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Absorption coefficient along a propagated ultra-short laser pulse in the air based on Kramers–Kronig relations

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In this manuscript, some effects such as nonlinear Kerr, stimulated Raman, and plasma generation effects lead to obtaining the nonlinear refraction index of the air along an intense ultra-short laser pulse based on one-dimensional propagation analysis. The variations in the pulse frequency by the self-phase modulation effect are investigated for achieving the functionality of the refractive index to the frequency. The mentioned functionality allows implementing Kramers–Kronig relations to measure the absorption coefficient. Results indicate that the front of the laser pulse faces a high rate of the energy loss whereas the back of the pulse experiences a gain. The implementation of Kramers–Kronig relations for a theoretical calculation of absorption coefficient variation along a laser pulse propagating in the air in which we have simultaneously taken into account the three above-mentioned effects distinguishes our work from other studies.

Keywords: ultra-short laser pulse, refractive index, self-phase modulation, absorption coefficient, Kramers–Kronig relations.

1. Introduction

The subject of the filamentation formation using propagation of the femtosecond laser pulse through the atmosphere was observed in 1995 [1]. This subject has attracted attention due to a wide range of applications, such as high intensity transportation of light bullets over a long distance, remote sensing in the atmosphere, triggering, guiding electric discharge, light detection, laser-plasma-based accelerators, and nonlinear light detection and ranging (LIDAR) [1–9]. The crucial and common point in these applications concerns the maximum propagation distance of the light filament. A long propagation distance of the laser pulse might exist thanks to nonlinear effects.

Actually, the propagation of the femtosecond laser pulse in the air is more interesting because its short-time interaction excites different nonlinear effects in comparison with continuous laser beam or a long laser pulse. For instance, the interaction between the high-peek-power laser pulse and the air gives rise to strong nonlinear effects such as stimulated Raman processes, self-focusing, multi-photon ionization, plasma generation, and self-phase modulation (SPM), which lead to strong modifications of ultra-short terawatt laser pulse behaviors [10–13]. Also, crossing of a laser pulse end through plasma leads to the generation of terahertz radiation [3, 14–16]. The propagation of intense laser pulses in plasma modifies the plasma density through a ponderomotive force and it also changes its refractive index and absorption coefficient along the pulse [17–19]. The propagation of a high-peek-power pulse over a long distance is possible because of the balancing between the above-mentioned nonlinear effects. According to the nonlinear Kerr-relation $n = n_0 + n_2 I$, self-focusing occurs for a laser pulse with radial dependence of intensity, where n and n_0 are nonlinear and linear refractive index, n_2 is Kerr coefficient, and I is laser pulse intensity. The enhancement of laser pulse intensity leads to inducing partial ionization in the air, and then plasma can be generated. The refraction under dense plasma limits the volume of self-focusing and diffracts the laser pulse. The competition between the self-focusing and diffraction processes leads to guiding of a laser pulse for a long distance. However, a continuous energy loss will deplete the energy in the channel and eventually unbalance this trapping process.

Spatiotemporal dependence of the laser pulse intensity causes variation in the refractive index and absorption coefficient along a laser pulse. The investigation of the refractive index and absorption coefficient helps us to select a suitable transverse and longitudinal profile of the intensity for the optimization of propagation distance.

SPRANGLE *et al.* studied the profile of the refractive index for an ultra-short intense laser pulse [20]. They have found the spectral broadening in the laser pulse using of SPM. Thus, the results show a 10% red-shift at the front of the pulse and a 40% blue-shift at the back.

In this manuscript, the 1D analysis of laser pulse propagation in the air has been assumed. The profile of the refractive index, which includes the influences of the nonlinear Kerr effect, the stimulated Raman effect, ionization, under dense plasma, has been found. Then, using SPM, the frequency dependence of the refractive index has been obtained. Finally, the absorption coefficient dependence on frequency and time has been measured based on Kramers–Kronig relations. Here, we have calculated all the mentioned quantities theoretically. Also, it should be mentioned that the proposed study is a novel method of measuring the absorption coefficient in assumed media.

