

# Near dipole-dipole effects on the propagation of few-cycle pulse in a dense two-level medium

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**Abstract:** The propagation behaviors, which include the carrier-envelope phase, the area evolution and the solitary pulse number of few-cycle pulses in a dense two-level medium, are investigated based on full-wave Maxwell-Bloch equations by taking Lorentz local field correction (LFC) into account. Several novel features are found: the difference of the carrier-envelope phase between the cases with and without LFC can go up to  $\pi$  at some location; although the area of ultrashort solitary pulses is larger than  $2\pi$ , the area of the effective Rabi frequency, which equals to that the Rabi frequency plus the product of the strength of the near dipole-dipole (NDD) interaction and the polarization, is consistent with the standard area theorem and keeps  $2\pi$ ; the large area pulse penetrating into the medium produces several solitary pulses as usual, but the number of solitary pulses changes at certain condition.

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## 1. Introduction

Recent advances in ultrafast laser technology have made it possible to generate few-cycle and high intense pulses [1, 2]. It has attracted much interest in the interaction of femtosecond pulse and atoms over the world. When many cycles are contained in the duration of pulse, the nonlinear properties of medium have been studied in an extensive area. In the resonant

medium, the light-matter interaction gives rise to the effect of self-induced transparency (SIT) according to the area theorem [3-5]. However, they are based on the slowly varying envelope approximation (SVEA) and the rotating-wave approximation (RWA). Clearly, these standard approximations are invalid if the pulse duration contains few-cycle or subcycle [6-11]. To analyze the nonlinear interaction between the few-cycle pulse and atoms based on numerical simulation, full-wave Maxwell-Bloch equations without SVEA and RWA are presented [6-10]. Hughes has reported that a large area ultrashort pulse gives rise to the area theorem to break down due to the carrier-wave Rabi flopping [6]. Very recently, Song *et al.* have found the area theorem invalid for an attosecond pulse propagating in a dense two-level medium [8]. Kalosha *et al.* have investigated the propagation of femtosecond pulse in a dense two-level medium, and reported the intrapulse third-order four-wave mixing (FWM) and the formation of optical subcycle pulses [12]. Femtosecond pulse behaviors in semiconductor have been also studied by numerical simulation [13] and in experiment [14]. When the laser pulse just contains one or two optical cycles, the carrier-envelope phase  $\Phi_{CE}$  (defined as the phase of the carrier wave with respect to the pulse peak [15-16]), i.e. the absolute phase, has an important influence on a large number of extremely nonlinear processes, such as photoionization [15-17], high-order harmonic generation [18] and photon emission [19], etc.. So, it is necessary to accurately determine the absolute phase in numerical investigation and in experiments.

In a dilute medium, it is valid to assume that only atoms interact with ultrashort pulse separately from each other. However, in a dense medium, where the atomic density is so high that there are, in the sense of average, many atoms within a cubic resonance wavelength such that the near-dipole-dipole interaction (NDD), which leads to Lorentz local-field correction (LFC), must be considered [20]. Up to now, there are many papers which have considered the NDD interaction based on the Lorentz-Lorentz relation [21] by employing SVEA and/or RWA. A set of generalized Maxwell-Bloch equations including the NDD effect is presented by Bowden *et al.* [20]. The LFC has been shown to produce many interesting phenomena [22-34]. Bowden and his co-workers have shown that a unique ultrafast intrinsic optical switch can be realized if the ratio of the peak Rabi frequency to the strength of the NDD interaction  $\epsilon$  is exactly chosen [22, 23]. As reported by Bowden and his co-workers, when optical pulse, whose temporal duration is much less than an induced-dipole dephasing time, incidents upon a thin film of homogeneously broadened two-level medium with the NDD interaction, the inversion  $w$  has a nearly step-function response to the peak Rabi frequency  $\Omega_0$  after the pulse has passed. The system, initially in the ground state,  $w=-1$ , is always returned to the ground state when the peak Rabi frequency is less than the strength of the NDD interaction  $\epsilon$ . However, the final state of the system is the fully excited state,  $w=1$ , when  $\Omega_0$  is nearly equal to  $\epsilon$ . Thus, the nearly step-function response of the system constitutes a new and unique optical switch, which does not have hysteresis and is independent of dissipation from incoherent effects [22].

