Laser and Particle Beams (2016), *34*, 171–177.
©Cambridge University Press, 2016 0263-0346/16
doi:10.1017/S0263034615000658

Resonant third harmonic generation in clusters with density ripple: Effect of pulse slippage

SHIVANI VIJ,[1] NITI KANT,[2] AND MUNISH AGGARWAL[3]

[1]Department of Applied Sciences, DAV Institute of Engineering and Technology, Jalandhar-144008, India
[2]Department of Physics, Lovely Professional University, Phagwara-144411, India
[3]Department of Physics, Lyallpur Khalsa College of Engineering, Jalandhar-144001, India

(RECEIVED 15 March 2015; ACCEPTED 9 June 2015)

Abstract

A model is presented for the resonant third harmonic generation of short pulse lasers in cluster plasma in the presence of density ripple. Because of ripple in cluster density and plasma electron density outside the cluster, the phase-matching condition for the third harmonic process is satisfied, leading to resonant enhancement of harmonic generation. We explore the impact of laser intensity, cluster size, and collisional frequency of electrons on the efficiency of third harmonic generation. Moreover, since the group velocity of the third harmonic wave is greater than that of the fundamental wave, it causes the slippage of the generated harmonic pulse out of the fundamental laser pulse and its amplitude increases with time.

Keywords: Atomic cluster; Density ripple; Harmonic generation

1. INTRODUCTION

The interaction of intense short pulse lasers with atomic clusters has been a fascinating field of research and has attracted considerable attention in the recent years. The variety of new and interesting nonlinear phenomena have been observed in such interaction (Sprangle *et al.*, 1990; McPherson *et al.*, 1994; Ditmire *et al.*, 1999; Kumarappan *et al.*, 2001; Fomichev *et al.*, 2003; Kim *et al.*, 2003; Zharova *et al.*, 2003; Tiwari & Tripathi, 2006*a*). Among these, higher-order harmonic generation has been an active research area that produces coherent radiation in the extreme ultraviolet (UV) or soft X-ray region (Reintjes *et al.*, 1977; Parra *et al.*, 2000), which can economically be used for routine laboratory work. Hence various authors have developed analytical methods to study harmonic generation from clusters (Krainov & Smirnov, 2002; Tiwari & Tripathi, 2004). The conversion efficiency of fundamental pulses to harmonics is low without a means for phase-matching between them. Several schemes have been implemented by various researchers theoretically as well as experimentally to make harmonic generation in clusters resonant (Fomytskyi *et al.*, 2004; Tiwari & Tripathi, 2006*b*; Shim *et al.*, 2007). Fomytskyi *et al.* (2004) have utilized the Lagrangian concept of a nonlinear oscillator driven by a sinusoidal force to show that there is a strong resonant enhancement of the third harmonic when the applied field frequency is close to one-third of the core eigen frequency. Tiwari and Tripathi (2006*b*) have studied analytically the third harmonic generation in the phase of exploding clusters. Recently Shim *et al.* (2007) have reported the first experimental controlled enhancement of the third harmonic generation from expanding argon gas cluster by pump-probe experiment. In their experiment, the gas jet was heated by the pump to generate clustered plasma, which then allowed to freely expand. A probe then generated third harmonic radiation at controlled time delays. They observed a sharp enhancement of third harmonic generation from cluster expansion at time delay $\Delta t \sim$ 200–300 fs. Hence the resonant harmonic generation in clusters which was studied earlier theoretically (Kim *et al.*, 2003; Fomyts'kyi *et al.*, 2004; Fomichev *et al.*, 2005), gets verified experimentally considering phase matching effects.

The basic mechanisms in laser–cluster interaction are: The laser field ionizes atoms of the cluster that is, a significant number of electrons are removed from the cluster and get free. The inner free electrons are first accelerated out from the cluster and then driven back into it by the combined effects of the incident laser field and the electrostatic field produced by the laser-driven charge separation. While the outer ionized electrons quickly expelled out of the cluster leaving a net positive charge. This positive charge and the high

Address correspondence and reprint requests to: Munish Aggarwal, Department of Physics, Lyallpur Khalsa College of Engineering, Jalandhar-144001, India. E-mail: sonuphy333@gmail.com

excitation energy acquired by the cluster cause it to undergo coulomb explosion and convert it into plasma balls via tunnel ionization. Also the recombination of a free electron with the parent ion leads to harmonic generation.

In our study, we want to enhance the third harmonic generation in clusters by satisfying the phase matching condition with density ripple on cluster density and plasma electron density outside the clusters. The wave vector $\vec{k}_0$ of density ripple acts as a virtual photon of zero quantum energy and momentum $\hbar\ k_0$ which provides additional momentum to the harmonic photon to satisfy the phase matching condition. The oscillatory velocity of the cluster electrons due to fundamental laser field at $(\omega_1, \vec{k}_1)$ beats with laser magnetic field to produce a second harmonic ponderomotive force at $(2\omega_1, 2\vec{k}_1)$. The oscillatory velocity due to this ponderomotive force beats with cluster density ripple $(0, \vec{k}_0)$ to exert a ponderomotive force at $(2\omega_1, 2\vec{k}_1 + \vec{k}_0)$. The oscillatory velocity due to this ponderomotive force at $(2\omega_1, 2\vec{k}_1 + \vec{k}_0)$ couples with cluster electron density oscillations at $(\omega_1, \vec{k}_1)$ to produce a nonlinear current at $(3\omega_1, 3\vec{k}_1 + \vec{k}_0)$, which drives the third harmonic radiation. In Section 2, we have derived the expression for the third harmonic current density. In Section 3, we have solved the wave equation for Gaussian beam in clustered plasma to find the amplitude of the third harmonic field. We have discussed our results in Section 4 and in the last section, conclusion of the present investigation is presented.