2. The basic formulation

The analysis of intense ultra-short laser pulse propagation through the air is considered here. A linearly polarized electric field of radiation is assumed which propagates along the z direction and is represented by,

$$\mathbf{E} = A(z, t)\exp\left[i(k_0 z - \omega_0 t)\right] + \text{c.c.} \tag{1}$$

where A, ω_0, and k_0 are electric field amplitude, frequency and wave number, respectively. The wave equation governing propagation of the electromagnetic radiation in a medium can be obtained by Maxwell's equations. According to Eq. (1) for time variations of the form $\exp(-i\omega_0)$, we can use $-i\omega_0$ instead of time derivative ($\partial/\partial t \to -i\omega_0$). Therefore the Ampere's law of the Maxwell's equations can be written as

$$\nabla \times \mathbf{H} = -\frac{i\omega_0}{c}\left(\mathbf{D} - \frac{4\pi}{i\omega_0}\mathbf{J}\right) \tag{2}$$

where **H** is the magnetic field strength vector, **D** is the electric displacement vector, **J** is current density vector and c is the speed of light. We may include the effect of **J** on wave propagation by introducing an effective dielectric constant. In Eq. (2) the quantity within the bracket may be treated as the effective displacement vector,

$$\mathbf{D}_{\mathrm{eff}} = \mathbf{D} + \frac{4\pi}{i\omega_0}\mathbf{J} = \varepsilon_{\mathrm{eff}}\mathbf{E} \tag{3}$$

Here, $\varepsilon_{\mathrm{eff}}$ is the effective dielectric constant. Tacking the curl of the Faraday's law of the Maxwell's equation and using Eqs. (2) and (3) for medium with magnetic permittivity equal to unity, one obtains the following equation

$$-\nabla^2\mathbf{E} + \nabla(\nabla \cdot \mathbf{E}) - \frac{\omega_0^2}{c^2}\varepsilon_{\mathrm{eff}}\mathbf{E} = 0 \tag{4}$$

We assume that the field variations in the z-direction are much larger than in x- and y-directions so that the waves can be treated as transverse in the zeroth order approximation and hence no net space charge is generated in the air, then $\nabla \cdot \mathbf{E} = 0$ [28]. Using Eq. (4), the wave equation governing the pulse propagation can be reduced as

$$\nabla^2\mathbf{E} + \frac{\omega_0^2}{c^2}\varepsilon_{\mathrm{eff}}\mathbf{E} = 0 \tag{5}$$

By substituting the electric field from Eq. (1) and using the paraxial approximation ($\partial^2 A/\partial z^2 = 0$), the wave equation is simplified to

$$2ik_0\frac{\partial A}{\partial z} + \left[n^2(z, t) - n_0^2\right]\frac{\omega_0^2}{c^2}A = 0 \tag{6}$$

where $n(z, t)$ and n_0 are the refractive index in configuration space variables and linear refractive index respectively. Different phenomena can have an effect on the refractive index. Here, the contributions of the Kerr effect, Raman scattering and plasma have been considered. The variation of the refractive index ($\delta n = n(z, t) - n$) is very small in comparison with its linear counterpart. Therefore, the wave equation (6) is given by

$$\frac{\partial A}{\partial z} = i\delta n\frac{\omega_0}{c}A \tag{7}$$

where the variation of the refractive index is $\delta n = \delta n_{\mathrm{Kerr}} + \delta n_{\mathrm{Raman}} + \delta n_{\mathrm{plasma}}$.

