



Laser Third Harmonic Generation in CNTs

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Abstract

In the current study, we have conducted a theoretical investigation into the enhanced third harmonic generation of cosh-Gaussian laser beams through their nonlinear interaction with vertically aligned arrays of carbon nanotubes. When a high-power cosh-Gaussian laser interacts with this nanoscale medium, the atoms within it are rapidly ionized, resulting in the formation of plasma. Consequently, the electrons, which possess a lighter mass, may be displaced relative to the ion cylinder. The nonlinearity is amplified due to the electrostatic restoring force acting on the electrons. This laser beam possesses sufficient potential to impart oscillatory velocity to the conducting electrons of the nanotubes, which are efficiently absorbed at the surface plasmon resonance frequency. We have derived an analytic expression for the nonlinear third harmonic current density and the laser's third harmonic field. The graphical profiles plotted indicate the efficient and tunable generation of the laser's third harmonic field, influenced by variations in the beam's decentered parameter, laser beam width, nanotube radii, inter-carbon nanotube separation, initial electric field amplitude of the laser beam, and electron-ion collisional frequency. The resonant field amplitude of the laser's third harmonic is observed when the laser beam frequency reaches $1/\sqrt{2}$ times the electron plasma frequency. As the transverse propagation distance of the laser beam approaches 0.95 times the initial beam width, enhanced third harmonic generation may occur. The decentered parameter of the laser beam significantly influences the field enhancement of the third harmonic without altering the laser beam frequency.

Keywords: Decentered parameter, Electron-ion collisional frequency, Radii, Surface plasmons resonance, Carbon nanotube, Third harmonic, Cosh-Gaussian laser

1. Introduction

Nowadays, interaction of various profile intense laser field with material is a fascinating field of research area owing to possess of interesting nonlinear phenomenon such as soft x-ray generation, charged particle acceleration, electrostatic wave excitation, terahertz radiation generation, self-focusing [1], electron heating, parametric instability and high harmonic generation. In nano dimension medium, the presence of large surface area (large surface to volume ration) promises the enhance the nonlinear interaction of laser beam. The resonant nonlinear phenomenon can be seen with the presence of effective surface plasmon oscillation on nano dimension regime [2]. As the laser beam frequency (electromagnetic radiation) becomes the $1/\sqrt{3}$ times electron plasma frequency[3], resonant nonlinear phenomenon is seen in spherical shape nanoparticles whereas if the laser beam frequency becomes the $1/\sqrt{2}$ times electron plasma frequency, resonant nonlinear phenomenon is seen in cylindrical shape nanoparticles [3] (carbon nanotube). By changing the shape and size of nanoparticles, the nonlinear phenomenon can be varied with without change of interacting electromagnetic radiation field.

Laser second and third harmonic generation scheme is developed by various experimental [4-6]. We have observed the higher order harmonic can be generated by the interaction of a pico second Nd:YAG laser beam with two target containing Cadmium and Palladium. A pump pulsed femtosecond laser with pulse time 50 fs interacts with gold nanoparticles has efficiently generated the second harmonic field [7]. In crystalline of Si nanoparticles, strong second harmonic field were investigated by using a pulse laser. A comparative theoretical and experimental scheme of laser third harmonic generation process has been developed in carbon nanotube. When an intense Gaussian laser beam interacts with cluster, directly converts it into the plasma balls via hydrodynamic expansion or Coulomb explosion. In this self-focusing phenomena takes place and by the result enhanced third harmonic field is generated [8]. An intense short pulse laser having frequency upto $1/\sqrt{3}$ times electron plasma frequency undergoes resonant third harmonic generation in gas jet of clustered plasma. In magnetized nanoclustered plasma, enhanced second harmonic amplitude can be obtained by using the X-mode laser having drift laser drift velocity upto one tenth of velocity of light in vacuum [8-10].