2. THIRD HARMONIC CURRENT DENSITY

Consider a clustered gas jet of argon which is formed by adiabatic expansion and hence cooling of gas into a vacuum. Backing pressure, temperature, geometry of nozzle, and other factors are adjusted in such a way that this structured nozzle produces clusters of radius r and space periodic density $n_c = n_{c0}^0 + n_c^0 \exp(\iota k_0 z)$. Here n_{c0}^0 is initial cluster density and n_c^0 is initial ripple density of cluster. Such density ripple in plasma and cluster can be produced by using a machining laser (Pai *et al.*, 2005; Liu & Tripathi, 2008) or circular grating axicon assembly to generate hydrogen and argon plasma waveguides in a cryogenic cluster jet in a space periodic manner (Layer *et al.*, 2007). Wave vector $\vec{k}_0$ of density ripple is considered to be parallel to z-axis. The free electron density inside cluster is n_e, while outside cluster due to intensity dependent ionization rate it varies sinusoidally as $n_0 = n_{00}^0 + n_0^0 \exp(\iota k_0 z)$, where n_{00}^0 is the initial plasma electron density and n_0^0 is the initial ripple density of plasma electrons. Such type of sinusoidal plasma density ripple has already been used by Xia (2014) with cosine component only. A laser beam of frequency ω_1 and wave number k_1 propagates through clustered plasma. The electric and magnetic field of laser beam are given by

$$\vec{E}_1 = \hat{x} A_1(z, t) e^{-i(\omega_1 t - k_1 z)}, \tag{1}$$

$$\vec{B}_1 = c\vec{k}_1 \times \vec{E}_1/\omega_1, \tag{2}$$

where $A_1(z,\ t) = F(z - v_{g1} t)$ is amplitude of electric field vector and $A_1|_{z=0} = A_0$, c is the speed of light in vacuum. The wave number k_1 and group velocity v_{g1} of the laser beam can be expressed as,

$$k_1 = \frac{\omega_1}{c}\left[1 - \frac{\omega_p^2}{\omega_1^2} - \frac{4\pi n_{c0}^0 r^3 \omega_{pe}^2}{3(\omega_1^2(1 + \iota\nu/\omega_1) - \omega_{pe}^2/3)}\right]^{1/2}, \tag{3}$$

$$v_{g1} = \frac{k_1 c^2}{\omega_1} \frac{1}{\left[1 + \dfrac{4\pi n_{c0}^0 r^3 \omega_{pe}^4}{9\left(\omega_1^2(1 + \iota\nu/\omega_1) - \omega_{pe}^2/3\right)^2}\right]}. \tag{4}$$

Here $\omega_p = \sqrt{4\pi n_{00}^0 e^2/m\gamma_0}$ is the frequency of plasma electrons, $\omega_{pe} = \sqrt{4\pi n_e e^2/m\gamma_0}$ is the plasma frequency inside the cluster, ν is the collisional frequency inside the cluster, $-e$ and m are electronic charge and rest mass, respectively, and γ_0 is the average value of relativistic factor, given as $\gamma_0 \approx (1 + a_0^2/2)^{1/2}$, $a_0 = eA_0/m\omega_1 c$, $a_0 < 1$.

When an intense ultra short laser pulse interacts with cluster, the cluster gets ionized. So a highly excited cluster consists of plasma with electrons and multicharged atomic ions. We assume this ionization to be homogeneous across the whole cluster volume so that we can consider the model of ionized cluster as two homogeneously charged spheres: One is positively charged and the other is negatively charged. The center of negatively charged sphere is shifted along the x-axis by a distance x with respect to center of positively charged sphere. The restoring force experienced by the cluster electrons is given as

$$\vec{F}_T = \frac{-4\pi}{3} n_e e^2 \vec{x} - e\vec{E}_1 \tag{5}$$

As the cluster expands, its size increases and average electron density inside it decreases. We consider the case when the plasma frequency ω_{pe} has gone down significantly. From the equation of motion of cluster electrons, their oscillatory velocity can be written as,

$$\vec{v}_{c1} = \frac{-\iota e\omega_1 \vec{E}_1}{m\gamma_0[\omega_1^2(1 + \iota\nu/\omega_1) - \omega_{pe}^2/3]}. \tag{6}$$

