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Nafis Ahmad, A. M. Alshehri, and A. Ibrahim
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Cite as: AIP Advances 10, 015312 (2020); doi: 10.1063/1.5118758
Submitted: 29 October 2019 • Accepted: 3 December 2019 • Published Online: 8 January 2020

Nafis Ahmad,a) A. M. Alshehri, and A. Ibrahim

AFFILIATIONS
Department of Physics, College of Science, King Khalid University, P.O. Box 9004, Abha 61413, Saudi Arabia

a)E-mail: nafis.jmi@gmail.com

ABSTRACT
Surface plasmon resonance enhanced high harmonic generation of intense short pulse lasers in xenon clusters is investigated. A laser pre-pulse partially ionizes the cluster atoms, turning them into plasma balls. As the main pulse arrives, plasma electrons execute large amplitude oscillations, creating a space charge field much higher than the laser field at surface plasmon resonance. The bound electrons under this field move out of the ions and return back with large residual energy. On recombination, they produce high harmonics. The number of photons emitted by the xenon clusters per second per frequency interval decreases with the emitted photon frequency.

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I. INTRODUCTION

High harmonic generation (HHG) of an infrared laser is a subject of continued interest due to its potential applications, e.g., X-ray generation, production of attosecond pulses, molecular tomography, etc.1–4 The HHG is understood as follows.5–7 A linearly polarized sub-picosecond laser pulse of ∼1013 to 1016 W/cm² intensity is impinged on a gaseous target. The laser partially tunnel ionizes the atoms, removing the outer electrons. The laser induces a bound electron of an ion to move out. The electron travels under the laser field and returns back to the ion with enhanced energy. As the electron recombines with the ion, it emits a photon of energy equal to the sum of the kinetic energy of the electron and the ionization energy of the ion to go to the next state of ionization.6,8 This way one produces vacuum ultraviolet (VUV) and X rays using the infrared laser. Strelkov9 has suggested a modification of the three step model where the last step of radiative recombination is replaced by two steps, radiationless transition to the autoionizing state (free electron is trapped by the parent ion) and relaxation with extreme ultraviolet emission. High harmonic generation has been reported in monomer gases10,11 and from solid targets.12–14 At higher intensities (I > 1017 W/cm²), an oscillating relativistic plasma mirror produces high harmonics with ultrashort IR pulses.15,16 Gases embedded with clusters offer an efficient nonlinear medium for high harmonic generation using attosecond laser pulses. The surface plasmon resonance very significantly lowers the laser intensity to realize nonlinear effects. L’Huillier and Balcou17 have experimentally observed the 29th harmonic in xenon, the 57th harmonic in argon, and the 135th harmonic in neon by using a Nd:glass laser with a wavelength of 1053 nm and a pulse duration of 1 ps. Ganeev et al.18 have demonstrated the enhancement of a single harmonic (13th, 17th, and 33rd harmonics) of ultrashort pulses in three types of nanoparticle-containing plasmas (In₂O₃, Sn, and Mn₂O₃, respectively). Kumar and Tripathi19 have theoretically investigated the second and third harmonic generation and nonlinear absorption by a short pulse laser in a gas embedded with anharmonic clusters. The second and third harmonic amplitudes increase sharply as the laser frequency ω approaches the surface plasmon resonance frequency, ωpe/√3 (ωpe being the plasma frequency of cluster electrons), and then falls off.

The increase in the photon flux and the maximum photon energy of the harmonic radiation is a continued quest of high harmonic sources. Donnelly et al.20 reported the generation of high order harmonics up to 31st order by using an 825 nm laser of 140 fs pulse duration in the intensity range of 1014–1015 W/cm² from clusters of ∼10⁷ argon atoms produced in a high pressure gas jet. Tisch et al.21 investigated high order harmonic generation from clusters of 1000 xenon atoms formed in a gas jet by using a 160 fs Ti:sapphire laser of 780 nm wavelength. They measured the dependence of the harmonic yield from clusters on the backing pressure to the gas jet.
and showed that Xe clusters may be a more efficient medium for HHG than Xe atoms in the moderate intensity range of $10^{13} \text{--} 10^{14}$ W/cm$^2$.

