uniform spherical cluster. Therefore, \(\xi_{(2)i}\) is proportional to the deviation \(\delta N\) from the uniform ion density. As a result, the first nonlinear term in Eq. (10) is proportional to the square of \(\delta N\), whereas the second term is linear in \(\delta N\).

Equation (10) involves a self-adjoint linear operator \(\hat{L}\) defined by:

\[ \hat{L}_{ik} \xi_{(3)k} \equiv -\frac{e^2}{m_e} \int d^3 r_1 n(r_1) \xi_{(3)k}(r_1) \frac{\partial^2}{\partial x_i \partial x_k} \frac{1}{|r - r_1|}. \]  
(13)

Let \(\Omega^2\) be an eigenvalue of this operator and \(\eta_i\) be the corresponding eigenvector, so that

\[ \hat{L}_{ik} \eta_k \equiv -\frac{e^2}{m_e} \int d^3 r_1 n(r_1) \eta_k(r_1) \frac{\partial^2}{\partial x_i \partial x_k} \frac{1}{|r - r_1|} = \Omega^2 \eta_i, \]  
(14)

with a normalization condition,

\[ \int n(r) \eta_i^*(r) \eta_i(r) d^3 r = 1. \]  
(15)

We can then rewrite Eq. (10) in the form

\[ m_e \left( 9 \omega_0^2 - \Omega^2 \right) \xi_{(3)i}(r) + m_e \left( \Omega^2 \delta_{ik} - \hat{L}_{ik} \right) \xi_{(3)k}(r) 
+ \frac{e^2}{24} \xi_{(1)k}(r) \xi_{(1)i}(r) \xi_{(1)m}(r) \int d^3 r_1 N(r_1) \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|} = 0. \]  
(16)

If \(9 \omega_0^2\) is close to \(\Omega^2\) and the nonlinear force is sufficiently small, then equation Eq. (16) implies that \(m_e (\Omega^2 \delta_{ik} - \hat{L}_{ik}) \xi_{(3)k}(r) \approx 0\). In other words, \(\xi_{(3)i}(r)\) must be an eigenvector of
the operator \( \hat{L} \) up to an overall multiplicative factor, i.e. \( \xi_{(3)i}(r) \approx C \eta_i \) with \( C \) a constant. In order to find the constant \( C \), we multiply Eq. (16) by \( n(r)\eta_i^*(r) \) and integrate the result over the volume. Since \( \hat{L} \) is a self-adjoint operator, this procedure annihilates the term involving \( (\Omega^2 \delta_{ik} - \hat{L}_{ik}) \). Solving for \( C \) gives

\[
C = - \frac{e^2}{24m_e(9\omega_0^2 - \Omega^2)} \int d^3r n(r)\eta_i^*(r)\xi_{(1)k}(r)\xi_{(1)l}(r)\xi_{(1)m}(r) \\
\times \int d^3r_1 N(r_1) \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|}.
\]  

(17)

As already noted, only the deviation \( \delta N \) from the uniform density profile contributes to the inner integral in Eq. (17), which allows us to disregard small non-uniformity in the quantity \( n(r)\eta_i^*(r)\xi_{(1)k}(r)\xi_{(1)l}(r)\xi_{(1)m}(r) \) within the cold electron core and treat this quantity as a constant. We then obtain

\[
C = - \frac{e^2 n\eta_i^* \xi_{(1)k} \xi_{(1)l} \xi_{(1)m}}{24m_e(9\omega_0^2 - \Omega^2)} \int d^3r \int d^3r_1 \delta N(r_1) \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|},
\]

where \( \xi_{(1)i} \) is related to the laser electric field by Eq. (12),

\[
\xi_{(1)i} = - \frac{eE_0}{m_e} \epsilon_i \frac{\omega_0}{\omega_i^2 - \Omega^2},
\]

(19)

and the outer integration needs to be performed over the spherical volume of the electron core.