In spite extensive works have investigated the NDD effect on the propagation behaviors of ultrashort pulse in a dense medium, but to our best knowledge, almost of them are based on SVEA and/or RWA. In this paper, we numerically investigate the extremely nonlinear properties of femtosecond pulse in a dense two-level medium based on the full Maxwell-Bloch equations and the Lorentz LFC is considered. Some essentially differences are found. The carrier-envelope phase  $\Phi_{CE}$  is sensitive to the NDD interaction. The effect of NDD arouses a decreasing in the center frequency and slightly increasing in the group velocity, which, combining with the self-phase modulation, lead to the change of the absolute phase  $\Phi_{CE}$ . The difference of  $\Phi_{CE}$  between the two cases with and without LFC can go up to  $\pi$  at some location, where Rabi frequency oscillates in opposite polarity. To be surprising, the area of solitary pulse, no matter LFC is considered or not, is evidently larger than  $2\pi$ . However, the area of the effective Rabi frequency, which equals to that Rabi frequency  $\Omega$  pluses the

product of the strength of NDD interaction  $\epsilon$  and the polarization  $u$ , i.e.  $\Omega + \epsilon u$ , keeps invariant area  $2\pi$  in a relative large density range. Moreover, the polarization  $u$  modifies the light-matter interaction. The interaction becomes stronger when the polarization following the Rabi frequency with identical direction, and becomes weaker when the direction of the polarization is opposite. When the area of input pulse is near a certain threshold, the large area pulse penetrating into the medium produces several solitary pulses as usual, however, the number of solitary pulses changes due to the light-matter interaction being enhanced or depressed.

This paper is organized as follows: In the second section, we present a one-dimension full wave Maxwell-Bloch equation for few-cycle pulse propagating in a dense two-level medium and without any approximations of RWA and SVEA, but the Lorentz LFC is considered. In the third section, we investigate the penetrating pulses' properties when a few-cycle pulse normally incident into a dense medium. Finally, we draw some conclusions in the final section.

## 2. Theoretical model

We model the light-matter interaction not only no SVEA and RWA but the NDD effect is included, and consider the propagation of an ultrashort pulse along  $z$  axis which incidents normally into an input interface of a resonant dense two-level medium at  $z=40 \mu\text{ m}$ . As Ref.[12], initially the pulse moves in the free space; then penetrating part propagates through the medium and finally exits again into the free space through the output interface at  $z=190 \mu\text{ m}$ . The medium zone length is  $L=150 \mu\text{ m}$ . According to the Lorentz-Lorenz relation [21], in a simple cubic lattice, the microscopic local electric field  $E_L$ , which couples with an atomic or molecular dipole moment is related to the macroscopic field  $E$  and volume polarization  $P$  in the isotropic homogeneous medium

$$E_L = E + \frac{P}{3\epsilon_0}, \quad (1)$$

where  $\epsilon_0$  is the electric permittivity in the vacuum.

The two-level model can be applied for the description of coherent effects for materials with a broad distribution of transitions such as inhomogeneously broadened resonance lines in gases and solids [3], and also a good approximation for inhomogeneous quasicontinuous energy bands as in semiconductors [35]. For the two-level medium,  $|1\rangle$  is the lower state of the atom,  $|2\rangle$  is the upper state. Under the condition that the electric field is linear polarization along the  $x$  axis and the magnetic field is along the  $y$  axis, the Maxwell's equations take the forms

$$\partial_t H_y = -\frac{1}{\mu_0} \partial_z E_x, \quad (2a)$$

$$\partial_t E_x = -\frac{1}{\epsilon_0} \partial_z H_y - \frac{1}{\epsilon_0} \partial_t P_x, \quad (2b)$$