Similarly from the equation of motion of plasma electrons, their oscillatory velocity can be written as,

$$\vec{v}_1 = \frac{e\vec{E}_1}{m\gamma_0 \iota\omega_1}. \tag{7}$$

The wave vector $\vec{k}_1$ of the fundamental laser increases more than linearly with the frequency ω_1 hence, $\vec{k}_3 > 3\vec{k}_1$; that is, the momentum $\hbar\vec{k}_3$ of a third harmonic photon is greater than the sum of the momenta of three pump photons $3\hbar k_1$. Hence the additional momentum is required to satisfy the phase matching condition. The density ripple provides this additional momentum to third harmonic photon to make the process resonant. For this the wave number k_0 should be as, $\vec{k}_0 = \vec{k}_3 - 3\vec{k}_1$. The laser exerts a ponderomotive force $\vec{F}_2 = (-e/2c)(\vec{v}_{c1} \times \vec{B}_1)$ on cluster electrons at $(2\omega_1, 2\vec{k}_1)$. This produces an oscillatory velocity,

$$\vec{v}_{c2} = \frac{\omega_1 e^2 k_1 A_1^2}{m^2\gamma_0[\omega_1^2(1+\iota\nu/\omega_1) - \omega_{pe}^2/3]} \frac{e^{-\iota[2\omega_1 t - 2k_1 z]}}{[4\omega_1^2(1+\iota\nu/2\omega_1) - \omega_{pe}^2/3]}\hat{z}. \tag{8}$$

Using $\vec{v}_{c2}$ in the equation of continuity, we obtain the nonlinear density perturbation at $(2k_1 + k_0)$,

$$n^{NL}_{c2\omega,2k_1+k_0} = \frac{\vec{v}_{c2}.(2\vec{k}_1 + \vec{k}_0)n_a}{2\omega_1}, \tag{9}$$

where $n_a = (4\pi/3)n_e n_c r^3$ is the average number of cluster electrons per unit volume.

Using Eq. (8) in Eq. (9) we get,

$$n^{NL}_{c2\omega,2k_1+k_0} = \frac{e^2 k_1(2\vec{k}_1 + \vec{k}_0)n_a A_1^2}{2m^2\gamma_0[\omega_1^2(1+\iota\nu/\omega_1) - \omega_{pe}^2/3]} \frac{e^{-\iota[2\omega_1 t - 2k_1 z]}}{[4\omega_1^2(1+\iota\nu/2\omega_1) - \omega_{pe}^2/3]}. \tag{10}$$

This nonlinear density perturbation at $(2\omega, 2k_1 + k_0)$ causes electrostatic perturbation of potential ϕ_2 as, $\phi_2 = \phi_2 e^{-\iota[2\omega_1 t - (2k_1+k_0)z]}$, which produces a self-consistent space charge field $\vec{E}_2 = -\vec{\nabla}\phi_2$. This space charge field produces linear density perturbation in cluster electrons as,

$$n^{L}_{c2\omega,2k_1+k_0} = \frac{(2k_1+k_0)^2\chi_{c2}\phi_2}{4\pi e}, \tag{11}$$

where $\chi_{c2} = -\dfrac{(4\pi/3)n^0_{c0} r^3\omega_{pe}^2}{(4\omega_1^2(1+\iota\nu/2\omega_1) - \omega_{pe}^2/3)}$.

Also the non-linear density perturbation in plasma electrons is

$$n^{NL}_{p2\omega,2k_1+k_0} = \frac{e^2 k_1(2\vec{k}_1 + \vec{k}_0)n_0 A_1^2 e^{-\iota[2\omega_1 t - 2k_1 z]}}{8m^2\gamma_0\omega_1^4}. \tag{12}$$

Now from Poisson's equation $\nabla^2\phi_2 = 4\pi e(n_{c2\omega,2k_1+k_0} + n_{p2\omega,2k_1+k_0})$, we get

$$\phi_2 = \frac{-4\pi e(n^{NL}_{c2\omega,2k_1+k_0} + n^{NL}_{p2\omega,2k_1+k_0})}{\epsilon_2'(2k_1+k_o)^2}, \tag{13}$$

where $\epsilon_2' = 1 - \omega_p^2/4\omega_1^2 - (4\pi/3)n^0_{c0}r^3\omega_{pe}^2/(4\omega_1^2(1+\iota\nu/2\omega_1) - \omega_{pe}^2/3)$.

Hence the net density of cluster electrons can be written as,

$$\begin{aligned} n_{c2\omega,2k_1+k_0} &= n^{NL}_{c2\omega,2k_1+k_0} + n^{L}_{c2\omega,2k_1+k_0} \\ &= n^{NL}_{c2\omega,2k_1+k_0}\frac{(1-\omega_p^2/4\omega_1^2)}{\epsilon_2'} \\ &\quad + n^{NL}_{p2\omega,2k_1+k_0}\frac{(4\pi/3)n^0_{c0}r^3\omega_{pe}^2}{(4\omega_1^2(1+\iota\nu/2\omega_1) - \omega_{pe}^2/3)\epsilon_2'}, \end{aligned} \tag{14}$$

where $n^{NL}_{c2\omega,2k_1+k_0}$ and $n^{NL}_{p2\omega,2k_1+k_0}$ are given by Eqs. (10) and (12), respectively.