It is apparent that, for the nearly spherical cluster, \( \eta_i \) has to be parallel to the laser electric field. We denote this direction with a subscript \( s \). Equations (18) and (19) together with the definition of \( \xi_{(3)i} \) and the normalization condition of Eq. (15) lead to

\[
D_{(3)s} = -e \int d^3r n\xi_{(3)s} = \frac{ne^3 \epsilon_s}{24m_e(9\omega_0^2 - \Omega^2)} \left[ \frac{eE_0}{m_e(\omega_0^2 - \Omega^2)} \right]^3 \\
\times \int d^3r \int d^3r_1 N(r_1) \epsilon_i \epsilon_k \epsilon_l \epsilon_m \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|}.
\]

(20)

In order to evaluate the integral on the right-hand side of this expression, we use a Legendre
polynomial expansion for \( N(r_1) \) in angular variable:

\[
N(r_1) = \sum_{l=0}^{\infty} N_l(r_1) P_l(\cos \theta_1), \tag{21}
\]

where \( \theta_1 \) is the angle between the vectors \( r_1 \) and \( \epsilon_i \). We find that only \( N_0, N_2, \) and \( N_4 \) contribute to the integral, and the result of the integration is

\[
\int d^3r \int d^3r_1 N(r_1) \epsilon_i \epsilon_k \epsilon_l \epsilon_m \frac{\partial^4}{\partial x_i \partial x_k \partial x_l \partial x_m} \frac{1}{|r - r_1|} = -\frac{16\pi^2}{5} \left[ r^2 \frac{\partial N_0}{\partial r} + \frac{4}{7r} \frac{\partial}{\partial r} (r^3 N_2) - \frac{8}{63r} \frac{\partial}{\partial r} \left( \frac{1}{r} \int_r^\infty dr \frac{N_4}{r^3} \right) \right]_{r=R_e}, \tag{22}
\]

where \( R_e \) is the radius of the cold electron core.

The total power radiated by an oscillating dipole with the amplitude defined by Eq. (20) is given by:

\[
P = \frac{e^2 \omega_p^4}{3c^3} \left[ \frac{3\pi}{10} \frac{\omega_0^2}{(9\omega_0^2 - \Omega^2)} m_e^2 R_e^3 (\omega_0^2 - \Omega^2) \right]^2 \times \left[ r^5 \frac{\partial N_0}{\partial r} + \frac{4r^2}{7} \frac{\partial}{\partial r} (r^3 N_2) - \frac{8r^2}{63r} \frac{\partial}{\partial r} \left( \frac{1}{r} \int_r^\infty dr \frac{N_4}{r^3} \right) \right]^2_{r=R_e}. \tag{23}
\]

IV. NUMERICAL MODEL

We use particle-in-cell code to simulate electron and ion dynamics self-consistently. The assumptions that the laser wavelength is much larger than the cluster size and that the motion of the particles is non-relativistic make the problem electrostatic. The laser electric field is introduced in the simulation as a spatially uniform function of time; the magnetic field is neglected. We assume that the laser is linearly polarized and that the cluster is axisymmetric with the axis of symmetry along the laser electric field. The code employs cylindrical coordinates.

Our numerical model has three important distinctions from earlier 2D simulations performed in Cartesian coordinates [11]. First, we address a 3D physical problem that is relevant to spherical clusters as opposed to rod-like clusters. The electrostatic potential falls off faster with distance for a spherical cluster, which changes the behavior of the extracted electrons significantly. Also, the electron eigenmodes are different for spherical and rod-like clusters.
Second, the electrostatic approximation makes our code faster than the earlier electromagnetic code since the size of our simulation box is only a small fraction of the laser wavelength. In experiments, the clusters are about 100 times smaller than the wavelength \[2\]. Our simulation box is typically five times larger than the cluster radius, which saves computation time while capturing most of relevant physics. Third, our code does not make use of periodic boundary conditions, which is convenient for modelling isolated clusters.

