

# Phase-matched relativistic second harmonic generation in clusters with density ripple



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## ABSTRACT

An intense short-pulse laser obliquely incident on a clustered gas quickly converts the atomic clusters into hot plasma balls. The laser beam produces a second harmonic due to nonlinear response of cluster and plasma electrons. For enhancement of efficiency of second harmonic generation, there should be appropriate phase-matching between the incident laser beam and the generated harmonic. To achieve the required phase-matching, the ripple in cluster density and plasma electron density outside the cluster is introduced. The efficiency of second harmonic generation is sensitive to the angle between ripple wave vector  $k_r$  and the direction of the incident laser beam.

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## 1. Introduction

During the last decade, many efforts have gone into the study of interaction of short intense laser pulses with atomic clusters [1–5]. Clusters are Vander-walls bonded aggregates of atoms or molecules having unique properties compared with atoms and molecules as well as with bulk matter. Due to their unique properties, these act as an efficient medium to absorb energy from an external electric field, such as lasers. The resonant absorption of laser energy by the cluster electrons leads to observation of important nonlinear phenomena like generation of fast electrons and ions [6], production of high charge states [7], neutron production [8], proton acceleration, X-ray generation [9] and harmonic generation [10,11], with high laser intensities up to  $10^{20}$  W/cm<sup>2</sup>. Among these, the phenomenon of harmonic generation from clusters has been interestingly explored by various research groups [12–16] because of its importance as a diagnostic tool in cluster experiments.

To broaden the practical applicability of harmonic generation, it is quite interesting to propose the methods and to investigate the conditions under which higher energy conversion efficiency can be obtained. Many methods and studies realizing this goal have been reported earlier by various researchers [10,11,17–21]. Fomytskyi et al. [10] have developed an analytical model 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 [11] have also developed an analytical formalism to study the third harmonic generation in the phase of exploding clusters. The first experimental observations of controlled enhancement of the third harmonic generation from expanding argon gas cluster was reported by Shim et al. [17]. In their pump-probe experiment, they generated clustered plasma by heating the gas jet using pump, then a probe generated the third harmonic radiation at controlled time delays. A sharp enhancement of this third harmonic radiation was observed at time delay  $\Delta t \sim 200 - 300$  fs. Kundu et al. [18] have analyzed the harmonic emission from short intense infrared laser-driven cluster nanoplasma by means of particle-in-cell simulations. They observed that the low-order harmonic yields were resonantly enhanced when the Mie plasma frequency  $\omega_{Mie} = \sqrt{Q/R^3}$ , (where  $Q$  and  $R$  are the total ionic charge and the cluster radius, respectively) of the expanding cluster resonates with the respective harmonic frequency. Recently Kumar and Tripathi [19] have developed a theoretical model to investigate the nonlinear absorption and harmonic generation of laser in a gas embedded with anharmonic clusters. They observed that anharmonicity in the response of electron clouds of clusters to an intense laser field is responsible for broadened surface plasmon resonance and laser absorption. The second and third harmonic generations, of high amplitude are observed than the one due to ponderomotive nonlinearity. Recently, the authors of this article have investigated the enhancement in the second harmonic generation in Ar cluster by applying

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wiggler magnetic field and observed the maximum second-harmonic power conversion efficiency of 13.3% [20].

The most successful model till date to explain laser-cluster interaction has been the nanoplasma model of Ditmire [22], which can be qualitatively summarized as follows. The laser field ionizes atoms of the cluster causing the cluster electrons to leave their parent atoms (called inner ionization). 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. Some electrons may get expelled out of the cluster (called outer ionization), leaving behind a net positive charge throughout the cluster. Due to this positive charge and the high excitation energy acquired by the cluster electrons, the cluster converts into plasma balls via tunnel ionization. These hot and dense nanometer-sized plasma balls, so called nanoplasmas, have unique and attractive properties, which have been interestingly studied in recent years due to the developments in the field of nanotechnology and nanoparticles.

In this paper, we present a theoretical model to generate second harmonic generation in clusters by an obliquely incident intense short laser pulse in a relativistic regime. In order to satisfy the phase matching condition, the density ripple on cluster density and plasma electron density outside the clusters has been introduced. To date, all the reported theoretical investigations of harmonic generation of an obliquely incident intense short laser pulse were taken from plasma [23–26]. We have considered atomic clusters to generate second harmonic, as they are highly efficient media to absorb energy from the incident laser pulse. Such study can serve as a spectroscopic tool to detect the presence of clusters, in their size measurement and other dynamics during the interaction of laser pulse with a gas jet.

