

EFFECT OF VARIATION OF LASER PARAMETERS ON FREQUENCY SHIFT IN TRANSPARENT BULK MEDIA

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ABSTRACT

The paper contains derivation of equations governing the propagation dynamics and parameters affecting the wide band dielectric in the interaction zone. Theoretical investigation of the variation of laser parameters during laser induced breakdown of wide band dielectrics is carried out in the paper. When the dielectric sample is irradiated with ultrashort laser pulses, the interaction results in transfer of pulse energy to electrons, and they get excited to conduction band through multiphoton ionization. Then the electron density increases beyond breakdown limit leading to formation of plasma. The change in refractive index triggers self phase modulation which upshifts laser frequency. The mathematical model illustrates dependence of beam width parameter and normalized laser frequency on propagation distance. This model also efficiently depicts shift in laser frequency, which is valid for higher intensities of the order $10^{17}/\text{cm}^2$.

Keywords: *Dielectrics, Frequency Upshift, Laser Induced Breakdown, WKB Approximation*

I. INTRODUCTION

The advents of femtosecond lasers have paced up the experimental as well as theoretical research in widening the scope of lasers in micromachining to keratoplasty. The study of physics of nonlinear optical processes of the irradiating a dielectric sample with laser pulses of few 10^{15} s can reveal the latent potentials of this interaction. This regime of laser matter interaction has seen extensive experimental advancement in past decades, but lacks theoretical background to explain the observed results.

The conduction band population increases as the laser pulses initiate the photo-excitation of electron from valence band via multiphoton ionization [5]. The electrons produced in conduction band absorb energy from laser pulse and collide with neighboring electron to transfer energy to other electrons, thus helping the electrons to overcome the ionization potential [9]. The electron-electron collision in addition to tunneling increases the electron density beyond the threshold value. This causes evolution of plasma and refractive index decreases causing the modulation of phase, ultimately leading to frequency broadening [11]. Theoretical investigation of the variation of laser parameters during laser induced breakdown of wide band dielectrics is carried out in the paper.

II. THEORETICAL ANALYSIS

2.1 Density Evolution

Consider a large band gap dielectric with energy band gap E_g occupying half space $z > 0$. An ultra-short laser pulse is incident normally on a $z = 0$ surface from free space. The laser field inside the dielectric is given by the equation below

$$E = \hat{x}E_0(t) \exp\left(-\frac{r^2}{r_0^2}\right) \exp(-i\omega t) \exp[-i(\omega t - kz)] \quad (1)$$

where r_0 is the spot size of the laser at $z = 0$. E_0 is related to the incident laser amplitude $E_{oi}(t)$ as $E_0(t) = [2/(1 + n)] E_{oi}$, where n is the refractive index of the dielectric and $k = (\omega/c)n$. For $z > 0$, we have $|\vec{E}| = \hat{x}Ae^{-i\phi}$ where $\phi(z, t) = \omega t - kz$ is the fast phase of the wave and $A(z, r, t)$ is a slowly varying complex amplitude of the wave. ($\omega = \partial\phi/\partial t$ and $k = -\partial\phi/\partial z$)

Li et al. (2000) have concluded from their experimental results that the net electron density evolution equation can be written as

$$\partial n_e / \partial t = W + \alpha n_e + \beta n_e \quad (2)$$

where $\alpha_0(Te)I$ distinguishes the rate of the collisional e-h production and $I(t)$ is the laser intensity of the laser pulse. $W = 2/9\pi (E_g/\hbar) (mE_g/\hbar^2)^{3/2} (|\vec{E}|/E_A)^{5/2} \exp(-E_A\pi/2|E|)$ and E_A is the strength of the atomic field. At enormously high laser intensities ($\gg 10^{17} \text{W/cm}^2$), when the electric field of laser $|E|$ exceeds the coulomb field of the atom E_A , the electron orbiting around tunnels out (rendered as free electron) and is the major contributor to the plasma formation. The evolution of plasma frequency ω_p ($\omega_p^2 = 4\pi me^2 n_o$, where n_o is the density) varies with time [4] as given below

$$\frac{\partial \omega_p^2}{\partial z} = \gamma (\omega_{pm}^2 - \omega_p^2) \quad (3)$$

III. LASER FREQUENCY UPSHIFT

Considering Gildenburg et al. (2002), the wave equation governing the laser pulse inside the dielectric, in the limit of $v_e^2 \ll \omega^2$ (assuming $\nabla E \approx 0$ is valid when $\omega_p^2 \ll \omega^2$) is given by

$$\nabla^2 \vec{E} - \frac{\epsilon_L}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{\omega_p^2}{c^2} \left(1 + \frac{i\nu'}{\omega}\right) \vec{E} \quad (4)$$