The macro-particles in our axisymmetric code are charged rings. Each particle has only radial and axial components of the velocity. The azimuthal component of the velocity is zero because of the axial symmetry. The electric field consists of two parts: the external (laser) field, which is a given function of time, and the Coulomb part. The Coulomb electric field is calculated by transferring the charge density onto the grid and then solving the Poisson’s equation using the multigrid method \[12, 13\]. The boundary condition for an isolated cluster is such that the electrostatic potential vanishes at infinity. Since our simulation box is finite, we need to find the value of the potential at the edge of the simulation box in order to formulate the boundary value problem for Poisson’s equation. Since the box is larger than the size of the cluster we use total charge and the dipole moment of the particles in the box to calculate the potential at the boundary. By doing so, we reduce the error in the boundary condition from the order of \(R/L\) to the order of \((R/L)^3\), where \(L\) is the size of the box and \(R\) is the cluster radius.

We assume that the cluster is fully ionized and located at the center of the simulation box (see Fig. 2). If an extracted electron reaches the edge of the simulation box, we remove it from the simulation. A macroparticle away from the axis represents more real particles than a macroparticle in the central region. When a large macroparticle moves close to the axis, we split it into several macroparticles in order to keep the number of macroparticles per cell much larger than unity.

In most of the simulations presented in this paper, 12000 grid points correspond to one laser wavelength e.g. 800nm. The initial cluster radius in a typical run is 30-50 grid points. Electron core can have a smaller radius than the ion background. A typical size of the simulation box is \(256 \times 256\) grid points. The total number of particles during a run varies from hundreds of thousands to several millions. The simulation of 10-20 laser periods takes less than half an hour on a 2.8 GHz Pentium 4 PC.

In order to test the code, we use problems that have analytical solutions and make the
quantitative comparison, as described below in subsections IV A and IV B.

A. Free core oscillations in uniform and nonuniform clusters

The first example is free oscillations of the electron core with radius \( R_e = \frac{3}{7} R \) inside a non-neutral cluster with radius \( R \). We excite the dipole mode by displacing the core with respect to the equilibrium by \( d = \frac{1}{14} R \). We perform simulations for a uniform and nonuniform ion density profiles. In the case of the uniform density, the frequency of the oscillations is \( \Omega_0 = \omega_{p0}/\sqrt{3} \). In the case of the nonuniform density profile, the frequency of the dipole mode \( \Omega \) is calculated using standard perturbation theory and is given by:

\[
\frac{\Omega - \Omega_0}{\Omega_0} \approx \frac{1}{2} \left\langle \frac{\delta N}{N_0} \right\rangle,
\]

where \( \delta N \) is the deviation from the uniform density \( N_0 \). The angular brackets in this equation stand for the averaging over the core volume. We use a parabolic density profile

\[
N(r) = N_0 + \Delta N \frac{r^2}{R^2},
\]

with \( \Delta N = 0; \pm 0.2 N_0 \). The dipole mode frequency shift for convex (\( \Delta N = 0.2 N_0 \)) and concave (\( \Delta N = -0.2 N_0 \)) profiles is then given by

\[
\frac{\Delta \Omega}{\Omega_0} = \frac{3}{10} \frac{\Delta N}{N_0} \left( \frac{R_e}{R} \right)^2 = \pm 0.011.
\]

We perform the simulations for three different profiles for \( K \approx 208 \) periods of the dipole mode. The frequency resolution is determined by the number of periods: \( \delta \Omega/\Omega_0 = 1/K \approx 4.8 \cdot 10^{-3} \). The time step in the simulation is 0.028 of the period of the dipole mode. The numerical frequency drift in leap-frog method is \( \delta \Omega_{num}/\Omega_0 = \frac{1}{24} \Omega_0^2 \Delta t^2 = 1.2 \cdot 10^{-3} \). The computed spectrum of the cluster dipole moment is shown in Fig. 3. The “nonuniform” shift in the frequency of the eigenmode obtained from the simulation agrees with the theoretical predictions. The code has sufficient resolution to reproduce this shift.
B. Driven core oscillations in a uniform cluster

The second example is the problem of cold electron oscillations inside a uniform spherical cluster in the presence of the external laser field, which also has an analytical solution. We choose the laser pulse with frequency much smaller than the plasma frequency: \( \omega_0 = \omega_p/6 \).