2.1. Variations of the refractive index

The interaction between the laser pulse and the electrons of non-ionized atoms and molecules, that are called bound electrons, leads to a variation in the medium refractive index. Also, the free electrons generated by ionization of medium atoms and molecules by an electromagnetic wave can change the refractive index through different mechanisms. In this section, the individual contributions of the bound electrons and free electrons have been derived. At first we begin with the inserted influences due to the Kerr nonlinear and stimulated Raman effects. Nonlinear polarization of the Kerr effect is shown by [21],

$$\mathbf{P}_{\mathrm{Kerr}} = \chi_{\mathrm{nl}} \langle \mathbf{E} \cdot \mathbf{E} \rangle_t \, \mathbf{E}(\mathbf{r}, t) \tag{8}$$

where χ_{nl} and $\langle \, \rangle_t$ are third-order of nonlinear susceptibility and time average among the electric field, respectively. Also, polarization of the stimulated Raman effect is given by [22]

$$\mathbf{P}_{\mathrm{Raman}} = \chi_l \, Q(t) \mathbf{E}(\mathbf{r}, t) \tag{9}$$

Here χ_l is the linear susceptibility, and $Q(t)$ is unit-less oscillator function that can be expressed as [20]

$$Q(\mathbf{r}, t) = -\frac{n_R n_0}{2\pi \chi_l} \int_{-\infty}^{t} \mathrm{d}t' \, W(t') \, R(t - t') \, I(\mathbf{r}, t') \tag{10}$$

where n_R is Raman effect contribution in nonlinear refractive index for pulse, $W(t)$ is the difference of the normalized population densities between two different states, in which, if all molecules are in a ground state ($W = -1$), $R(t)$ is the Green function in the Raman effect, that is given by [20]

$$R(t) = \frac{\omega_R^2 + \Gamma_2^2}{\omega_R} \exp(-\Gamma_2 t) \sin(\omega_R t) \tag{11}$$

Here ω_R and Γ_2 are time characteristics of the stimulated Raman effect. Recent experiments show that for the propagation of shorter than ~100 fs laser pulses with $\lambda = 800$ nm through the air, the effective parameters (for the short-pulse regime) are $n_2 \approx n_R = 3\times10^{-19}$ cm^2/W, $\omega_R = 1.6\times10^{13}$ s^{-1} and $\Gamma_2 = 1.3\times10^{13}$ s^{-1} [27]. Finally, by considering Kerr and Raman effects, the polarization can be expressed by

$$\mathbf{P} = \chi_l \mathbf{E}(\mathbf{r}, t) + \chi_{\mathrm{nl}} \langle \mathbf{E} \cdot \mathbf{E} \rangle \, \mathbf{E}(\mathbf{r}, t) + \chi_l \, Q(t) \mathbf{E}(\mathbf{r}, t) \tag{12}$$

Using $\langle \mathbf{E} \cdot \mathbf{E} \rangle = A^2/2$, the effective susceptibility of the medium is given by,

$$\chi_{\mathrm{eff}} = \chi_l + \chi_{\mathrm{nl}} \frac{A^2}{2} + \chi_l \, Q(t) \tag{13}$$

At this case, by $n = \sqrt{1 + 4\pi\chi_{\mathrm{eff}}}$ the refractive index can be written as

$$n^2 = 1 + 4\pi\left[\chi_l + \chi_{\mathrm{nl}}\frac{A^2}{2} + \chi_l Q(t)\right] \tag{14}$$

We can write the refractive index as $n = n_0 + n_2 I + n_{\mathrm{nl\,Raman}}$. Laser pulse intensity can be introduced as $I = cn_0|A|^2/8\pi$. By assuming that the influence of the nonlinear effect is smaller than n_0, we can write $n_{\mathrm{nl}} = n_2 I + 2\pi/(n_0\chi_l Q(t))$. Substituting $Q(t)$ from Eq. (10), the variation of the refractive index is

$$\delta n_{\mathrm{Kerr}} + \delta n_{\mathrm{Raman}} = n_2 I - n_R\int_{-\infty}^{t} \mathrm{d}t'\, W(t')\, R(t-t')\, I(\mathbf{r}, t') \tag{15}$$