where  $\mu_0$  is the magnetic permeability in the vacuum. In Eq. (2b) the macroscopic nonlinear polarization  $P_x = Ndu$  is connected with the off-diagonal density matrix element  $\rho_{12} = (u + iv)/2$ , the population difference  $w = \rho_{22} - \rho_{11}$  between the upper and lower states, which are determined by the Bloch equations with LFC,  $N$  is the density of the two-level medium and  $d$  is the dipole moment. Substituting the local field  $E_L$  in Eq. (1) for the electric field in the interaction Hamiltonian operator and Eq. (12) in Ref. [7], the Bloch equations with LFC can be easily derived as follows:

$$\partial_t u = -\omega_0 v - \gamma_1 u, \quad (3a)$$

$$\partial_t v = \omega_0 u + 2(\Omega + \epsilon)w - \gamma_1 v, \quad (3b)$$

$$\partial_t w = -2(\Omega + \epsilon)v - \gamma_2(w - w_0), \quad (3c)$$

where  $\Omega = dE_x/\hbar$  is the Rabi frequency,  $\hbar$  is the Planck's constant divided by  $2\pi$ ,  $\gamma_1$  and  $\gamma_2$  are the polarization and population relaxation constant, respectively,  $\omega_0$  is the transition frequency of the two-level medium, and  $w_0$  is the initial population difference of the system.  $w_0 = -1$  at  $t=0$  means all the atoms initially should be in their lower states. The NDD parameter  $\epsilon = Nd^2/3\epsilon_0\hbar$  has unit of frequency, which presents the strength of the NDD interaction. For  $\epsilon = 0$ , i.e. the NDD effect is not considered, Eq. (3) is consistent with Eq. (2) in Ref. [12].

We employ a Yee's leap-frog finite-difference time-domain (FDTD) discretization scheme [36] and the iterative predictor-corrector method [7] for solving the equations. Mur absorbing boundary conditions [37] were incorporated with FDTD discretization to avoid the influence of the finite space computational domain. The spatiotemporal step is set to ensure  $c\Delta t \leq \Delta z$  [38], i.e.  $\Delta t = 1.25 \times 10^{-17}$  s and  $\Delta z = 7.5 \times 10^{-9}$  m. The initial condition is

$$E_x(t=0, z) = E_0 \operatorname{sech}[1.76(z/c - z_0/c)/\tau_p] \cos[\omega_p(z/c - z_0/c)] \quad \text{and}$$

$$H_y(t=0, z) = \sqrt{\epsilon_0/\mu_0} E_x(t=0, z),$$

where  $z_0 = 25 \mu\text{m}$ ,  $E_0$  is the peak amplitude of the

incident pulse,  $\tau_p$  is the full width at half-maximum (FWHM) of the pulse intensity envelope, and  $c$  is the light velocity in the vacuum. The choice of  $z_0$  ensures that the pulse penetrates negligibly into the medium at  $t=0$ . The medium is initialized with  $u=v=0$ , and the population difference  $w_0 = -1$  at  $t=0$ . In the following numerical analysis, all the material parameters we adopt are based on Ref. [12]:  $\tau_p = 5$  fs,  $\omega_p = \omega_0 = 2.3 \text{ fs}^{-1}$  ( $\lambda = 830$  nm),  $d = 2 \times 10^{-29}$  Asm,  $\gamma_1^{-1} = 0.5$

ps,  $\gamma_2^{-1} = 1$  ps. For these parameters, the density  $N = 4.4 \times 10^{20} \text{ cm}^{-3}$  gives  $\epsilon = 0.0667 \text{ fs}^{-1}$

<sup>1</sup>(Corresponding to  $\omega_c = 0.2 \text{ fs}^{-1}$  in Ref. [12]) and  $\Omega_0 = dE_0/\hbar = 1 \text{ fs}^{-1}$  corresponds to the electric

field of  $E_x=5\times 10^9$  V/m or an intensity of  $I=6.6\times 10^{12}$  W/cm<sup>2</sup>. The input envelope area can be

get by  $A=\Omega_0\tau_p\pi/1.76$ .