The amplitude of the laser field grows linearly to its maximum value \( (eE_0/mc\omega_0 = 0.063) \) during the first several cycles and then remains constant. The simulation is done for 20 laser field periods. The radius of the electron core is smaller than the radius of the ion sphere and the core does not touch the surface of the cluster during the oscillations. The ion density profile is fixed in the simulation. The top graph in Fig. 4 shows the position of individual particles after 17 periods. One can see that the electrons preserve the initial spherical shape predicted by the theory [8]. The bottom graph in Fig. 4 shows the evolution of the second time derivative \( \ddot{D} \) of the cluster dipole moment \( D \). The dashed lines indicate the amplitude of \( \ddot{D} \) calculated analytically. The response of the uniform cluster has only the frequency of the laser field and a small component at \( \Omega_0 = \omega_p/\sqrt{3} \), which is excited by the leading edge of the laser pulse. The time step in this simulation is \( \frac{1}{10} \) of the Mie mode period \( (2\pi/\Omega_0) \); as a result the computed eigenfrequency is 1.4% larger than \( \Omega_0 \).

V. NONLINEAR RESPONSE OF THE COLD ELECTRON CORE
(SIMULATION RESULTS)

A. Cluster with a fixed ion density profile

This section addresses the case in which the excursion of the extracted electrons is much larger than the cluster radius. In this regime, most of the extracted electrons escape from the cluster and never come back. As a result, the cluster contains only cold electrons that are trapped inside the electrostatic potential well and respond to the laser field adiabatically.

In order to simulate the response of the confined electrons only, we choose an initially spherical cluster in which the radius of the cold electron core is less than the radius of the ion sphere, so that the core is surrounded by a positively charged ion shell. Such a configuration can be created by a very short pump laser pulse that removes the edge electrons before a lower amplitude probe pulse arrives. Starting from this configuration, we simulate the cluster response to the probe pulse.
We first present simulation results for the case of immobile ions with a radial density profile given by Eq. (25), with \( \frac{\Delta N}{N_0} = -0.2 \). The laser pulse is 60 periods long. The amplitude of the laser field rises from zero to its maximum value during the first 6 periods and then remains constant. We perform simulations with different amplitudes and frequencies of the laser field in order to compare the frequency and amplitude scalings of the cluster response, which may be evaluated by Eq. (20) using the ion profile of Eq. (25). The expression obtained may be rewritten in the following normalized form:

\[
\frac{D_{(3)s}}{Q_e R_e} = 0.062 \frac{\Delta N}{N_0} \frac{R}{R_e} \left( \frac{c}{\omega_{p0} R} \right)^3 \left( \frac{e E_0}{m_e c \omega_0} \right)^3 \frac{\omega_{p0}^5 \omega_0^3}{(9 \omega_0^2 - \Omega^2) (\omega_0^2 - \Omega^2)^3},
\]

where \( Q_e \) and \( R_e \) are the charge and the radius of the electron core, \( \omega_{p0} \) is the plasma frequency corresponding to \( N_0 \), and \( \Omega = \Omega_0 + \delta \Omega = 0.989 \cdot \omega_{p0}/\sqrt{3} \) is the eigenfrequency in nonuniform cluster.

At each time step, the code computes the first time derivative \( \dot{D} \) of the cluster dipole moment \( D \). This gives \( \dot{D} \) as a function of time. In order to calculate the cluster response at the third harmonic, we Fourier transform \( \dot{D} \) and filter out all frequencies except those in the third harmonic vicinity, which finally gives us the computed value of \( D_{(3)s} \).