Now we try to calculate the influence of plasma on the refractive index. The refractive index in plasma is expressed by $n = (n_0^2 + \omega_p^2/\omega_0^2)^{1/2}$, where $\omega_p^2 = 4\pi e^2 n_e/m$ is the plasma frequency, e, n_e and m are charge, density and mass of electron, respectively. For high frequencies, $\omega_0 \gg \omega_p$, we can use the approximated value $n \approx$ $\approx n_0 - \omega_p^2/(2n_0\omega_0^2)$ for n. Therefore, the plasma influence is given by

$$\delta n_{\mathrm{plasma}} = -\frac{2\pi e^2 n_e}{n_0 m \omega_0^2} \tag{16}$$

The plasma medium is generated on interaction between the laser pulse and the air. Then, the rate of electron density can be obtained as [23, 24]

$$\frac{\partial n_e}{\partial t} = w n_n - \eta n_e - \beta_r n_e^2 \tag{17}$$

The above-mentioned equation involves ionization, recombination and attachment processes. Here, w is the photoionization rate coefficient, n_n is the density of the neutral atoms, η is the electron attachment rate coefficient, and β_r is the recombination coefficient rate. Because of short interaction time for the ultra-short laser pulse, recombination and electron attachment processes can be neglected. Also, ionization occurs by multi-photon and tunneling processes, and avalanche ionization is not significant. For each of these processes, the ionization rate takes different value according to the Keldysh parameter $\gamma_K = 2.31(U_{\mathrm{ion}}/\lambda^2 I)^{1/2} \times 10^6$, where, U_{ion} is the ionization energy in electron-volts, λ is wavelength in micrometers [25]. The value of the Keldysh parameter identifies which one of ionization processes occurs. Namely, for $\gamma_K \gg 1$ and $\gamma_K \ll 1$ multi-photon and tunneling occur, respectively, the middle of an analytical fit is employed for the middle regime ($\gamma_K \sim 1$) [20]. In this study, we assume that the multi -photon ionization occurred. In this regime, the ionization rate is [26]

$$w = \frac{2\pi\omega_0}{(l-1)!}\left[\frac{I(\mathbf{r}, t)}{I_{mp}}\right]^l \tag{18}$$

where $I_{mp} = \hbar\omega_0^2/\sigma_{mp}$ and σ_{mp} is the cross-section whose value for a short laser pulse is 6.4×10^{-18} cm^2 [12]. The integer l is the minimum number of photons that is necessary for ionization. Using Eqs. (17) and (18), we can write electron density as

$$n_e(t) = n_{n_0}\left[1 - \exp\left(-\frac{2\pi\omega_0}{I_{mp}^l(l-1)!}\int_0^t I^l \mathrm{d}t'\right)\right] \tag{19}$$

Here n_{n_0} is the density of neutral atoms in the absence of the laser pulse.

2.2. Self-phase modulation

The variable refractive index along the laser pulse propagating in the air can result in the instantaneous frequency spread due to self-phase modulation. Because of time dependence of the refractive index due to nonlinear Kerr, Raman, and plasma generation effects, the phase of the pulse becomes modulated.

The solution of Eq. (6) has the form $A(z, \tau) = B(z, \tau)\exp[i\theta(z, \tau)]$, where B is the amplitude and θ is the phase function. So, the instantaneous frequency of the pulse can be expressed as

$$\omega(z, \tau) = \omega_0 - \frac{\partial\theta}{\partial\tau} \tag{20}$$

Substituting A in the wave equation (7), we obtain

$$\frac{\partial\theta}{\partial\tau} \approx \frac{\omega_0}{c}\delta n(z, \tau) \tag{21}$$

Therefore, the variation of frequency along the pulse is given by

$$\delta\omega(z, \tau) = -\frac{\omega_0}{c}\int_0^z \frac{\partial\delta n(z', \tau)}{\partial\tau}\mathrm{d}z' \tag{22}$$

Using the variation of the refractive index due to Kerr, stimulated Raman, and plasma generation processes, we obtain

$$\delta\omega(z, \tau) = -\frac{n_2\omega_0}{c}\frac{\partial I}{\partial\tau}z + \frac{n_R\omega_0 z}{c}\int_0^\tau W(\tau')\frac{\partial R(\tau-\tau')}{\partial\tau}I(\mathbf{r}, \tau)\mathrm{d}\tau' + \frac{1}{2cn_0\omega_0}\frac{\partial\omega_p^2}{\partial\tau}z \tag{23}$$