The dispersion relation for a semiconductor of dielectric constant ϵ_L is found by using equations (2) and (4) in WKB approximation, given by $\epsilon_L \omega^2 = \omega_{po}^2 + k^2 c^2$ and we get

$$2ik \frac{\partial A}{\partial z} + iA \frac{\partial k}{\partial z} + \nabla_{\perp}^2 A + \frac{2iA\epsilon_L}{c^2} \frac{\partial \omega}{\partial t} + \frac{iA\epsilon_L}{c^2} \frac{\partial k}{\partial t} = \frac{A}{c^2} (\omega_p^2 - \omega_{po}^2) + \frac{i\nu' \omega_p^2 A}{\omega c^2} \quad (5)$$

Differentiating the above equation with respect to t (using $\partial k/\partial t = -\partial\omega/\partial z$) and taking group velocity of the form $v_g = (c/\varepsilon_L)(\varepsilon_L - \omega_{po}^2/\omega^2)^{1/2}$ for under-dense plasma $\omega_p^2 \ll \omega^2$, we get

$$\frac{\partial \omega^2}{\partial t} + v_g \frac{\partial \omega^2}{\partial z} = \frac{1}{\varepsilon_L} \frac{\partial \omega_{po}^2}{\partial t} \quad (6)$$

Now as per the analysis introduced by Liu and Tripathi (2000), for an initially Gaussian beam, results in the beam width parameter [7]

$$\frac{\partial^2 f}{\partial \xi^2} = \frac{1}{f^3 \Omega^2 \varepsilon_L} - \frac{1}{\varepsilon_L \Omega} \frac{\partial f}{\partial \xi} \frac{\partial \Omega}{\partial \xi} - \frac{1}{\varepsilon_L} \frac{\Omega_{p2}^2}{\Omega^2} \eta^2 f \quad (7)$$

Introducing dimensionless quantities for paraxial ray approximation [11] $\xi = z'/\omega_o r_o^2$, $\xi = z'/R_d$, where R_d

is defined as the Rayleigh length ($R_d = k r_o^2$), $\tau' = \gamma_o t'$, η is the normalized propagation distance given by

$$\eta = \omega_o r_o / c, \quad \Omega_{pm} = \omega_{pm} / \omega_o, \quad \Omega_{p2} = \omega_{p2} / \omega_o, \quad \text{and} \quad \Omega = \omega / \omega_o \quad \text{where} \quad \gamma_o = (\pi/2)^{1/2} (I_o / \hbar) (1/g^2)$$

and the value of g is given by $g = E_A / E_{oo}$.

Applying transformation rules on the equations we finally get the equations governing the laser frequency

$$\frac{\partial \Omega^2}{\partial \xi} = \frac{\gamma \eta^2}{\varepsilon_L^{1/2}} (\Omega_{pm}^2 - \Omega_{po}^2) \quad (8)$$

$$\frac{\partial \Omega_{po}^2}{\partial \tau'} = V (\Omega_{pm}^2 - \Omega_{po}^2) \quad (9)$$

$$\frac{\partial \Omega_{p2}^2}{\partial \tau'} = -\Omega_{p2}^2 V - \frac{\gamma}{\gamma_o} (\Omega_{pm}^2 - \Omega_{po}^2) \quad (10)$$

where $V = (1/f^2) \exp(-gf)$, $\gamma = (\pi/2)^{1/2} (I_o / \hbar) (|E|/E_A)^2 \exp(-E_A/|E|)$. Then the equations

(7),(8),(9), and (10) are solved numerically as well as plotted using MATLAB 2013a and ORIGIN 9.0.

IV. FIGURES

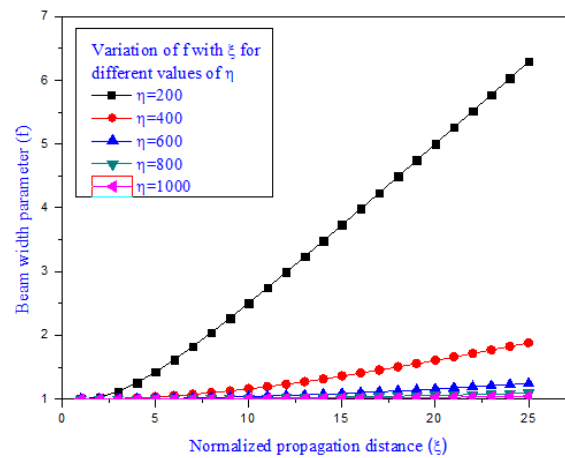


Figure 1: Variation of beam width parameter (f) with normalized propagation distance (ξ) for different values of normalized spot size (η) of laser beam. The other parameters are $\Omega_{pm}^2 = 0.005$ and $\varepsilon_L = 1$.