In the first set of simulations, we choose the laser frequency such that \( 3 \omega_0 = 1.06 \cdot \Omega_0 \) and vary the laser amplitude. According to Eq. (27), the cluster response at the third harmonic should cubically depend on amplitude. Figure 5 shows comparison between the simulation results and the theoretical prediction given by Eq. (27). For low laser amplitudes the linear cluster displacement is smaller than the size of the electron core and the results of the simulation are in good agreement with the theory. As the amplitude of the core oscillations increases, the average ion density seen by the electron core decreases, moving the resonance frequency \( \Omega \) away from \( 3 \omega_0 \), which explains the deviation from the theoretical curve in Fig. 5.

In the second set of simulations, we keep the laser field amplitude fixed and we vary the laser frequency in the proximity of the “resonance” frequency \( \Omega/3 \). The results are presented in Fig. 6 that shows the cluster dipole moment at the third harmonic as a function of the laser frequency. We observe very good agreement between simulations and theory in this figure.
B. Enhancement of the 3rd harmonic during cluster expansion

As already stated, the frequency of the dipole mode $\Omega$ is larger than the triple laser frequency at solid state densities. As the cluster expands, its density and the eigenfrequency $\Omega$ decrease, so that $\Omega$ can eventually cross the resonance value $3\omega_0$.

In order to simulate this process, we use the same setup as for induced oscillations in subsection IV B, except we let the ions move. Almost all the electrons remain inside the cluster during the simulation. Only a small fraction of electrons crosses the boundary at the end of the simulation. This configuration allows us to demonstrate the harmonic generation in the expanding cluster due to the cold electrons alone.

We choose the initial conditions in such a way, that the third harmonic is slightly bellow the eigenfrequency ($3\omega_0 = 0.88\Omega$). We first take an artificially large ion mass $m_i = 50000m_e$ in order to slow down cluster expansion and make the effect more pronounced. Figure 7 shows the result of this simulation. We observe that:

a) The electron response is virtually linear at the beginning of the laser pulse.

b) As the cluster expands and the resonance frequency matches the third harmonic frequency, significant energy goes into the third harmonic.

c) The third harmonic is still present later in time, but it is not as strong as during resonant enhancement.

Next, we take the parameters close to the real experiments [14]: $\lambda = 1\mu$, $\omega_p/\omega_0 = 6.6$ and $m_i = 4000m_e$. This corresponds to a deuterium cluster with the density of $N_0 = 4.85 \times 10^{22}\text{cm}^{-3}$. The cluster radius is $R = 4.6\text{nm}$, the core radius is $R_e = 2.5\text{nm}$. The field amplitude is $eE_0/m_e\omega_0 = 0.0315$, which corresponds to the core displacement $d = 0.16\text{nm}$ and the extracted electron excursion $\zeta = 2.4\text{nm}$. The result of the simulation is shown in Fig. 8. A new element that we observe in this simulation is the electron leakage through the ion shell during the relatively fast expansion of the shell. It takes only 15 laser periods for all electrons to leave the expanding cluster, which is evident from the signal depletion in Fig. 8.
C. Role of extracted electrons

How will the situation change if there is no ion shell preventing electrons from leaving the cluster? In order to answer this question, we do another simulation with the same parameters as in section V B, but setting the initial radius of the ion sphere $R$ equal to the radius of the electron core $R_e$, so that the cluster is initially neutral. Here we take $m_i = 50000m_e$ in order to suppress cluster expansion early in time. The result of this simulation is presented in Fig. 9. In comparison with the previous case (Fig. 7), we now observe third harmonic generation at the early stage of the interaction. The underlying reason is that the extracted electrons excite the third harmonic transiently as they move back and forth across the edge, before leaving the cluster for good.

Figure 10 shows that there are electrons that are not confined inside the cluster after 7 laser periods. The ion density is still very close to uniform at this moment. It appears that mostly the extracted electrons produce the nonlinear response at the early stage. Later in time, the extracted particles are nearly gone, but the ion density profile becomes strongly nonuniform because of the cluster expansion. At this stage, the mechanism for harmonic generation is the same as in the case presented in Fig. 7.

In order to verify this interpretation, we perform another simulation with the same parameters, except that we freeze the ions to keep their density uniform. Figure 11 shows that, in this case, the third harmonic arises at the early stage only (due to the extracted electrons).