In this paper, we study the generation of high order harmonics of the Gaussian laser beam from xenon clusters. The laser is assumed to propagate in a gas embedded with clusters with constant spot size, i.e., without convergence or divergence. The clusters on the laser axis experience the highest laser field, while the ones away from the axis experience a radially decreasing field. The laser field tunnel ionizes the atoms of clusters within a few laser periods to a certain stage of ionization, turning the clusters into plasma balls. The response of these plasma balls to the laser results in the oscillation of the electron sphere (with respect to the ion sphere) and creation of a space charge field. At surface plasmon resonance, this space charge field acquires a much larger value than the laser field and induces the bound electrons of cluster ions to tunnel out. After tunneling out, the electrons move in the oscillatory space charge field and return back to the original ions with enhanced energy, producing high frequency photons on recombination. For off-axis tunneling, the probability of electrons from the ions strongly decreases with radial distance as the laser field of the Gaussian beam falls off; hence, the region of high harmonic generation is severely restricted to the vicinity of the laser axis, within a fraction of laser spot size.

In Sec. II, we study the trajectories of electrons in the laser and space charge fields and deduce the frequency of radiation produced on radiative recombination. In Sec. III, we study the probability of emission of photons on recombination of electrons with parent ions and calculate the total number of photons emitted by the cluster per second per unit frequency interval when illuminated by the laser beam. The results are discussed in Sec. IV.

II. TRAJECTORIES OF ELECTRONS IN LASER AND SPACE CHARGE FIELD

Consider a gas embedded with xenon clusters of interior atomic density $n_i$, cluster radius $r_i$, and the number of clusters per unit volume $N$. A laser propagates through the gas with the electric field

$$\vec{E}_L = \hat{x} A_L e^{-i (\omega - k_i z) t},$$

$$A_L^2 = \frac{n_i e^2}{\lambda_0^2} F(t - z/r).$$

For now, we choose $F(t - z/r) = 1$.

The laser turns the clusters into plasma balls, by ionizing the Xe atoms to the charge state $Z$. However, $Z$ would be high on the laser axis and will decrease with $r$, the transverse distance from the laser axis. As a consequence, the interior electron density of clusters $n_e = Z n_i$ would vary with the radial position $r$ of the cluster.

The cluster located between $r$ and $r + dr$ have their electron sphere oscillating with respect to the ion sphere. For electron displacement $\Delta$, the space charge field $\vec{E}_S$ in the overlap region of the ion sphere and the electron sphere is

$$\vec{E}_S = \frac{n_e e^2}{4 \epsilon_0} \Delta.$$  

Solving the equation of motion for cluster electrons under the laser and space charge fields, one obtains

$$\ddot{\Delta} = \frac{e E_L}{m (\omega_L^2 - \omega_{pe}^2/3)},$$

$$\ddot{E}_S = \frac{\omega_{pe}^2/3}{(\omega_s^2 - \omega_{pe}^2/3)} \vec{E}_L,$$

where $\omega_{pe} = (n_i e^2/\epsilon_0 m)^{1/2}$, $-e$ and $m$ are the electronic charge and mass, and $\epsilon_0$ is the free space permittivity. At $\omega_{pe} \sim \sqrt{3} \omega_L$, the oscillating space charge field could be much larger than the laser field. However, $\omega_{pe}$ is a function of radial location $r$ of the clusters; hence, the enhancement factor of the field is different at different $r$.

A Xe ion, charged to the charge state $Z$, is induced by the laser to tunnel out its $Z + 1$st electron, say at time $t_1$. If this electron is captured by the Xe ion in an energy level of $\omega_{ei}$, the time of recapture is $t_2$, and the electrons of cluster ions to tunnel out. After tunneling out, the electrons move in the oscillatory space charge field and return back to the original ions with enhanced energy, producing high frequency photons on recombination. For off-axis tunneling, the tunneling probability of electrons from the ions strongly decreases with radial distance as the laser field of the Gaussian beam falls off; hence, the region of high harmonic generation is severely restricted to the vicinity of the laser axis, within a fraction of laser spot size.