In this theoretical investigation, we study the cosh-Gaussian laser beam third harmonic generation in vertical aligned arrays of carbon nanotube. As the cosh-Gaussian beam interacts with carbon nanotube medium, preformed plasma is taken into place. By this, electrons are displaced with respect ions and a self-consisted space charge will be produced which give rise to restoration force. When the laser beam frequency is become considerably high enough and

if the electrons excursion comparatively become equal to the radius of carbon nanotube, then the average restoration force leads to generation of third harmonic. The schematic of cosh-Gaussian laser beam third harmonic generation process is shown in Fig. 1. The nonlinear interaction of cosh-Gaussian laser beam with carbon nanotube medium is governed in Sec. 2. The analytic derivation scheme of laser third harmonic field is given in Sec. 3. Sec. 4 provides the results and discussion of this theory. The summary and conclusions is given in Sec. 5.

2. Nonlinear Interaction

Herein, we have considered that vertically aligned carbon nanotubes are mounted in a dielectric substrate (say Si). Let each carbon nanotube is identical and the radius of carbon nanotube is taken as r_c . The length of carbon nanotube is grown along the z-direction which perpendicular to the XY-plane and is denoted as L. The separation between consecutive each vertically aligned carbon nanotube is considered as d, N nanotubes are comprised per unit area an XY-plane and the interior electron density is represented as n_{e0} . Let a high power cosh-Gaussian laser beam with frequency ω and wave number k propagates along z-direction and polarized along y-direction in arrays of vertically aligned carbon nanotubes. As the intense field associated with cosh-Gaussian laser beam interacts with arrays of carbon nanotube, the atoms of carbon nanotube are eventually ionized and become into the plasma plume balls. The general electric field profile of cosh-Gaussian laser beam [4] can be written as

$$\vec{E} = \hat{y}E_0 \cosh\left(\frac{yd}{w_0}\right) \exp\left(-\frac{y^2}{w_0^2}\right) e^{-i(\omega t - kz)}, \quad (1)$$

where d is a dimension less parameter associated with hyperbolic cosine term which is known as beam decentered parameter, w_0 represents the cosh-Gaussian laser beam width. As the laser beam interacts with arrays of carbon nanotubes, the electrons of ionized atoms impart oscillatory velocity. Thus, electrons might be segregated from the ion cylinder with excursion $\vec{\Delta}$. Since, we have considered the electron excursion perpendicular to the axis of vertically aligned carbon nanotube

The expression of electron excursion and oscillatory velocity can be solved by using the Eq. as

$$\vec{\Delta} = \frac{e\vec{E}}{m \left[\omega^2 - \frac{\omega_{pe}^2}{2} + i\nu_{ei}\omega \right]}, \quad (2)$$

$$\vec{v} = \frac{(-i\omega)e\vec{E}}{m \left[\omega^2 - \frac{\omega_{pe}^2}{2} + i\nu_{ei}\omega \right]}, \quad (3)$$

The sequence of harmonics upto third order might be arisen owing to presence of anharmonicity and this can be written as

$$\vec{\Delta} = \hat{r}(\Delta_1 + \Delta_2 + \Delta_3), \quad (4)$$

where first order excursion Δ_1 possess the phase variation as $\exp[-i(\omega t - kz)]$ with frequency ω and wave number k , second order excursion Δ_2 possess the phase variation as $\exp[-i2(\omega t - kz)]$ with frequency 2ω and wave number $2k$ and third order excursion Δ_3 possess the phase variation as $\exp[-i3(\omega t - kz)]$ with frequency 3ω and wave number $3k$.