The clustered plasma is a dispersive medium, where refractive index increases with wave frequency. Hence, the wave number,  $k_2$ , of the second harmonic increases more than twice the wave number,  $k_1$ , of the incident laser. Due to mismatch in wave numbers, there will be low efficiency harmonic generation. To increase the efficiency of harmonic generation, the wave vector,  $\vec{k}_o$ , of density ripple which acts as a virtual photon of zero quantum energy and momentum  $\hbar\vec{k}_o$ , provides the additional momentum to the second harmonic photon. The laser imparts an oscillatory velocity  $\vec{v}_1$  to cluster and plasma electrons at  $(\omega_1, \vec{k}_1)$  and exerts a ponderomotive force  $\vec{F}_{2\omega_1}$  on them at  $(2\omega_1, 2\vec{k}_1)$ . This ponderomotive force  $\vec{F}_{2\omega_1}$  and the force due to self-consistent field  $\vec{E}_2$  (which arises due to space-charge oscillations) induce oscillatory velocities  $\vec{v}_{c2}$  and  $\vec{v}_2$  on cluster and plasma electrons respectively. The  $\vec{v}_{c2}$  and  $\vec{v}_2$  couple with cluster electron density and plasma electron density respectively, at  $(0, \vec{k}_o)$ , to produce second harmonic current and density oscillations  $n_2$  at  $(2\omega_1, 2\vec{k}_1 + \vec{k}_o)$ . This second-harmonic current generates second-harmonic radiation. In Section 2, we have derived the expression for the second harmonic current density. In Section 3, we have solved the wave equation for a Gaussian beam in clustered plasma to find the amplitude of the second harmonic field and hence the second harmonic power density. We have discussed and concluded our analysis in Sections 4 and 5 respectively.

## 2. Second-harmonic current density

Consider a preionized, density modulated plasma embedded with a clustered gas jet of argon which is formed by adiabatic expansion, subsequent cooling and condensation of gas into

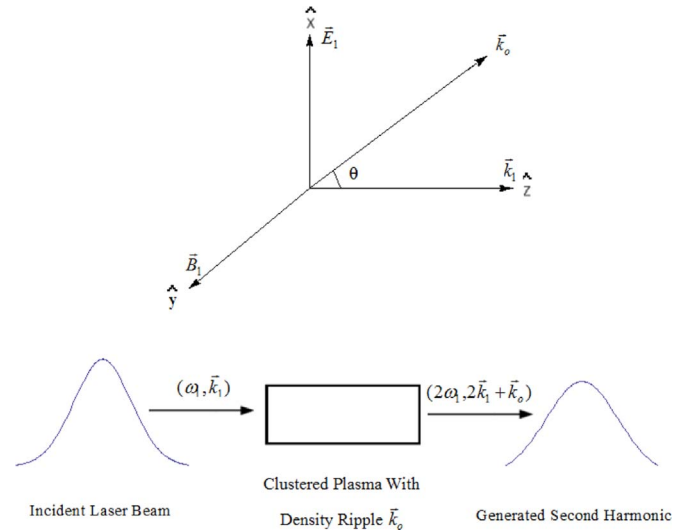


Fig. 1. Schematic of second-harmonic generation process.

vacuum. Geometry of nozzle, backing pressure, temperature and other thermodynamic parameters are adjusted in such a way that this structured nozzle produces clusters of radius  $r$  and space periodic density  $n_c = n_{co}^o + n_c^o \exp(i\vec{k}_o \cdot \vec{r})$ . Here  $n_{co}^o$  and  $n_c^o$  are the initial cluster density and the initial ripple density of cluster respectively. Such density ripple in plasma and cluster can be produced by using a machining laser [27,28] or circular grating axicon assembly to generate hydrogen and argon plasma waveguides in a cryogenic cluster jet in a space periodic manner [29]. One more scheme of plasma density-structure fabrication by laser machining has been reported by Kuo et al. [30]. Wave vector  $\vec{k}_o$  of density ripple is considered to be inclined at an angle  $\theta$  with the  $z$ -axis (Fig. 1). Each cluster has free electron density  $n_e$  inside it, while outside the cluster it contains ripple as  $n_o = n_{oo}^o + n_o^o \exp(i\vec{k}_o \cdot \vec{r})$ . Here  $n_{oo}^o$  is the initial plasma electron density and  $n_o^o$  is the initial ripple density of plasma electrons. Such type of sinusoidal plasma density ripple has been used by Xia [31], with cosine component only.

A laser beam of frequency  $\omega_1$  and wave number  $k_1$  is launched into the clustered gas along  $\hat{z}$  direction. The electric and magnetic fields of the laser beam are given as

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

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

where  $A_1(z, t) = F(z - v_{g1}t)$  is the amplitude of electric field vector and  $A_1|_{z=0} = A_o$ ,  $c$  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,

$$k_1 = \frac{\omega_1}{c} \left[ 1 - \frac{\omega_p^2}{\omega_1^2} - \frac{4\pi n_{co}^o r^3 \omega_{pe}^2}{3(\omega_1^2 - \omega_{pe}^2/3)} \right]^{1/2}, \quad (3)$$

$$v_{g1} = \frac{k_1 c^2}{\omega_1} \left[ \frac{1}{1 + \frac{4\pi n_{co}^o r^3 \omega_{pe}^4}{9(\omega_1^2 - \omega_{pe}^2/3)^2}} \right]. \quad (4)$$

Here  $\omega_p = \sqrt{4\pi n_{oo}^o e^2 / (m\gamma_o)}$  is the frequency of plasma electrons,  $\omega_{pe} = \sqrt{4\pi n_e e^2 / (m\gamma_o)}$  is the cluster electron plasma frequency,  $-e, m$

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