Similarly the net density of plasma electrons will be

$$n_{p2\omega,2k_1+k_0} = n^{NL}_{p2\omega,2k_1+k_0}\left(1 + \frac{\omega_p^2}{4\omega_1^2\epsilon_2'}\right) + n^{NL}_{c2\omega,2k_1+k_0}\left(\frac{\omega_p^2}{4\omega_1^2\epsilon_2'}\right). \tag{15}$$

The second-order oscillatory velocity $\vec{v}_{c2}$ of cluster electrons beats with first-order magnetic field B_1 to exert a ponderomotive force at $(3\omega_1, 3\vec{k}_1)$ that is, $\vec{F}_3 = (-e/2c)(\vec{v}_{c2} \times \vec{B}_1)$. This force produces a third-order oscillatory velocity,

$$\vec{v}_{c3} = \frac{-3\iota\omega_1 e^3 k_1^2 A_1^3}{2m^3\gamma_o[\omega_1^2(1+\iota\nu/\omega_1) - \omega_{pe}^2/3]} \frac{e^{-\iota[3\omega_1 t - 3k_1 z]}}{[4\omega_1^2(1+\iota\nu/2\omega_1) - \omega_{pe}^2/3]} \frac{1}{[9\omega_1^2(1+\iota\nu/3\omega_1) - \omega_{pe}^2/3]}\hat{x}. \tag{16}$$

The third harmonic nonlinear current density due to atomic cluster at $(3\omega_1, 3\vec{k}_1 + \vec{k}_0)$ is,

$$\begin{aligned} \vec{J}^{NL}_{c3} &= -n_a e\vec{v}_{c3} - \frac{n_{c2\omega,2k_1+k_0}e\vec{v}_{c1}}{2} \\ &= \frac{\iota\pi n^0_c n_e r^3\omega_1 e^4 k_1 A_1^3}{m^3\gamma_0\left[\omega_1^2(1+\iota\nu/\omega_1) - \omega_{pe}^2/3\right]} \frac{e^{-\iota[3\omega_1 t - (3k_1+k_0)z]}}{\left[4\omega_1^2(1+\iota\nu/2\omega_1) - \omega_{pe}^2/3\right]} \\ &\quad \left[\frac{2k_1}{\left[9\omega_1^2(1+\iota\nu/3\omega_1) - \omega_{pe}^2/3\right]} + \frac{(2k_1+k_0)(1-\omega_p^2/4\omega_1^2)}{3\gamma_o\left[\omega_1^2(1+\iota\nu/\omega_1) - \omega_{pe}^2/3\right]\epsilon_2'} + \frac{\pi n^0_0(2k_1+k_o)e^2}{3m\gamma_0\omega_1^4\epsilon_2'}\right]\hat{x}. \end{aligned} \tag{17}$$

Similarly we can obtain third harmonic nonlinear current

https://doi.org/10.1017/S0263034615000658 Published online by Cambridge University Press

density due to plasma electrons at $(3\omega_1, 3\vec{k}_1 + \vec{k}_0)$ as,

$$\vec{J}_{p3}^{NL} = \frac{\iota n_0^0 e^4 k_1 A_1^3 e^{-\iota[3\omega_1 t-(3k_1+k_0)z]}}{8m^3\gamma_0\omega_1^5}\left[\frac{k_1}{3} + \frac{(2k_1+k_0)(1+\omega_p^2/4\omega_1^2\epsilon_2')}{2\gamma_o} + \frac{2\pi n_a e^2\omega_1^2}{m\gamma_0[\omega_1^2(1+\iota\nu/\omega_1)-\omega_{pe}^2/3]}\frac{(2k_1+k_0)}{[4\omega_1^2(1+\iota\nu/2\omega_1)-\omega_{pe}^2/3]\epsilon_2'}\right]\hat{x}. \tag{18}$$

The self-consistent field $\vec{E}_3$ of laser imparts an oscillatory velocity $\vec{v}_3^L = e\vec{E}_3/m\gamma_0\iota 3\omega_1$ to plasma electrons at frequency $3\omega_1$ and linear current density at third harmonic due to this field is,

$$\vec{J}_{p3}^{L} = \frac{-n_0^0 e^2\vec{E}_3}{m\gamma_0\iota 3\omega_1}. \tag{19}$$

Hence net current density becomes, $\vec{J}_3 = \vec{J}_{p3}^L + \vec{J}_{p3}^{NL} + \vec{J}_{c3}^{NL}$.

3. THIRD HARMONIC FIELD

The wave equation for third harmonic field E_3 can be derived from Maxwell's equations as,

$$\nabla^2\vec{E}_3 = \frac{4\pi}{c^2}\frac{\partial\vec{J}_3}{\partial t} + \frac{1}{c^2}\frac{\partial^2\vec{E}_3}{\partial t^2} \tag{20}$$