By using the equation of motion and harmonics, we can get the expressions of excursions Δ_1 , Δ_2 and Δ_3 as follows

$$\Delta_1 = \frac{eE}{m \left[\omega^2 - \frac{\omega_{pe}^2}{2} \left(1 - \frac{|\Delta_1|^2}{4\pi r_{c0}^2} \right) + i\nu_{ei}\omega \right]}, \quad (5)$$

$$\Delta_2 = -\frac{\omega_{pe}^2}{4\pi r_{c0}} \frac{\Delta_1^2}{\left[4\omega^2 - \frac{\omega_{pe}^2}{2} + i2\nu_{ei}\omega \right]}, \quad (6)$$

$$\Delta_3 = -\frac{\omega_{pe}^2}{24\pi r_{c0}^2} \frac{\Delta_1^3}{\left[9\omega^2 - \frac{\omega_{pe}^2}{2} + i3\nu_{ei}\omega \right]} \left[1 + \frac{3\omega_{pe}^2}{\pi \left(4\omega^2 - \frac{\omega_{pe}^2}{2} + i2\nu_{ei}\omega \right)} \right]. \quad (7)$$

The first order excursion Δ_1 is very sensitive to the effective surface oscillations of vertically aligned carbon nanotube and its nonlinearity. Since, second and third order excursions are also depend square and cube of first order excursion Δ_1 respectively and hence, this suggests that second and third order excursion is also very sensitive to the effective surface oscillations of vertically aligned carbon nanotube and its nonlinearity.

3. Third Harmonic Generation

If the interaction length is taken longer as compared with inverse of wave numbers $(k_3 - 3k_1)^{-1}$, then we can replace the ∇ operator as $\nabla \rightarrow i3k_1$. On applying the above criterion, we can simplify as

$$\vec{E}_3 = \left(\frac{e^2 \vec{E}^3}{m^3 \times 24\pi r_{c0}^3 \omega_{pe}^6} \right) \times \frac{\omega_{pe}^6}{(8\omega^2 + i2\nu_{ei}\omega) \left(\omega^2 - \frac{\omega_{pe}^2}{2} + i\nu_{ei}\omega \right)^2} \times \left[1 + \frac{6\omega_{pe}^2}{2\pi \left(4\omega^2 - \frac{\omega_{pe}^2}{2} + i2\nu_{ei}\omega \right)} \right], \quad (8)$$

The above Eq. (8) represents the electric field amplitude of third harmonic and or analytic results, we have to normalize this third harmonic electric field with initial electric field amplitude of laser beam E_0 .

4. Results and Discussion

Hyperbolic Cosine-Gaussian laser beam third harmonic generation is analytically studied in arrays of vertically aligned carbon nanotube. When a high power cosh-Gaussian laser beam interacts with nano dimension medium of carbon nanotube, the atoms of this medium are ionized and very quickly converted into the plasma plume balls. This leads to formation of preformed plasma in nano dimension medium. In nano dimension medium, presence of effective surface plasmons oscillation causes to resonant in nonlinear phenomenon (say third harmonic generation). A possible schematic diagram of laser third harmonic generation in carbon nanotube is shown in Fig. 1. We have derived an analytic formalism of cosh-Gaussian laser beam third harmonic generation in arrays of vertically aligned carbon nanotube. The electric field amplitude of third harmonic is directly proportional to the cube of electric field amplitude of laser beam. Herein, we have normalized this third harmonic electric field amplitude with E_0 . In graphical representation, we will see the effect of laser beam frequency, beam decentered parameter, laser beam width, laser beam transverse propagation distance from the origin, carbon nanotube radii, inter carbon nanotube separation and electron ion collisional frequency on the generated third harmonic nonlinear current density and electric field amplitude of laser third harmonic.

Fig. 2 shows the normalized third harmonic current density as a function of normalized cosh-Gaussian laser beam frequency for different values of normalized initial electric field amplitude of laser beam. The nonlinear current density attains two different peaks at normalized laser beam frequency $\omega/\omega_{pe} \sim 0.353$ and $\omega/\omega_{pe} \sim 0.703$ respectively. The amplitude of appeared second peak acquired large peak as compared with first one peak. Since we know that on the surface of carbon nanotube (cylindrical nanoparticle), the effective surface plasmon oscillations occurs as the laser beam frequency become $\frac{1}{\sqrt{2}}$ times of electron plasmon frequency. Here, in second peak we can see that the laser beam frequency is approached near the 0.703 (which is typically most appropriate with the value $\frac{1}{\sqrt{2}}$) times of electron plasma frequency. In this way, we can see that nonlinear resonant phenomenon will be occurred at the second peak and this results the attainment of large amplitude of nonlinear third harmonic current density. It is to be noticed that as one increases the initial electric field amplitude of laser beam, the nonlinear third harmonic current density is enhanced. The initial electric field

amplitude of laser beam might be changed the dynamics of charge particle present in the carbon nanotubes. This leads to more generation of nonlinear third harmonic current density. It was found that the rate of enhancement of nonlinear third harmonic current density at second peak is larger as compared with first one peak with the enhancement of initial laser electric field amplitude.

