Modeling the Femtosecond Pulse Interaction with Fused Silica

Supriya Rai
Department of Physics
Lovely Professional University, Jalandhar, Punjab, India

Abstract— An explicit model of physics behind laser breakdown of fused silica by femtosecond laser pulses is developed. When the ultra short laser pulse propagates for time interval shorter than time required for heat conduction, diffusion, electron to lattice energy transfer and other relaxation processes, ergo interaction is spatially localized. Multi-photon absorption and tunneling of electrons from valence band to conduction band are the two competing mechanisms for swift production of electron hole pairs, supplemented by Joule heating and avalanche ionization. The rapid rise in free carrier density in the interaction zone subsequently influences refractive index, modifying the phase of the laser and causing frequency shift. This model is best fit for higher frequencies of the order 1017 W/cm2 under the application of WKB and SEWA approximation.

Key words: Fused Silica, Femtosecond Pulse

I. INTRODUCTION

The developments in the field of interaction of dielectrics with intense ultra-short laser pulses can be accredited to advent of femtosecond lasers. This regime of laser matter interaction has seen extensive experimental advancement in past decades, but lacks theoretical background to explain the observed results.

Lenzer et al. (1998) illustrates laser induced breakdown of dielectrics as three steps: (i) the excitation of electrons from valence band to the conduction band via multiphoton ionization (MPI) and/or tunneling, (ii) electron-electron collisional ionization (avalanche process) due to Joule heating, and (iii) transfer of the plasma energy to the lattice. Theoretically the generation of electron hole pairs and flow of energy has been understood as initial two processes store energy in the plasma while the third process releases the deposited energy to the lattice. Stuart et al. (1996) tested their hypothesis in femtosecond regime and concluded multiphoton ionization alone is enough to cause production of electron densities above breakdown threshold. Quere et al. (2001) have concluded that breakdown in dielectrics is produced by multiphoton absorption by valence electrons, which is enhanced by tunneling as well as there was no sign of electron avalanche. Dias et al. (1997) reported for the first time frequency upshifts of the order 25 microns. Liu and Tripathi (2000) have studied laser frequency upshift, defocusing, and ring formation in gases interacting with ultrashort laser beams. However, the propagation dynamics of ultrashort laser beam during laser induced breakdown in dielectrics remains less investigated, hence a potential area of research.

When the intense femtosecond laser beam interacts with dielectric sample, causes the inceptive deposition of energy in valence band through multi-photon ionization and creates quasi free electron population in conduction band. The generated electrons in conduction band exploit latter part of pulse to increase their energy. The hot electrons collide and transfer energy to other electrons increasing free electron density. Consequently, the change in refractive index in focal volume leads to phase modulation and frequency shift. In this paper we have derived equation for propagation of dielectrics and focused on encompassing control parameters and the effect of variation of those parameters for optimization of the shift either sides of laser frequency.

II. THEORETICAL ANALYSIS

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

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

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 $E_o(t) = [2/(1 + n)]\, E_{1'}$, where n is the refractive index of the dielectric and k = (ω/c)n. For z > 0, we have $|\vec{E}| = \hat{x}Ae^{-i\phi}$ where ϕ (z, t) = ωt − kz is the fast phase of the wave and A(z, r, t) is a slowly varying complex amplitude of the wave.(ω = ∂ϕ/∂t and k = −∂ϕ/∂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 α_0(Te)I distinguishes the rate of the collisional e-h production and I(t) is the laser intensity of the laser pulse.

$$W = 2/9\pi\left(E_g/\hbar\right)\left(mE_g/\hbar^2\right)^{3/2}\left(|\vec{E}|/E_A\right)^{5/2}\exp\left(-E_A\pi/2|E|\right)$$

and E_A is the strength of the atomic field.