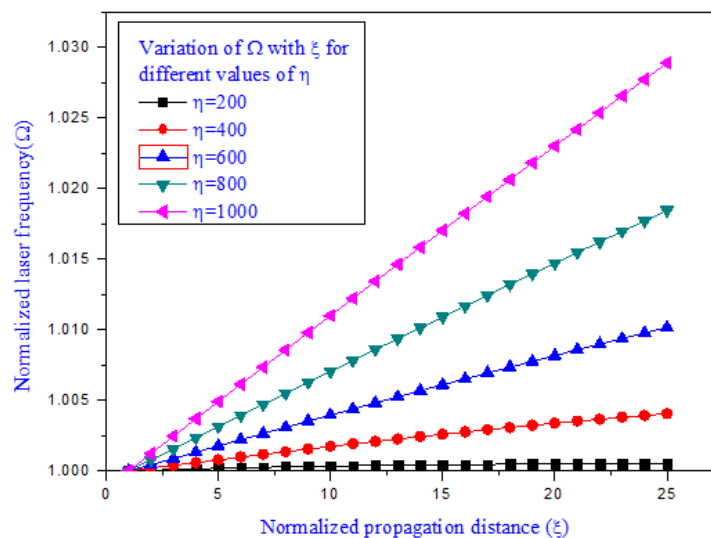


Figure 2: Variation of normalized laser frequency (Ω) with normalized propagation distance (ξ) for different values of normalized spot size (η) of laser beam. The other parameters are $\Omega_{pm}^2 = 0.005$ and $\varepsilon_L = 1$.

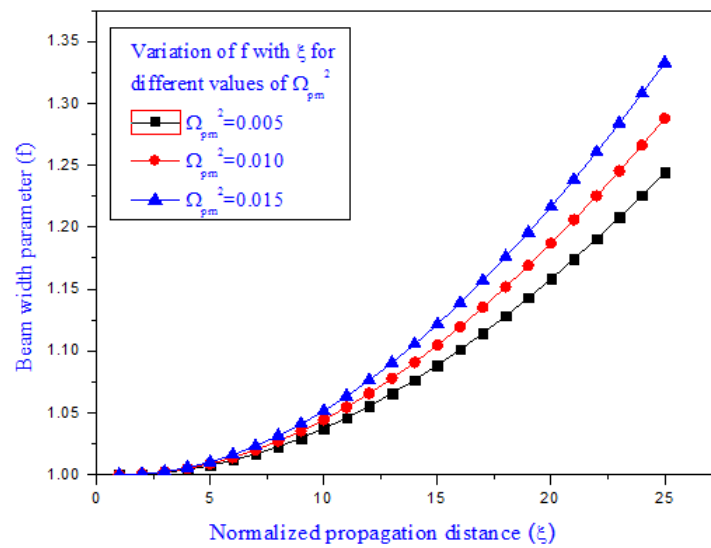


Figure 3: Variation of beam width parameter (f) with normalized propagation distance (ξ) for different values of Ω_{pm}^2 . The other parameters are normalized spot size (η) = 600 and $\varepsilon_L = 1$.