The variation of the normalized third harmonic current density as a function of normalized cosh-Gaussian laser beam frequency for different values of inter carbon nanotube spacing $b(nm)$ is plotted in Fig. 3. The electric field amplitude of laser third harmonic is decreased with increase in inter carbon nanotube separation. As one increases the inter carbon nanotube separation, the electron density of vertically aligned carbon nanotube will be decreased. This shows that the laser imparts less oscillatory velocity to the electrons of carbon nanotubes and results the less generation of nonlinear third harmonic current density. The rate of variation of third harmonic current density was found very less at the first place of surface plasmon resonance as compared to second place of surface plasmon resonance with the variation of inter carbon nanotube separation.

5. Summary and Conclusions

In this theoretical work we have investigate the cosh-Gaussian laser beam third harmonic generation in vertically aligned carbon nanotube arrays. Our theoretical result is in good agreement with the theoretical results of laser third harmonic generation. Here we have studied the effect of field optimization property of cosh-Gaussian laser beam in third harmonic generation has used the simple Gaussian laser beam for harmonic generation. The results of various graphical plot promise the tunable and controlled generation of third harmonic nonlinear current density and laser electric field third harmonic. The resonant enhanced nonlinear current density and third harmonic field has been seen with the presence of surface plasmons oscillations on carbon nanotube. We have found the particular value of laser beam transvers propagation distance, laser beam frequency and laser beam width on the extreme enhanced condition of third harmonic field. Both electron-ion collisional frequency and beam decentered parameter (associated with hyperbolic cosine term) plays the sensitiveness behaviour on the generated third harmonic field. This laser third harmonic field amplitude can be tuned and controlled by varying the carbon nanotube elements (radii, interatomic separation, electron density and height) and cosh-Gaussian laser beam parameters.

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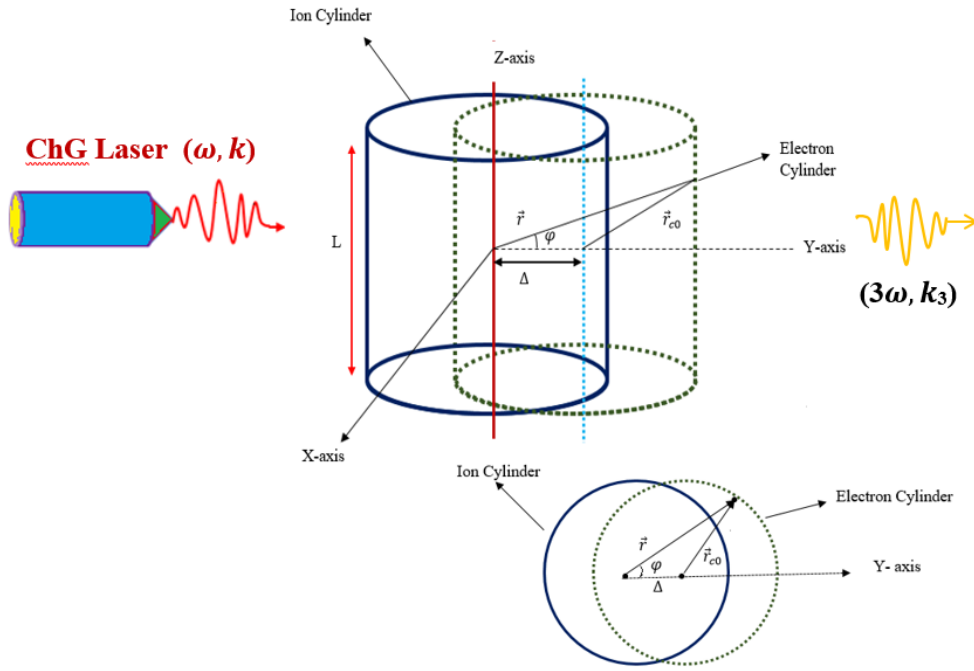