At enormously high laser intensities (>> 10^{17}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

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

III. LASER FREQUENCY UP-SHIFT

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

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

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

$$2ik\frac{\partial A}{\partial z}+iA\frac{\partial k}{\partial z}+\nabla_{\perp}^{2}A+\frac{2iA\varepsilon_L}{c^2}\frac{\partial \omega}{\partial t}+\frac{iA\varepsilon_L}{c^2}\frac{\partial k}{\partial t}=\frac{A}{c^2}\left(\omega_p^{2}-\omega_{po}^{2}\right)+\frac{i\nu\omega_p^2 A}{\omega c^2} \quad (5)$$

Differentiating the above equation with respect to t (using $\partial k/\partial t = -\partial\omega/\partial z$) and taking group velocity of the form $v_g=(c/\varepsilon_L)\left(\varepsilon_L-\omega_{po}^{2}/\omega^2\right)^{1/2}$ for under-dense plasma $\omega_p^{2} << \omega^2$, 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 of the form $G_0=\frac{E_{00}^2}{f^2}\exp(-r^2/r_0^2f^2)$, following the same approach results in the equation of beam width parameter [6]

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

Introducing dimensionless quantities for paraxial ray approximation [10] $\xi=z'c/\omega_o r_o^2$, $\xi=z'/R_d$,

where R_d is defined as the Rayleigh length ($R_d=kr_o^2$), $\tau'=\gamma_o t'$, η is the normalized propagation distance given by $\eta=\omega_o r_o/c$, $\Omega_{pm}=\omega_{pm}/\omega_0$, $\Omega_{p2}=\omega_{p2}/\omega_0$, and $\Omega=\omega/\omega_0$ where $\gamma_o=(\pi/2)^{1/2}(I_o/\hbar)(1/g^2)$ 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

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

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

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

where

$$V=\left(1/f^2\right)\exp(-gf),$$

$$\gamma=(\pi/2)^{1/2}(I_o/\hbar)\left(|E|/E_A\right)^2\exp\left(-E_A/|E|\right).$$

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

IV. Results and Discussion

When Ti: Sapphire laser (intensity of 5×10^{17}W/cm^2, I_o=9 eV at $\omega_o=2\times10^{15}$rad/s) with standard output in 25fs is 0.5mJ at a repetition rate of 1KHz, is made to irradiate dielectric sample of fused silica, the incident laser beam undergoes frequency broadening owing to change in the refractive index of the material governed by the equation $n(I)=n_0+n_2I^2+n_3I^3+\cdots$, where n_0 is absolute refractive index, n_2 and n_3 are other coefficients and I is intensity of laser pulse.

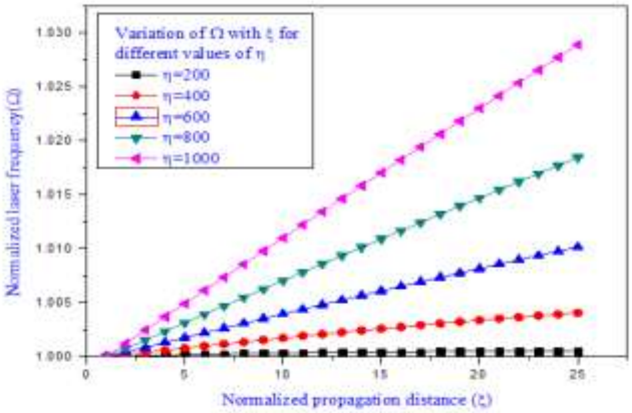

Fig. 1: Variation of normalized laser frequency (Ω) with normalized propagation distance (ξ) for different values of normalized spot size (η) of laser beam. The other parameters are Ω_{pm}^2 = 0.005 and ε_L =1.

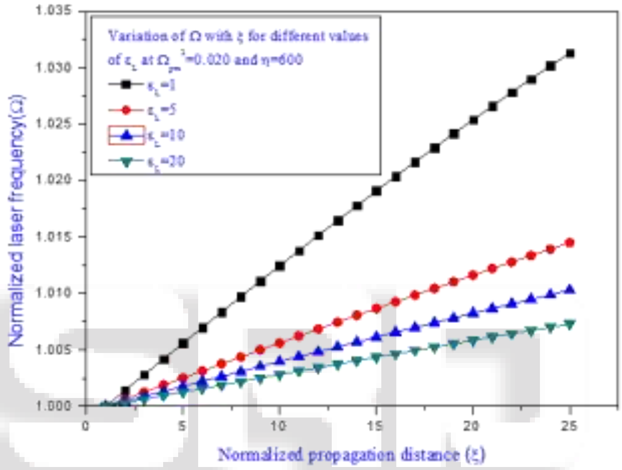

Fig. 2: Variation of normalized laser frequency (Ω) with normalized propagation distance (ξ) for different values of ε_L. The other parameters are normalized spot size (η) = 600 and ε_L =1 Ω_{pm}^2=0.005.