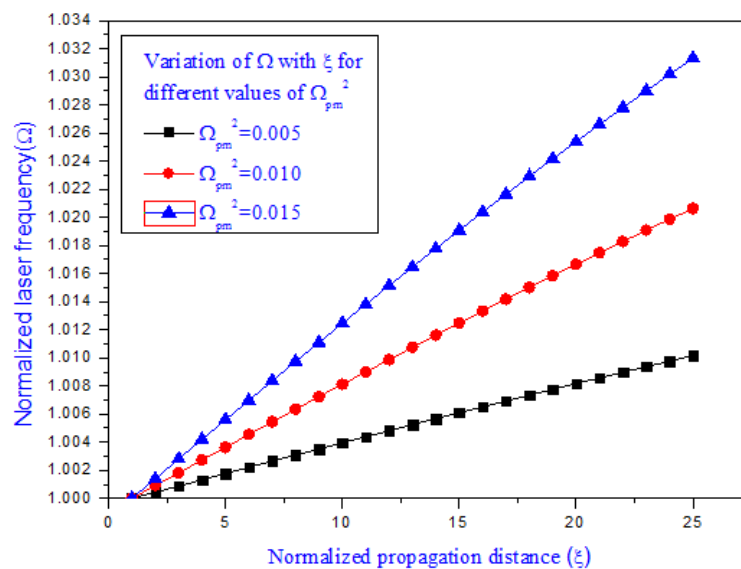


Figure 4: Variation of normalized laser frequency (Ω) with normalized propagation distance (ξ) for different values of Ω_{pm}^2 . The other parameters are normalized spot size (η) = 600 and $\varepsilon_L = 1$.

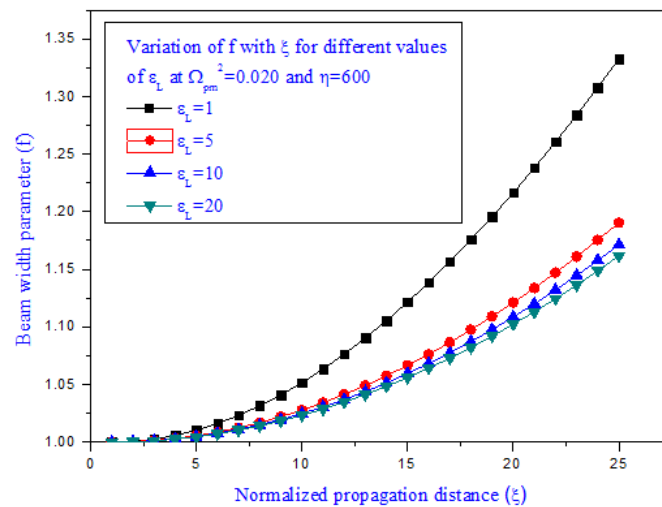


Figure 5: Variation of beam width parameter (f) with normalized propagation distance (ξ) for different values of dielectric constant (ϵ_L) of laser beam. The other parameters are $\Omega_{pm}^2 = 0.005$ and $\eta=600$.

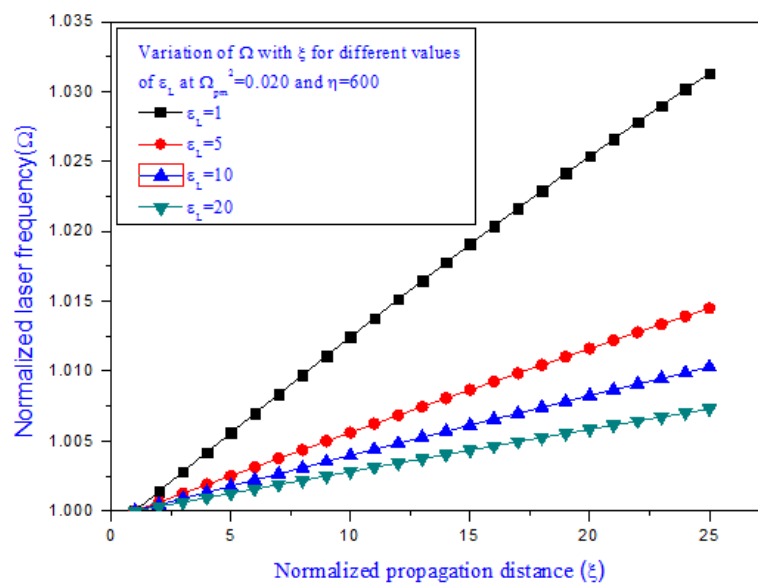


Figure 6: Variation of normalized laser frequency (Ω) with normalized propagation distance (ξ) for different values of dielectric constant (ϵ_L) of laser beam. The other parameters are $\Omega_{pm}^2 = 0.005$ and $\eta=600$.