Fig. 1

Fig. 1: Schematic diagram of laser third harmonic generation in carbon nanotube

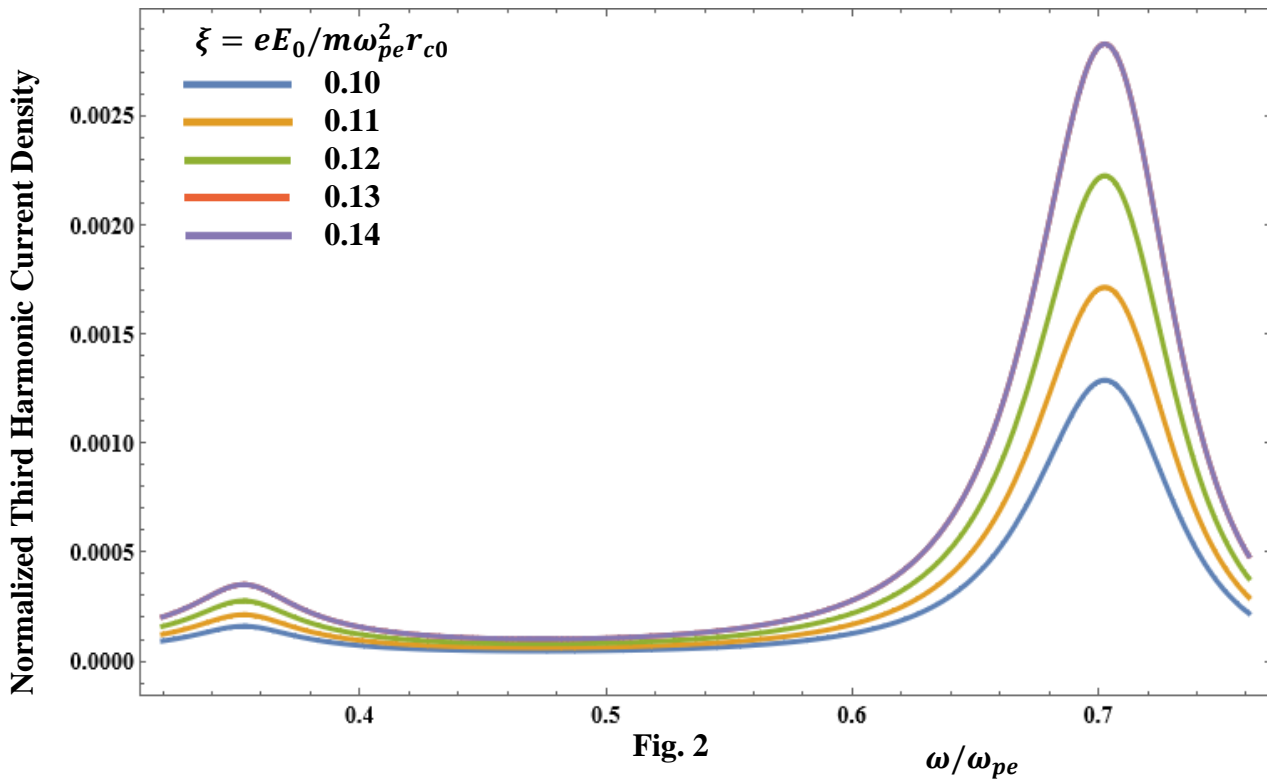


Fig. 2

Fig. 2: Variation of normalized third harmonic nonlinear current density with normalized laser beam frequency for different values of $\xi = eE_{0L}/m\omega_{pe}^2 r_{c0}$ when $d=1$, $n_{ce0}/n_0 = 0.01$, $r_{c0} = 20 \text{ nm}$, $v_{ei}/\omega_{pe} = 0.01$, $y/w_0 = 0.9$