2.3. Variations of the absorption coefficient

The Kramers–Kronig relations link the real and imaginary parts of frequency dependence of quantities such as the refractive index. Namely, they allow one to calculate the real/imaginary part of the quantities at the particular frequency from the knowledge of the frequency dependence of the imaginary/real part of quantities. In this work, using the refractive index profile (Eqs. (15) and (16)) and spectral broadening (Eq. (23)) along the laser pulse, we can determine the frequency dependence of the refractive index. Then, the frequency dependence of absorption coefficient has been studied as an imaginary part of the refractive index by the following relation [21]:

$$\kappa(\omega) = -\frac{2\omega}{\pi}\int_0^{+\infty}\frac{n(\omega')-1}{\omega'^2-\omega^2}\,\mathrm{d}\omega' \tag{24}$$

where $\kappa(\omega)$ is the absorption function.

3. Results and discussion

In this section, graphs for the variation of the refractive index, frequency, and absorption coefficient have been plotted. We consider a short laser pulse with duration $\tau_l = 500$ fs, frequency $\omega_0 = 2.43\times10^{15}$ s^{-1}, amplitude $A(z, \tau) = (8\pi I_0)^{1/2}\sin(\pi\tau/\tau_l)$ and peak intensity $I_0 = 5\times10^{15}$ W/cm^2. In Figure 1, the variations of the refractive index due to Kerr and stimulated Raman effects, as given by Eq. (15), *versus* the pulse time $\tau = (t - z/v_g)$ are plotted. As shown in Fig. 1, the variation of the refractive index related to the Kerr effect is maximum ($\delta n_{\mathrm{Kerr}} = 1.5\times10^{-5}$) at $\tau = 250$ fs, because the intensity of the pulse in the center is bigger than its front and behind. The behavior of the plot of the Raman effect contribution is the same as the Kerr effect. But the maximum variation due to the Raman effect is shifted to 70 fs the back of the pulse.

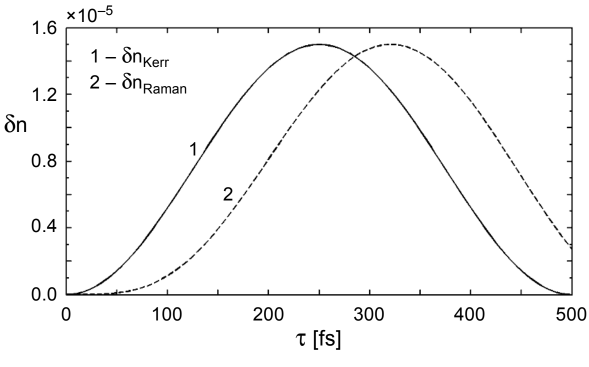

Fig. 1. Variations of the refractive index due to the Kerr effect (solid curve) and Raman effect (dashed curve) *versus* the pulse time τ.

Figure 2 shows the variations of the refractive index due to plasma generation along the pulse. For plasma contribution, we assume that the multi-photon ionization mechanism of O_2 is dominant. Therefore the ionization rate is given by Eq. (18), with $l = 8$. As shown in Fig. 2, the variation of the refractive index is not significant in front of the laser pulse, because the ionization has not started yet and the laser pulse face the natural medium. But, increasing intensity and starting plasma generation lead to refractive index decreasing with a high rate until the ionization stops behind the laser

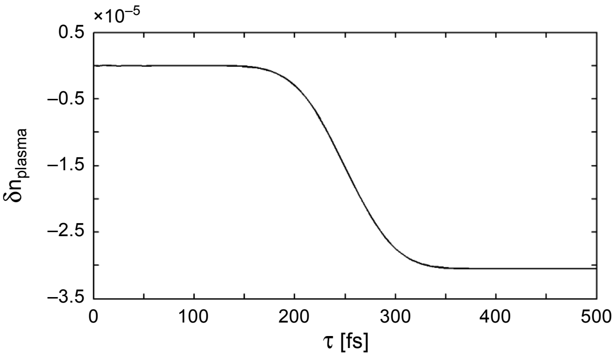

Fig. 2. Variations of the refractive index due to plasma generation *versus* the pulse time τ.