VI. ACCUMULATION OF HOT ELECTRONS

If the laser electric field is not strong enough to completely remove the extracted electrons from the cluster, then two electron populations will coexist [8]. In order to model the buildup of the hot electron population, we choose an initially neutral spherical cluster of a radius $R = 40\text{nm}$ with an immobile uniform ion background. The laser field is taken to be sub-relativistic with $eE_0/m_e c \omega_0 = 0.025$. The laser wavelength is $\lambda = 0.8\mu$. The corresponding excursion of an extracted electron ($\zeta = eE_0/m_e c \omega_0^2$) is 10% of the cluster radius.

We keep track of the electrons that travel more than 80% of $\zeta$ from the cluster surface at least once. This selects the electrons that gain the largest energy during a laser cycle.
The top graph of Fig. 12 shows the distribution of such electrons over axial momentum $p_z$. The corresponding distribution of the remaining electrons (cold electron core) is shown in the bottom graph of Fig. 12. We observe an order of magnitude difference between the temperatures of these two populations after 20 laser periods. As the fast electrons from the narrow edge layer expand, they spread over the entire cluster volume. They therefore contribute to the electron density inside the cold core, causing the core to expand. This expansion allows some cold electrons to come close enough to the edge to be extracted by the laser field during its next period. As a result, the number of hot electrons grows in time as shown in Fig. 13.

Figure 14 shows partial contributions of the two electron populations to the oscillating dipole moment of the cluster. Early in time, the electron response is linear. As the laser heats more electrons, the response exhibits nonlinearity. The spectrum indicates the presence of the third harmonic. One may think that the third harmonic stems from the stochastic motion of the hot electrons. A more detailed analysis shows that this is not the case. Although there is third harmonic component in the hot electron dipole moment, it is significantly smaller than that of the core electrons. The motion of the core electrons appears to be affected by the nonuniform space-charge of the hot electron halo and it is no longer harmonic as it would be in the absence of the hot population.

VII. SUMMARY

The underlying premise of the present work is that a laser-irradiated cluster generally contains two electron populations, the cold core and the heated halo. Electrons in the core move collectively in step with the applied laser field. On the other hand, the stochastically heated halo electrons move in a chaotic manner.

Our analytic model deals with the oscillatory motion of the core in a potential well provided by the ions of the cluster. The nonlinear response of the core is due to nonuniformity of the ion density, which could be due to some intrinsic nonuniformity or may arise naturally in the process of cluster expansion. When the third harmonic of the laser frequency is in resonance with the Mie frequency, which is the experimentally relevant case, the cluster exhibits enhancement of third harmonic generation. In the present work, this resonant enhancement has been confirmed through PIC simulations.
Our simulation work also confirms the presence of the halo electrons. Here the vacuum heating mechanism is operative. The characteristic energy of halo electrons increases in time due to vacuum heating and the number of halo electrons increases as well. Since the halo is less dense than the cold core and its motion is incoherent albeit nonlinear, the halo contributes less than the core to the third harmonic signal. Also, the timing of the halo response is different.

It remains to be a challenge to observe such two-component electron dynamics experimentally and to quantitatively compare our description of enhanced harmonic generation with experimental data.

Acknowledgments

This work was supported by NSF FOCUS center and by the U.S. Department of Energy Contract No. DE-FG03-96ER-54346.

Figure Captions

FIG. 1: Electron core in a uniform spherical cluster [8]. The core adiabatically responds to the laser field. The electron and ion spheres are concentric in the case when there is no field.

FIG. 2: Simulation setup.

FIG. 3: Spectra of the free electron core oscillations inside a spherical cluster. The figure shows the Fourier amplitude $|\dot{D}_\omega|$ of the cluster dipole moment in a spherical cluster with a radial density profile given by Eq. (25). The curves correspond to concave ($\Delta N = -0.2N_0$), flat ($\Delta N = 0$), and convex ($\Delta N = 0.2N_0$) density profiles.