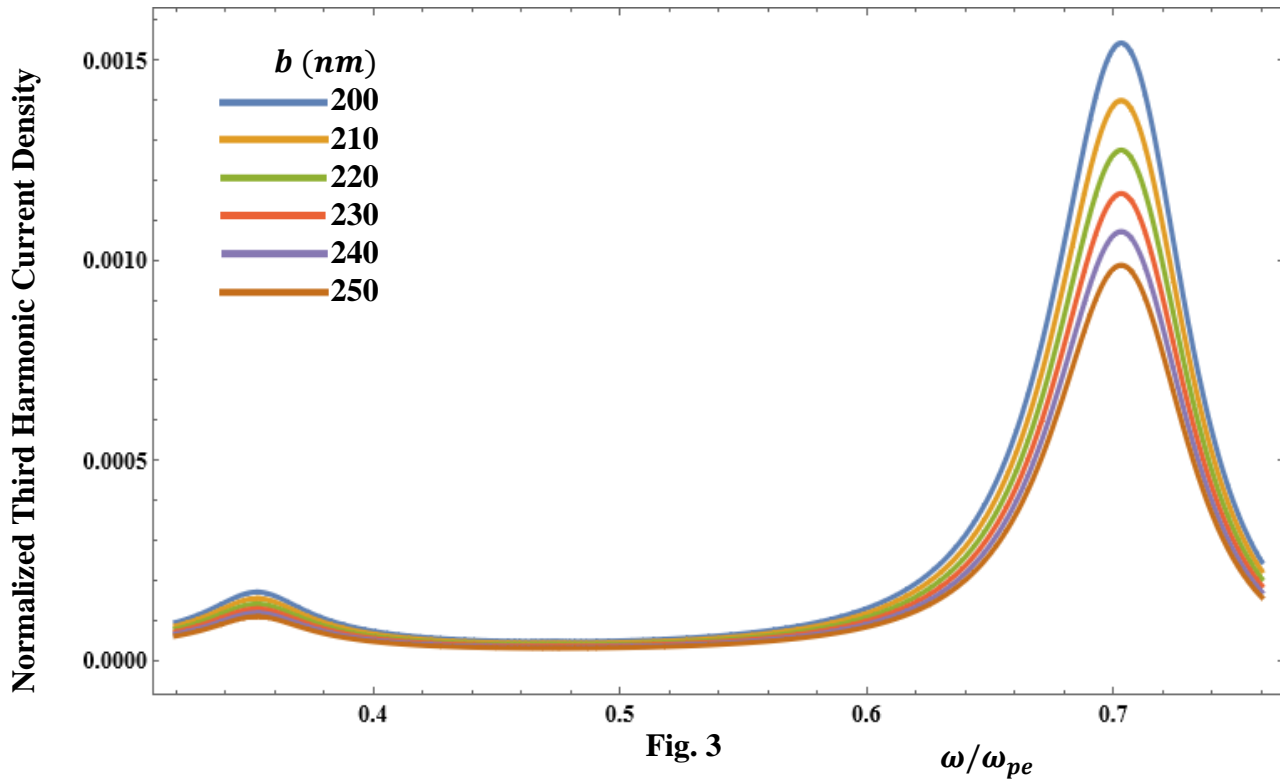


Fig. 3: Variation of normalized third harmonic nonlinear current density with normalized laser beam frequency for different values of inter carbon nanotube separation b when $d=1$, $n_{ce0}/n_0 = 0.01$, $r_{c0} = 20 \text{ nm}$, $v_{ei}/\omega_{pe} = 0.01$, $y/w_0 = 0.9$, $\xi = eE_{0L}/m\omega_{pe}^2 r_{c0} = 0.1$