Using $\vec{J}_3$ from Eqs. (17)–(19), the wave equation becomes,

$$\frac{\partial^2\vec{E}_3}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2\vec{E}_3}{\partial t^2} - \frac{4\pi}{c^2}\frac{\partial\vec{J}_{p3}^L}{\partial t} = \alpha A_1^3 e^{-\iota[3\omega_1 t-(3k_1+k_0)z]}\hat{x} \tag{21}$$

where

$$\alpha = \frac{-12\pi\iota\omega_1}{c^2}\left[\frac{\iota\pi n_c^0 n_e r^3\omega_1 e^4 k_1}{m^3\gamma_0(\omega_1^2(1+\iota\nu/\omega_1)-\omega_{pe}^2/3)}\frac{1}{(4\omega_1^2(1+\iota\nu/2\omega_1)-\omega_{pe}^2/3)}\left\{\frac{2k_1}{(9\omega_1^2(1+\iota\nu/3\omega_1)-\omega_{pe}^2/3)} + \frac{(2k_1+k_0)(1-\omega_p^2/4\omega_1^2)}{3\gamma_0(\omega_1^2(1+\iota\nu/\omega_1)-\omega_{pe}^2/3)\epsilon_2'} + \frac{\pi n_0^0(2k_1+k_0)e^2}{3m\gamma_0\omega_1^4\epsilon_2'}\right\} + \frac{\iota n_0^0 e^4 k_1}{8m^3\gamma_0\omega_1^5}\left\{\frac{k_1}{3} + \frac{(2k_1+k_0)(1+\omega_p^2/4\omega_1^2\epsilon_2')}{2\gamma_0} + \frac{2\pi n_a e^2\omega_1^2}{m\gamma_0(\omega_1^2(1+\iota\nu/\omega_1)-\omega_{pe}^2/3)}\frac{(2k_1+k_0)}{(4\omega_1^2(1+\iota\nu/2\omega_1)-\omega_{pe}^2/3)\epsilon_2'}\right\}\right]. \tag{22}$$

We assume the solution of Eq. (21) as

$$\vec{E}_3 = \hat{x}A_3(z,t)e^{-\iota[3\omega_1 t-(3k_1+k_0)z]} \tag{23}$$

Using Eqs. (21) and (23), we obtain

$$\frac{\partial A_3}{\partial z} + \frac{3\omega_1}{c^2k_3}\left(1 - \frac{\omega_p^2}{18\omega_1^2(1+i\nu/3\omega_1)}\right)\frac{\partial A_3}{\partial t} - \frac{2\iota\pi n_c^0 r^3\omega_{pe}^2}{3k_3c^2((1+\iota\nu/3\omega_1)-\omega_{pe}^2/27\omega_1^2)}A_3 = \alpha_3[F(z-v_{g1}t)]^3 \tag{24}$$

where $\alpha_3 = \alpha/2\iota k_3$.

Using the transformations $\zeta = z - v_{g1}t$ and $\eta = z$, we get,

$$\frac{\partial A_3}{\partial\eta} + \beta\frac{\partial A_3}{\partial\zeta} - qA_3 = \alpha_3' A_0\exp\left(\frac{-3\zeta^2}{\zeta_0^2}\right) \tag{25}$$

where

$$\beta = 1 - \frac{v_{g1}\left(1-\omega_p^2/(18\omega_1^2(1+i\nu/3\omega_1))\right)}{v_{g3}\left(1+4\pi n_c^0 r^3/\left(1-27\omega_1^2(1+\iota\nu/3\omega_1)/\omega_{pe}^2\right)^2\right)},$$

$$q = 2\iota\pi n_c^0 r^3\omega_{pe}^2/3k_3c^2((1+\iota\nu/3\omega_1)-\omega_{pe}^2/27\omega_1^2)$$

and $$\alpha'_3 = \left(\frac{-\iota\pi}{4}\right)\left(\frac{\omega_{pe}}{\omega_1}\right)^2\left(\frac{k_1}{k_3}\right)\left(\frac{eA_0}{mc\omega_1}\right)^2\frac{n_c^0 r^3 k_1}{\left((1+\iota\nu/\omega_1)-\omega_{pe}^2/3\omega_1^2\right)\left((1+\iota\nu/2\omega_1)-\omega_{pe}^2/12\omega_1^2\right)}\left[\frac{1}{3((1+\iota\nu/3\omega_1)-\omega_{pe}^2/27\omega_1^2)} + \frac{(1+k_0/2k_1)(1-\omega_p^2/4\omega_1^2)}{\gamma_0\left((1+\iota\nu/\omega_1)-\omega_{pe}^2/3\omega_1^2\right)\epsilon_2'} + \frac{(1+k_0/2k_1)(\omega_p/\omega_1)^2}{2\gamma_0\epsilon_2'}\right] + \left(\frac{-3\iota}{8}\right)\left(\frac{\omega_p}{\omega_1}\right)^2\left(\frac{k_1}{k_3}\right)\left(\frac{eA_o}{mc\omega_1}\right)^2 k_1\left[\frac{1}{6} + \frac{(1+k_0/2k_1)(1+\omega_p^2/4\omega_1^2\epsilon_2')}{2\gamma_0}\right].$$

For Gaussian profile, we can use $F(z-v_{g1}t) = A_o\exp(-k_i\eta)\exp(-\zeta^2/\zeta_0^2)$, where $\zeta_0 = \tau_0 v_{g1}$ is initial laser pulse length and τ_0 is laser pulse duration. Following Liu and Parashar (2007), k_i the absorption coefficent can be written as

$$k_i = \frac{2\pi}{3}\frac{\omega_1}{c}\frac{n_c^0 r^3\omega_{pe}^2\nu/\omega_1^3}{[1-(\omega_{pe}^2/3\omega_1^2)(1+\eta_1/f)^{-3}]^2 + [\nu/\omega_1]^2} \tag{26}$$

where $\eta_1 = \sqrt{(Z_i \nu \tau_0 m)/(2m_i)}(eA_0 t)/(m\omega f r)$

$$1/(1 + \iota\nu/\omega_1 - \omega_{pe}^2/3\omega_1^2)^2.$$

Here Z_i is the charge state of cluster atom, m_i is mass of ion and f is beam width parameter.