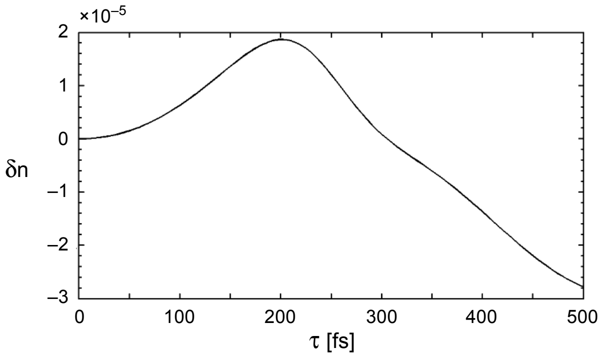

Fig. 3. The sum of the Kerr, Raman, and plasma generation effects influence on the refractive index *versus* the pulse time τ.

pulse, so that the rate of variation is to be zero at this zone. As demonstrated in Figs. 1 and 2, the domain of variations due to plasma generation is bigger than the Kerr and Raman effects.

The sum of the Kerr, Raman, and plasma generation effects influence *versus* the pulse time τ is plotted in Fig. 3. According to Fig. 3, the maximum increasing of the refractive index ($\delta n = 1.87\times10^{-5}$) occurs in $\tau = 200$ fs and the maximum decreasing is related to the back of the laser pulse ($\delta n = -2.78\times10^{-5}$).The weakness of the Kerr and Raman effects at the back of the pulse and the incidence of the back of the pulse with the ionized medium lead to maximum decreasing of the refractive index at this area. Because of the short duration of the laser pulse, the variations in the laser pulse intensity is large and sudden. Also, the nonlinear effects are a sensitive function of the pulse intensity. Therefore, the variations of the nonlinear effect along the laser pulse are large. The encounter of the laser pulse front with non-ionized medium and the end of laser pulse with plasma leads to a large difference between the refractive index in front

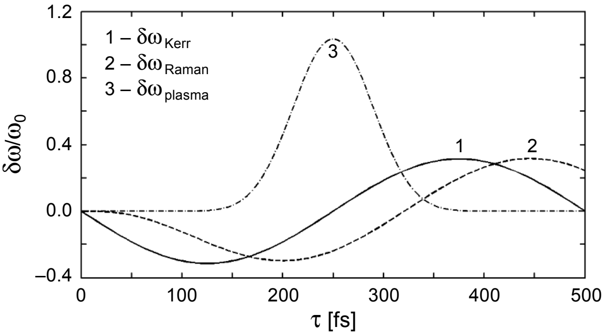

Fig. 4. The relative variations in frequency due to the Kerr (solid curve), Raman (dashed curve), and plasma generation (dot-dashed curve) effects *versus* the pulse time τ.

and end of the pulse. By considering these phenomena, the large change in the index is acceptable.

The relative variations of the frequency due to the Kerr, Raman, and plasma generation effects *versus* the pulse time after propagating for 100 cm in the air are plotted in Fig. 4. Figures show that the variations in frequency due to the Kerr and stimulated Raman effects have the same behavior along the laser pulse. As is obvious from the figure, the frequency in front of the laser pulse is decreased, then it is increased at the end of the pulse. But the plot related to the Raman effect has a little ($\Delta\tau = 35$ fs) shift to the back due to variations in the refractive index. The maximum and minimum values of the contribution of the Kerr effect are $\delta\omega_{\mathrm{Kerr}}/\omega_0 = 0.314$ and $\delta\omega_{\mathrm{Kerr}}/\omega_0 = -0.314$, but for the Raman effect they are $\delta\omega_{\mathrm{Raman}}/\omega_0 = 0.316$ and $\delta\omega_{\mathrm{Raman}}/\omega_0 = -0.298$. According to Fig. 3, the variations rate of the refractive index in front of and at the end of the laser pulse related to plasma generation is not significant. Therefore, the relative variations of frequency at this point are very small. But at the center of the pulse we can see a high decreasing rate of the refractive index that leads to a high increase in the frequency at this area ($\delta\omega_{\mathrm{plasma}}/\omega_0 = 1.035$).