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The total number of photons emitted by the cluster illuminated by the laser beam per second per unit frequency interval is

$$N_{\text{ph}} = \int P_{\text{rec}} P_{\text{emi}} \left(4\pi/3\right) r_1^3 n_0 n_l \omega_l l \, dr. \quad (13)$$

$P_{\text{emi}}$ and $P_{\text{rec}}$ depend on the laser induced space charge field inside the clusters which depends on $r$. Keldysh' has given $P_{\text{emi}}$ (emission probability) as a function of laser field which we replace by the space charge field,

$$P_{\text{emi}} = \frac{1}{I} = (\frac{\pi}{2})^{1/2} \left(\frac{E_l}{\hbar}\right) \left(\frac{|E_0|}{E_A}\right)^{1/2} \exp(-E_A/|E_0|), \quad (14)$$

where $E_A = (4/3) \sqrt{2mE_l^3/\hbar}$ is the atomic field, $E_l$ is the ionization energy, and $\hbar$ is reduced Planck’s constant.

Corkum' has obtained the dipole moment $d(\omega)$ of radiative recombination of the electron with the parent ion. The power radiated by a dipole oscillating at frequency $\omega$ is

$$P_{\text{rad}} = \frac{\hbar \omega}{12 \pi c} |d(\omega)|^2.$$

$P_{\text{rec}}$ may be taken as $nP_{\text{rad}}/\hbar \omega = (\mu_0/12 \pi c \hbar) \omega^3 |d(\omega)|^2$.

Corkum' has plotted $|d(\omega)|^2$ as a function of frequency in atomic units. One atomic unit of dipole moment is $8 \times 10^{-30}$ C m. In the plot, $|d(\omega)|^2$ appears to decrease with frequency as some negative power of frequency ($-\omega^\lambda$). One may take

$$P_{\text{rec}} = P_{\text{rad}}(\omega/\omega_0)^{-1}, \quad (15)$$

where $P_{\text{rec}}$ is a constant,

$$P_{\text{rec}} = (\mu_0/12 \pi c \hbar) \omega_0^3 |d(\omega_0)|^2, \quad (16)$$

where $\omega_0$ is a normalizing frequency. We choose $\omega_0 = 25 \omega_l$ for which Corkum' has given the value of $|d(\omega_0)|^2 = |d(25 \omega_l)|^2$.

III. RADIATIVE RECOMBINATION AND HIGH HARMONIC GENERATION

Let $P_{\text{rec}}(\epsilon)$ or $P_{\text{rec}}(\omega)$ be the probability of capture of the returning electron by the parent Xe ion with the emission of a photon of frequency $\omega$,

$$\hbar \omega = \epsilon + \epsilon_{z_l}, \quad (10)$$

where $\epsilon_{z_l}$ is the ionization energy of the Xe ion in the charge state $Z$ for the release of the $Z + 1$ electron. The probability of emission of the $Z + 1$ electron under the space charge field at time $t_i$ may be taken as $P_{\text{emi}}(t_i)$ or $P_{\text{emi}}(\omega)$ as $\omega$ is related to $\epsilon$ which depend on $t_i$.

The number of photons of frequency $\omega$ emitted by one cluster in one laser period is

$$= P_{\text{rec}} P_{\text{emi}} \left(4\pi/3\right) r_1^3 n_0. \quad (11)$$

The number of photons emitted by the clusters lying over a ring of radius $r$ and width $dr$ and length along $z = l$ per second is

$$= P_{\text{rec}} P_{\text{emi}} \left(4\pi/3\right) r_1^3 n_0 n_l (2\pi r dr) / T, \quad (12)$$

where $T = 2\pi/\omega_l$ and $l$ is the length of the laser illuminated plasma channel.

![FIG. 1](image1.png) Emitted photon frequency ($\omega$) as a function of $\omega_{0}/\omega_{l}$ for the following parameters: $eA/m_{0}c = 0.01$ (corresponding to a laser intensity of $\sim 3 \times 10^{14} \text{ W/cm}^2$ for a wavelength of $1.064 \mu\text{m}$), $3\omega_{c}/\omega_{l} = 0.9$, and $E_{l} = 71.8 \text{ eV}$.