From the solution of Eq. (25), normalized third harmonic wave amplitude can be deduced as,

$$\frac{|A_3|}{|A_0|} = \left| \frac{e^{-k_i\eta} e^{q\zeta/\beta} \alpha'_3 e^{q^2\zeta_0^2/12\beta^2} \sqrt{3\pi}\zeta_0}{6\beta} \left[\mathrm{erf}\left(\frac{6\beta\zeta + q\zeta_0^2}{2\sqrt{3}\beta\zeta_0} \right) - \mathrm{erf}\left(\frac{6\beta(\zeta - \beta\eta) + q\zeta_0^2}{2\sqrt{3}\beta\zeta_0} \right) \right] \right|, \tag{27}$$

where $\mathrm{erf}(\phi)$ is error function of argument ϕ. Or

$$\frac{|A_3|}{|A_0|} = \left| \frac{e^{-k_i z'\zeta_0} e^{(q\zeta_0(z'-t')/\beta)} \alpha'_3 e^{q^2\zeta_0^2/12\beta^2} \sqrt{3\pi}\zeta_0}{6\beta} \left[\mathrm{erf}\left(\sqrt{3}(z' - t') + \frac{q\zeta_0}{2\sqrt{3}\beta} \right) - \mathrm{erf}\left(\sqrt{3}((z' - t') - \beta z') + \frac{q\zeta_0}{2\sqrt{3}\beta} \right) \right] \right| \tag{28}$$

where $z' = z/\zeta_0$ and $t' = v_{g1}t/\zeta_0$.

The third harmonic Poynting vector can be written as

$$\vec{P}_3 = \frac{c}{8\pi} \vec{E}_3^{\star} \times \vec{H}_3 = \frac{c^2}{24\pi\omega_1}(3\vec{k}_1 + \vec{k}_0)|A_3|^2. \tag{29}$$

Also the Poynting vector for the fundamental wave can be written as

$$\vec{P}_1 = \frac{c}{8\pi} \vec{E}_1^{\star} \times \vec{H}_1 = \frac{c^2}{8\pi\omega_1} \vec{k}_1 |A_1|^2. \tag{30}$$

From Eqs. (29) and (30), we get efficiency of third harmonic generation as,

$$\left| \frac{\vec{P}_3}{\vec{P}_1} \right| = \frac{1}{3}(3 + k_0/k_1) \left| \frac{\vec{A}_3}{\vec{A}_0} \right|^2 e^{-2\zeta^2/\zeta_0{}^2}. \tag{31}$$

4. NUMERICAL RESULTS AND DISCUSSION

We have solved Eq. (28) numerically for the following set of parameters: Laser frequency $\omega_1 = 2.126 \times 10^{15}$ rad/s, laser intensity $I \approx 10^{17}$ W/cm^2, laser pulse duration $\tau_0 = 100$ fs, plasma frequency $\omega_p = 1.256 \times 10^{14}$, plasma frequency inside the cluster $\omega_{pe} = 7.565 \times 10^{15}$ rad/s, initial ripple density of plasma electrons $n_0^0 = 5 \times 10^{18}$ cm^{-3}, free electron density inside the cluster $n_e \approx 1.8 \times 10^{22}$ and $\nu/\omega_1 = 0.01$.

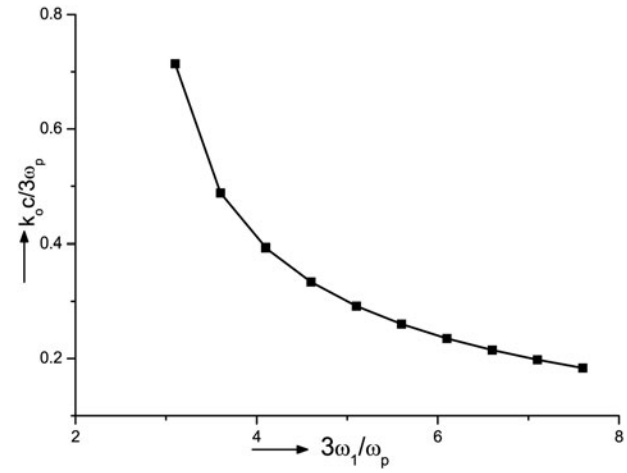

Fig. 1. Variation of normalized wave number of density ripple $k_0 c/\omega_p$ with normalized third harmonic frequency $3\omega_1/\omega_p$ for the following set of parameters, $\omega_{pe} = 7.565 \times 10^{15}$ rad/s, $n_c^0 r^3 = 2 \times 10^{-4}$, and $\omega_p = 1.256 \times 10^{14}$ rad/s.