Figure 1 illustrates that the rate of frequency increase with propagation distance for beams with larger spot sizes is large in comparison to smaller values. 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 of frequency shift on laser spot size. It is evident from the Figure 2, where the dimensionless frequency downshifts with distance and its rate of downshift has strong correlation with relative permittivity. The density varies with the increase in the value of free charge carrier density Ω_{pm}^{2}. 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

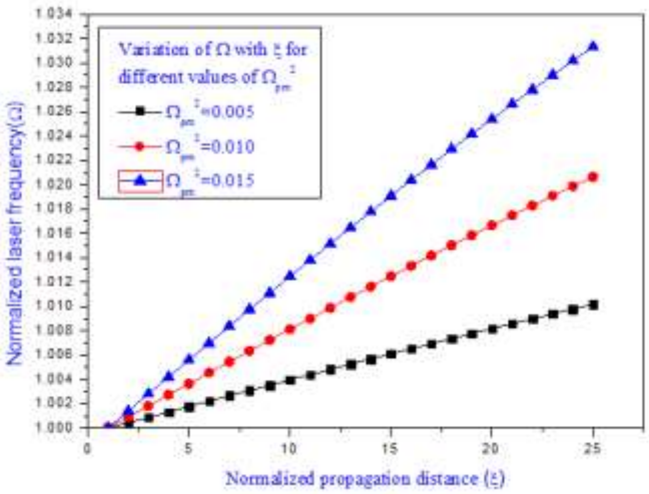

Fig. 3: Variation of normalized laser frequency (Ω) with normalized propagation distance (ξ) for different values of Ω_{pm}^2. The other parameters are normalized spot size (η) = 600 and ε_L =1.

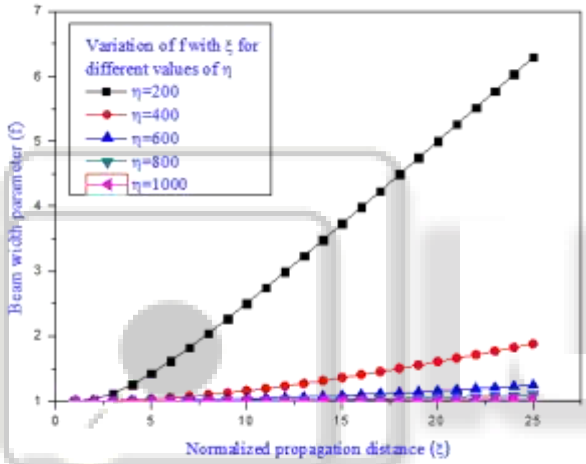

Fig. 4: Variation of beam width parameter (f) with normalized propagation distance (ξ) for different values of normalized spot size (η) of laser beam. The other parameters are Ωpm2 = 0.005 and ε_L =1.

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. The frequency shift saturates with propagation distance, which is shown in Figure 3. Figure 4 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.

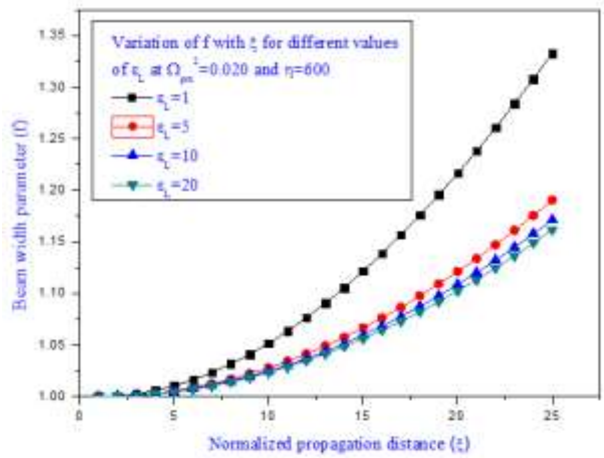

Fig. 5: Variation of beam width parameter (f) with normalized propagation distance (ξ) for different values of dielectric constant (ε_L) of laser beam. The other parameters are Ω_{pm}^2 = 0.005 and η=600.