![FIG. 2](image2.png) Total number of photons ($N_{\omega_0}$) emitted per second per unit frequency interval by the xenon cluster as a function of $\omega_{0}/\omega_{l}$ for typical parameters: $|E_{0}|/\omega_{l} = 10^{11} \text{ V/m}$ (corresponding to $|E_{0}|/\omega_{l} = 2 \times 10^{10} \text{ V/m}$ at $\omega_{l} = 0.9 \omega_{0}/\sqrt{3}$), $\omega_{l} = 1.77 \times 10^{10} \text{ rad/s}$, $r_{c} = 5 \text{ nm}$, $n_{0} = 3 \times 10^{22} \text{ m}^{-3}$, $n_{l} = 10^{24} \text{ m}^{-3}$, $l = 200 \text{ nm}$, and $t_{0} = 10 \mu\text{m}$.
Using $P_{\text{ion}}$ and $P_{\text{em}}$ from Eqs. (14) and (15), we evaluate the integral in Eq. (13) after expressing frequency in terms of the electron energy that depends on the space charge field and hence on the radial location of the cluster, $r$.

For $|E_{\text{i},\omega}| = 10^{15}$ V/m (corresponding to $|E_{\text{L},\omega}| = 2 \times 10^{10}$ V/m at $\omega_L = 0.9 \omega_0/\sqrt{3}$ and laser intensity = $0.53 \times 10^{14}$ W/cm$^2$), $E_{\text{i}} = 7.18$ eV, $\omega_0 = 1.77 \times 10^{15}$ rad/s, $r_c = 5$ nm, $n_c = 3 \times 10^{21}$ m$^{-3}$, $n_a = 10^{28}$ m$^{-3}$, $l = 200$ µm, and $r_0 = 10$ µm. In Fig. 2, we have plotted the total number of photons ($N_{\text{ph}}$) emitted per second by xenon clusters as a function of $\omega/\omega_L$. The number of photons decreases as the ratio of emitted photon frequency to laser frequency increases from 25 to 80. The total number of high harmonic photons emitted by the laser pulse is equal to $N_{\text{ph}}$ multiplied by the pulse duration.

IV. DISCUSSION

The space charge field induced by the laser inside the clusters has a very significant influence on high harmonic generation from xenon clusters. For a Gaussian laser beam, the clusters near the laser axis show strong emissions, while the ones a fraction of laser spot size away contribute a little as the tunneling probability of bound electrons from the ions decreases quite sharply. One can generate up to the 80th harmonic by using a 1064 nm laser of $\sim 10^{14}$ W/cm$^2$ intensity in xenon clusters of 5 nm radius. The total number of photons emitted by the xenon clusters per second per frequency interval decreases with the emitted photon frequency.

The present treatment is applicable to other clusters also, e.g., Ar; one must choose appropriate ionization potential and the electron density in cluster turned plasma balls. For argon clusters, one would expect cluster atoms to be ionized to the first state of ionization or second state of ionization. For singly ionized argon ions, the ionization energy (to go to the next state of ionization) is 27.6 eV, while for doubly ionized ions, it is 40.7 eV. These are smaller than the ionization energies used in the above calculations of xenon. As a consequence, one would have a higher probability of tunneling of bound electrons from ions at the same laser intensity. However, the energy of emitted photons will be smaller. We have carried the calculation for argon clusters with doubly ionized cluster ions for the following parameters: $|E_{\text{i},\omega}| = 4.2 \times 10^{10}$ V/m (corresponding to $|E_{\text{i},\omega}| = 8 \times 10^{10}$ V/m at $\omega_0 = 0.9 \omega_0/\sqrt{3}$ and laser intensity = $0.85 \times 10^{13}$ W/cm$^2$), $E_{\text{i}} = 40.7$ eV, $\omega_0 = 1.77 \times 10^{15}$ rad/s, $r_c = 5$ nm, $n_c = 3 \times 10^{22}$ m$^{-3}$, $n_a = 10^{26}$ m$^{-3}$, $l = 200$ µm, and $r_0 = 10$ µm. Figure 3 shows the variation of the number of photons emitted per frequency interval per second by the doubly ionized argon clusters as a function of frequency of emitted photons, which is close to xenon but at lower laser intensity.

ACKNOWLEDGMENTS

The authors express their gratitude to the Deanship of Scientific Research at King Khalid University for funding this work through the Group Research Program under Grant No. G.R.P. 230/40. The authors are also grateful to Professor V. K. Tripathi, IIT Delhi, for the helpful discussions.

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