We consider Ar cluster of size $r = 100$ Å with density $n_c^0 = 2 \times 10^{14}$ cm^{-3}. In Figure 1, we have plotted normalized wave number of density ripple, $k_0 c/\omega_p$ with normalized third harmonic frequency $3\omega_1/\omega_p$ for $n_c^0 r^3 = 2 \times 10^{-4}$. The normalized wave number of density ripple required for phase matching, decreases with the increase in third harmonic frequency. Figure 2 shows the variation of normalized third harmonic wave amplitude $|A_3|/|A_0|$ with normalized distance of propagation z' for different values of intensity parameter $a_0 = eA_0/m\omega_1 c = 0.1$, 0.15, 0.2. The other parameters are $\omega_{pe}/\omega_1 = 3.558$, $n_c^0 r^3 = 2 \times 10^{-4}$, $t' = 4$, $f = 1$ and $\sqrt{Z_i m/m_i}\omega_{pe} t = 60$. The third harmonic amplitude increases sharply at z' =4. For z' =4 in Eq. (28), the exponential term tends to zero and it gives the maximum amplitude of generated third harmonic wave. It is clear from Figure 2 that as we increase the value of a_0 from 0.1 to 0.2, the normalized

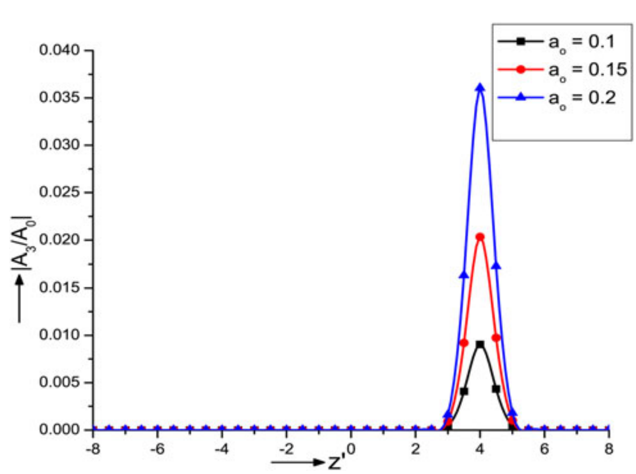

Fig. 2. Variation of normalized third harmonic wave amplitude $|A_3|/|A_0|$ with normalized distance of propagation z' for different values of normalized intensity parameter a_0 with $\omega_{pe}/\omega_1 = 3.558$, $n_c^0 r^3 = 2 \times 10^{-4}$, $\nu/\omega_1 = 0.01$, $\omega_1 = 2.126 \times 10^{15}$ rad/s, $\omega_p = 1.256 \times 10^{14}$ rad/s, $\tau_0 = 100$ fs, and $t' = 4$.

https://doi.org/10.1017/S0263034615000658 Published online by Cambridge University Press

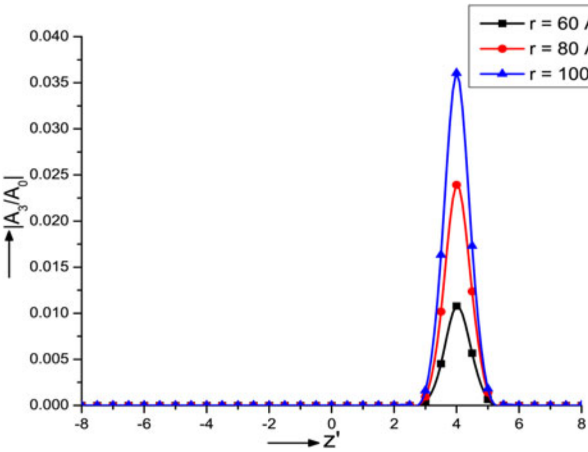

Fig. 3. Variation of normalized third harmonic wave amplitude $|A_3|/|A_0|$ with normalized distance of propagation z' for different values of cluster radius r and fixed value of $a_0 = 0.2$. The other parameters are same as taken in Figure 2.

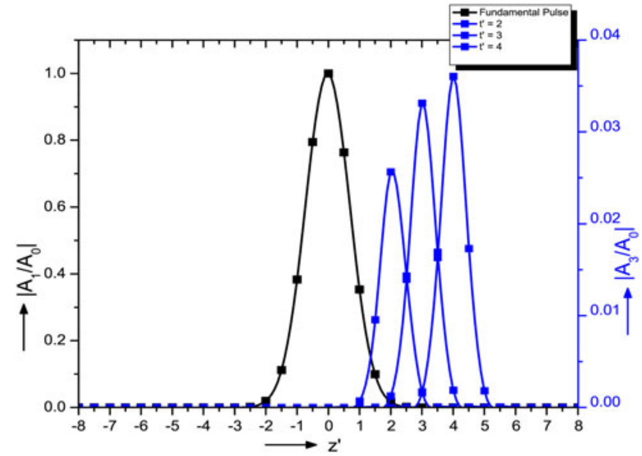

Fig. 5. Variation of normalized third harmonic and fundamental wave amplitude with normalized distance of propagation z' for different value of t' and fixed value of $a_0 = 0.2$. The other parameters are same as taken in Figure 2.