FIG. 4: Driven core oscillations in a uniform spherical cluster. The top figure shows the cluster configuration after 17 laser periods. The bottom graph shows the time evolution of the second time derivative of the cluster dipole moment.

FIG. 5: Normalized dipole moment of the cluster at the third harmonic ($|D_{(3)s}|/QeRe$) as a function of the laser field amplitude. The dots are the simulation results, the solid line is the theoretical prediction given by Eq. (27). The cluster is nonuniform with a density profile given by Eq. (25), where $\Delta N/N_0 = -0.2$.

FIG. 6: Normalized dipole moment of the cluster at the third harmonic ($|D_{(3)s}|/QeRe$) as a function of the laser frequency. The dots are the simulation results, the solid line is the theoretical prediction given by Eq. (27). The cluster is nonuniform with a density profile given by Eq. (25), where $\Delta N/N_0 = -0.2$.

FIG. 7: Enhancement of the third harmonic during expansion of a cluster with artificially heavy ions ($m_i = 50000m_e$). Initial cluster consists of a cold electron core inside an uncompensated ion shell. The core oscillates inside the expanding cluster. The enhancement happens when the core eigenfrequency matches the third harmonic of the laser frequency. The upper graphs show the incident laser field as a function of time and its spectrum. The lower left graph shows the time evolution of the second time derivative $\ddot{D}$ of the cluster dipole moment $D$. The three lower right graphs show the spectra of $\ddot{D}$ corresponding to the time intervals $A$, $B$, and $C$ shown on the lower left graph.

FIG. 8: The third harmonic generation in an expanding Deuterium cluster. Initial cluster consists of a cold electron core inside an uncompensated ion shell. The upper graphs show the incident laser field as a function of time and its spectrum. The lower graphs show the time evolution of the second time derivative $\ddot{D}$ of the cluster dipole moment $D$ and the corresponding spectrum. As the cluster expands the electrons leak out from the core, which explains the decay of the cluster response in the left bottom graph.
FIG. 9: The third harmonic generation during expansion of initially neutral cluster with artificially heavy ions \((m_i = 50000m_e)\). The upper graphs show the incident laser field as a function of time and its spectrum. The lower graphs show the time evolution of the second time derivative \(\ddot{D}\) of the cluster dipole moment \(D\) and the corresponding spectrum. The harmonic generation at early time is caused by the extracted electrons. After the extracted electrons leave, the third harmonic disappears. Later in time the third harmonic emerges again when its frequency matches the core eigenfrequency.

FIG. 10: Configuration of initially neutral cluster with artificially heavy ions \((m_i = 50000m_e)\) after approximately 7 laser periods.

FIG. 11: Transient generation of the third harmonic by the extracted electrons in an initially neutral and uniform cluster with immobile ions. As the extracted electrons leave, the third harmonic disappears.

FIG. 12: The distribution of the extracted electrons (top graph) and the cold core electrons (bottom graph) over axial momentum \(p_z\) after 20 laser periods.

FIG. 13: Formation of the two component electron distribution. “Cold electrons” are the electrons that never traveled more than 80% of the typical electron excursion \(\zeta\) from the cluster surface. “Hot electrons” are the electrons which have crossed this boundary at least once. “Hot removed electrons” are hot electrons that reached the boundary of the simulation box and were removed from the simulation. The area plot shows the corresponding fraction for each group of the electrons as a function of time.

FIG. 14: Harmonic generation in the presence of the hot electron halo. The left column shows the time dependence of the laser field, the second time derivative \(\ddot{D}\) of the cluster dipole moment and the contribution to \(\ddot{D}\) from the cold and hot electrons. The right column shows the corresponding spectra.
Fig. 1
Fig. 2
Fig. 3
Fig. 4
Fig. 5

Normalized laser field \( \left( \frac{3eE_0}{m_e \omega^2 \rho R} \right) \)
Fig. 6
Fig. 7
Fig. 8
Fig. 9
Fig. 10
Fig. 11
Fig. 12
Fig. 13