The net normalized variations of the laser pulse frequency, namely the sum of Kerr, Raman and plasma generation effects contribution has been shown in Fig. 5 The maximum red-shift occurs in front of the laser pulse in $\tau = 140$ fs. So that, we can see a 50% red-shift at this point. Also, the maximum blue-shift occurs at the center of the pulse ($\tau = 260$ fs), as one can see a 0.83% blue-shift at this area.

Now, using the data indicated in Figs. 3 and 5, we have found the frequency dependence of the pulse refractive index, then investigated the absorption coefficient profile along the laser pulse, as given by Eq. (24). The variations of the absorption coefficient of the laser pulse *versus* the pulse time were plotted in Fig. 6. With starting a ionization process in front of the pulse, much of the laser energy is lost. Then, while the ionization rate is decreased, the absorption coefficient is decreased also. So that absorption gets to zero in $\tau = 144$ fs and is converted to a gain. The existence of

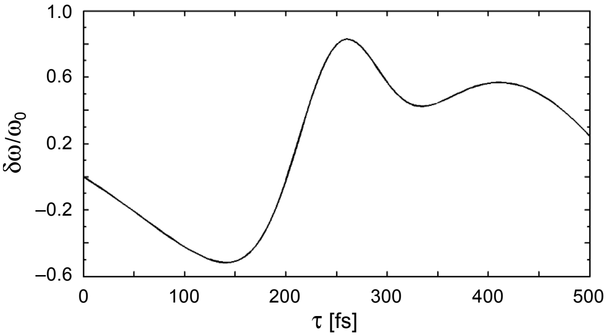

Fig. 5. Total normalized variations in the frequency *versus* the pulse time τ.

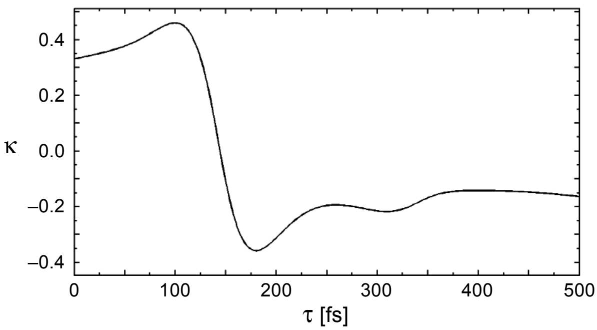

Fig. 6. The variations of the absorption coefficient of the laser pulse *versus* the pulse time τ.

the gain at the center and back of the pulse is related to a recombination process and energy coming back to the electromagnetic field from electrons and ions. It is clear that the energy loss is bigger than the gain when comparing absorption and gain coefficient along the laser pulse.

4. Conclusions

In this work, the absorption coefficient of the laser pulse propagating through the air is investigated. The results show that the existence of the nonlinear Kerr, stimulated Raman, and plasma generation effects leads to variations in the refractive index of the pulse along the propagation. Therefore, the phase of the laser pulse becomes modulated through the dependence of the refractive index on time. Using the self-phase modulation of the laser pulse, we can find the frequency distribution along the pulse. So that, a 50% red-shift and a 85% blue-shift of the laser pulse frequency after propagating for 100 cm have been observed. This shift of the pulse frequency leads to white light generation. By obtaining data from variations in the frequency and refractive index, we can find the frequency dependence of the refractive index. Using the refractive index as a function of frequency and the Kramers–Kronig relation, calculation of the imaginary part of the refractive index, namely absorption coefficient, becomes possible. Results show that the absorption along the laser pulse is varying. So that, at first by starting ionization process in front of the laser pulse, a high energy loss is observed. Then, through recombination and coming back energy from electrons and ions to the laser pulse process, the absorption is converted to a gain at the back of the pulse. The knowledge about an absorption coefficient can help to control the distance of the laser pulse propagation.

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