third harmonic amplitude increases from 0.0090 to 0.0362. The normalized amplitude of the third harmonic also depends upon cluster size and increases with increase in the cluster size at $a_0 = 0.2$, as depicted in Figure 3. Since atomic clusters are very efficient at absorbing laser energy, so it would be significant to study the effect of laser absorption on third harmonic wave amplitude. Hence in Figure 4 we plot normalized third harmonic wave amplitude with normalized distance of propagation for different values of normalized collisional frequency $\nu/\omega_1 = 0.01$, 0.02, 0.03. In this plot we observe that if we increase the collision frequency then the laser absorption increases and correspondingly amplitude of third harmonic wave decreases significantly. Therefore, at low value of collisional frequency one can get higher efficiency of third harmonic generation. Figure 5 shows the variation of normalized amplitude of the fundamental wave and third harmonic wave with normalized distance of propagation for different values of normalized time $t' = 2$, 3, and 4 at $a_0 = 0.2$. The other parameters are same as taken in Figure 2. It is clear from Figure 5 that initially the third harmonic wave is generated in the domain of the fundamental laser pulse and as time passes, it starts slipping out of the domain of fundamental laser pulse and its amplitude increases with time. Our results are in contrast in case of plasma where the amplitude of generated harmonic wave saturates with time (Kant & Sharma, 2004; Rajput *et al.*, 2009; Kumar *et al.*, 2013). It is due to the group velocity mismatch between the fundamental laser and third harmonic pulse. The group velocity of third harmonic pulse is greater than that of fundamental laser pulse which causes pulse slippage of third harmonic pulse. Figure 6 shows the variation of normalized third harmonic wave amplitude $|A_3|/|A_0|$ with normalized third harmonic frequency. One can clearly see from the figure that initially the third

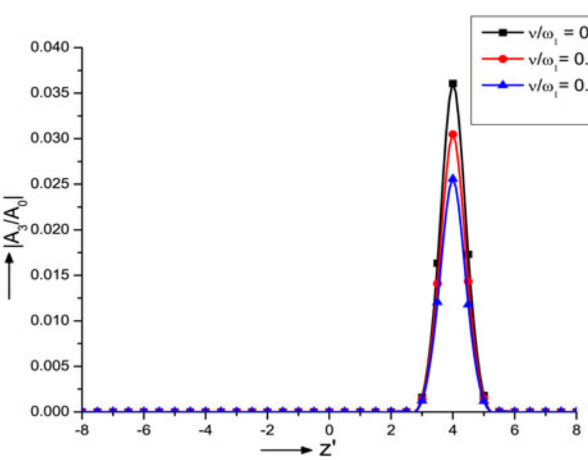

Fig. 4. Variation of normalized third harmonic and fundamental wave amplitude with normalized distance of propagation z' for different value of normalized collisional frequency ν/ω_1 and fixed value of $a_0 = 0.2$. The other parameters are same as taken in Figure 2.

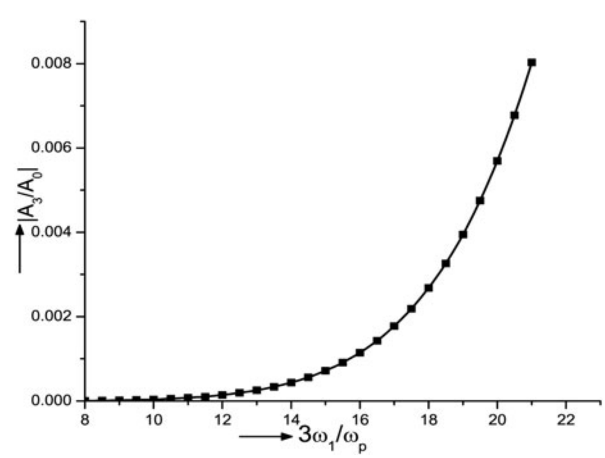

Fig. 6. Variation of normalized third harmonic amplitude $|A_3|/|A_0|$ with normalized third harmonic frequency $3\omega_1/\omega_p$ at $a_0 = 0.2$ and $z' = 4$ for the same parameters as taken in Figure 2.

harmonic amplitude increases gradually with third harmonic frequency and after $3\omega_1/\omega_p = 16$ it increases rapidly.

5. CONCLUSION

Resonant third harmonic generation in clusters with density ripple is analyzed. Density ripple of cluster electrons provides additional momentum to satisfy the required phase matching condition and makes the third harmonic generation process resonant. Third harmonic efficiency depends upon the intensity of the fundamental wave as well as on the size of the cluster. A significant enhancement in the third harmonic efficiency is observed with the increase in the intensity of the fundamental laser pulse and the cluster radius. The increase in collisional frequency of electrons causes more absorption of laser beam by cluster and hence decrease in third harmonic efficiency. Third harmonic efficiency is maximum at low value of collisional frequency. An interesting effect of third harmonic pulse slippage is also seen. Since the group velocity of third harmonic pulse is greater than that of fundamental pulse,the third harmonic pulse moves ahead of the fundamental pulse and its amplitude increases with time. The effect of harmonic generation can serve as a diagnostic tool for the presence of clusters and measurement of their size in cluster experiments. The extreme UV and X-rays emitted in harmonic generation can be used in lithography (Kubiac *et al.*, 1996) and microscopy (Kirz *et al.*, 1995).

ACKNOWLEDGMENT

The authors are grateful to Prof. V. K. Tripathi for his valuable suggestions.

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