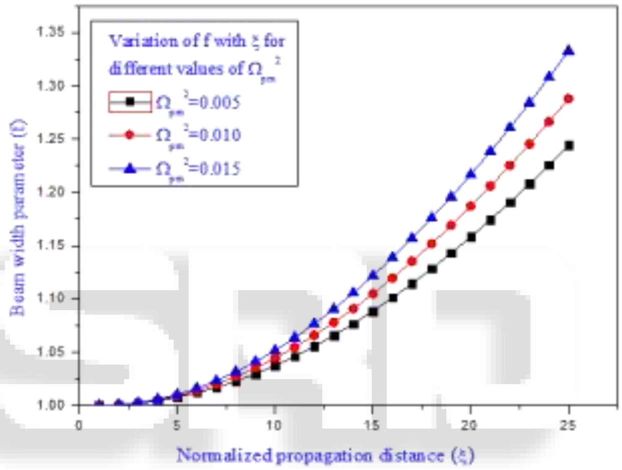

Fig. 6: Variation of beam width parameter (f) with normalized propagation distance (ξ) for different values of Ω_{pm}^2. The other parameters are normalized spot size (η) = 600 and ε_L =1.

As the dielectric constant increases the laser beam has to face more resistance in propagation (Figure 5) where the curves of beam width parameter versus normalized distance slopes down for higher value of dielectric constant. Figure 6 shows the nonlinear dependence of beam width parameter with propagation distance for different values of Ω_{pm}^2.

V. CONCLUSIONS

The paper focuses on developing a numerical model of laser induced breakdown in large band gap dielectrics. The study comprises of irradiating the sample of fused silica in the time scale of few femtoseconds at high intensities. The multi-photon absorption becomes instrumental in production of seed electron-hole pairs via tunneling of electron to conduction band. The beam width parameter increases monotonically with the normalized propagation distance for decreasing values of laser spot sizes and dielectric constant but increasing molecular densities. 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. Our model is applicable and most suitable for modeling the femtosecond laser induced breakdown in large band gap dielectrics at higher intensities. It proves to be potential area of research (theoretical as well as experimental) as applications range from enhancing possibilities in 3D optical storage to keratoplasty. The study of control parameters will help us to exploit the potential unleashed in the interaction between ultrashort lasers and dielectric.

ACKNOWLEDGMENT

The author thanks the initial motivation of Dr. Niti Kant, LPU, however, the results and conclusions are the views of author.

REFERENCES

[1] Bakunov M.I., Bystrov A.M., and Gildenburg V.B. (2002) “Frequency self upshifting of intense microwave radiation producing ionization in a thin gaseous layer”, American Institute of Physics, Phys. Plasmas, 9, 2803.
[2] Dias J.M., Stenz C., Lopes N., Badiche X., Blasco F., Dos Santos A., Oliveira e Silva L., Mysyrowicz A., Antonetti A., and Mendonça J.T. (1997) “Experimental Evidence of Photon Acceleration of Ultrashort Laser Pulses in Relativistic Ionization Fronts”, Phys. Rev. Lett., 78, 4773.
[3] Kumar N. and Tripathi V.K. (2005) “Non-paraxial theory of self defocusing /focusing of a laser in a multiple –ionizing gas”, *Appl. Phys*., 82, 53
[4] Lenzner M., Krüger J., Sartania S., Cheng Z., and Spielmann Ch.(1998) “Femtosecond Optical Breakdown in Dielectrics”, Phys. Rev. Lett., 80, 4076.
[5] Li Ming, Menon Saipriya, Nibarger John P., and Gibson George N. (1999) “Ultrafast Electron Dynamics in Femtosecond Optical Breakdown of Dielectrics”, Phys. Rev. Lett., 82, 2394.
[6] Liu C.S. and Tripathi V.K. (2000) “Laser frequency upshift, self-defocusing, and ring formation in tunnel ionizing gases and plasmas”, *Phys. Plasmas*, 7, 4360.
[7] Quéré F., Guizard S. and Martin P. (2001) “Time-resolved study of laser-induced breakdown in dielectrics”, *Europhys. Lett.* , 56, 136.
[8] Rethfeld B. (2006) “Free-electron generation in laser-irradiated dielectrics” PHYSICAL REVIEW B, 73, 035101.
[9] Stuart B. C, Feit M. D, Rubenchik A. M, Shore B. W, and Perry M. D (1996) “Nanosecond to picosecond laser induced breakdown in dielectrics” Phys. Rev. B, 53,1761.
[10] Verma U. and Sharma A.K. (2010) “Laser frequency upshift and self defocusing under avalanche breakdown of air”, *Phys. Plasmas*, 23, 234.