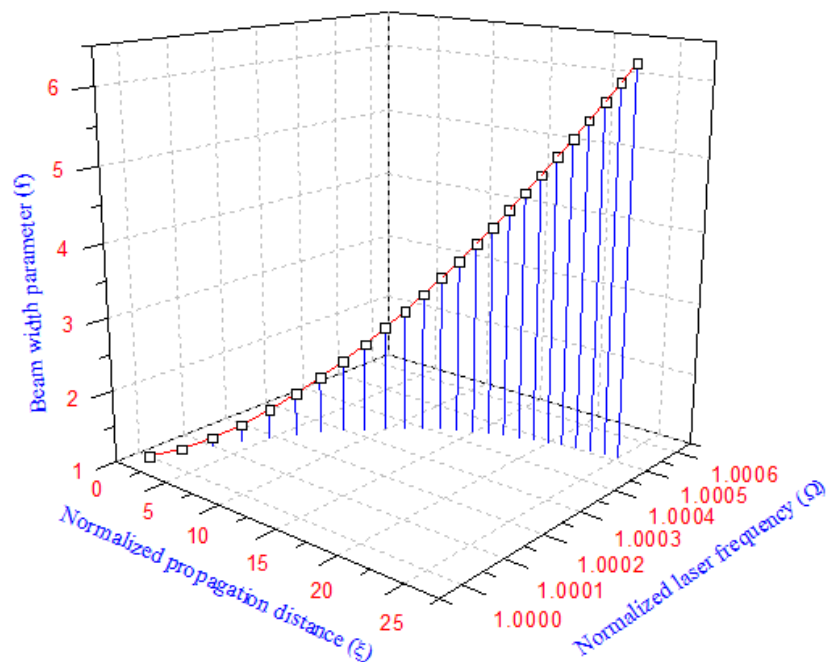


Figure 7: Laser frequency shift with propagation distance

V. RESULTS AND DISCUSSIONS

The material chosen for numerical appreciation is a dielectric sample of fused silica. The Ti: Sapphire laser system is used whose standard output in 25 fs is 0.5 mJ at a repetition rate of 1 KHz. The laser had an intensity of $5 \times 10^{17} \text{ W/cm}^2$, $I_0 = 9 \text{ eV}$ at $\omega_0 = 2 \times 10^{15} \text{ rad/s}$ which emits red and near-infrared light in the range from 650 nm to 1100 nm. Figure 1 elicits the frequency shift with respect to distance (ξ) and dielectric constant ϵ_L . Figure 2 represents the monotonic increase in the beam width parameter with respect to normalized distance at different normalized spot sizes and reveals that the rate of divergence due to diffraction for higher laser spot sizes vary smoothly. Figure 3 illustrates that the rate of frequency increase with propagation distance for beams with larger spot sizes is large in comparison to others. The atoms or ions in the influence of electric field of the fast moving electrons and slow moving ions causing perturbation to energy level is proportional to electric field resulting in shift in frequency, hence pronouncing a strong dependence on laser spot size. As the dielectric constant increases the laser beam has to face more resistance in propagation (Figure 4) where the curves of beam width parameter versus normalized distance slopes down for higher value of dielectric constant. Light traverses in a region in which the refractive index varies in time, but not spatially, the wavelength remains constant. However, the phase velocity of the propagating wave does change; it increases if the index is decreasing. When the wavelength remains constant while the speed increases an observer at a fixed location will therefore see more peaks per second, measurement then results in a higher frequency. Even if the wave moves out of the spatial region in which the index was time-varying, the frequency upshift persists in picture as a spatial change in refractive index does not affect the frequency. In the case of a time-decreasing index, the medium has done work on the photon and made it more energetic. Ionization of a transparent material causes a time-decreasing refractive index, because the effective index of plasma is lower than that of the surrounding medium. The

change in refractive index induces self phase modulation and ultimately frequency broadening. It is evident from the Figure 5, where the dimensionless frequency downshifts with distance and its rate of downshift has strong correlation with relative permittivity. Figure 6 shows the nonlinear dependence of beam width parameter with propagation distance for different values of Ω_{pm}^2 . The density varies with the increase in the value of free charge carrier density Ω_{pm}^2 , there is a modulation in refractive index of the dielectric, causing self phase modulation and ultimately frequency upshift. The frequency shift saturates with propagation distance, which is shown in Figure 7.

VI. CONCLUSIONS

Frequency shift plays a significant role in influencing the variation of parameters of the incident laser beam due to the interaction between plasma formed in focal volume of sample and ultra-short intense laser beam. Our model is applicable and most suitable for modeling the femtosecond laser induced breakdown in large band gap dielectrics at higher intensities. Normalized frequency up-shifts with the normalized propagation distance, for increasing normalized spot sizes of laser. While with increase in the value of dielectric constant, the normalized frequency shows a downshift and beam width parameter rises with normalized propagation distance. It proves to be potential area of research (theoretical as well as experimental) as applications range from enhancing possibilities in micromachining to multiphoton mediated subpico-processing of human chromosomes. The study of variation of laser parameters will help us to exploit the potential unleashed in the interaction between ultrashort lasers and dielectric.

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