### 3. Numerical results

We simulate the light-matter interaction modeled by Eqs. (2) and (3) for different input pulse parameters and medium densities. Similarity to the case of RWA [3], we rotate the vector ( $u$ ,  $v$ ,  $w$ ) but remain the  $2\omega$  part, and find it still valid that the effect of NDD arises a dynamic shift  $\Delta_L = \epsilon w$  in the transition frequency as stated in Refs.[23,30-34,39]. In a medium, whose density is not enough large, a few-cycle pulse has very high Rabi frequency much larger than  $\epsilon u$  for a given area due to ultrashort duration and medium can be nearly completely inversed, whose average is near to zero, so the effect of NDD is negligible. For dense medium, which is weakly excited and  $\epsilon u$  is comparable with the Rabi frequency, the NDD interaction makes some essential differences. Firstly, the reflected, penetrating, and transmitting pulses are investigated for different medium densities and different input pulse magnitude  $\Omega_0$ . It is found that the spectra of these fields are similar to those in Ref. [12] and suffer slight redshift due to the dynamic shift in the transition frequency of medium. Except for the spectra of pulses, we pay more attention on the absolute phase, the area evolution and the number of solitary pulses.

In many research areas, the absolute phase of few-cycle pulse plays an important role due to the extreme short pulse duration. Due to the strongly influence of the absolute phase on a large number of extremely nonlinear interactions and the sensitivity to the effect of NDD, the absolute phase should be accurately determined. Though the very short relaxation times cause an effect of weak damping upon the h.s. pulses [4], the oscillating pulses investigated in this paper do an evidently likesoliton behavior and propagate a distance much longer than that limited by the usual Beer's law. So we still consider the pulses as stable solitons. Through fitting the curve gotten by the numerical simulation in an analytic expression  $\Omega(t) = \Omega_m \sec h(1.76(t-t_0)/\hat{\tau}_p) \cos(\omega_c(t-t_0) + \phi(t))$ , where  $\Omega_m$  is the predicted maximum of the envelope,  $\omega_c$  is the central carrier frequency,  $t_0$  is the delay,  $\hat{\tau}_p$  is the predicted duration and  $\phi(t)$  the temporal pulse phase, we can determine the area, the center frequency and the absolute phase  $\Phi_{CE}$ , i.e.  $\phi(t_0)$ .

To determine the evolution of the absolute phase in the medium, we investigate it in detail. As an example in Fig. 1, for  $\Omega_0=1.8$  fs<sup>-1</sup> corresponding to  $5.1\pi$  and  $\epsilon=1/3$  fs<sup>-1</sup>, in the case without LFC, the absolute phase  $\Phi_{CE}$  is -0.48 rad at  $z=79$   $\mu$  m. However, when the LFC is considered,  $\Phi_{CE}$  changes into 2.75 rad at the same location, and then the difference of  $\Phi_{CE}$  is about  $1.0\pi$ , i.e. the Rabi frequency oscillates in opposite direction. When we observe the pulse at  $z=80$   $\mu$  m, the absolute phase in the case without LFC is 2.12 rad and changes into -2.07 rad in the case with LFC. So, if we confine the difference of  $\Phi_{CE}$  in the range  $(-\pi, \pi]$ , the difference of  $\Phi_{CE}$  between two cases becomes into  $0.67\pi$ , i.e. the difference changes about  $\pi/3$  during the length increasing 1  $\mu$  m. Although the difference keeps changing in the medium, it remains constant after transmitting into free space. As an example, the difference remains  $1.0\pi$  if pulses exit into free space at  $z=79$   $\mu$  m, i.e. the medium zone length  $L=39$   $\mu$  m. Numerical results show that the absolute phases of pulses transmitting into free space are constant for both cases, but the difference varies linearly in the rate of  $-0.36\pi/\mu$  m with the medium zone length increasing, see Fig. 2. The difference goes back to the same value every about 4.5  $\mu$  m and is near to zero for the medium zone length  $L=40.5$   $\mu$  m. For other input pulse and medium density, consistent conclusion can also be drawn. It is clear that the carrier-envelope phase  $\Phi_{CE}$  of the pulse is sensitive to the NDD effect.

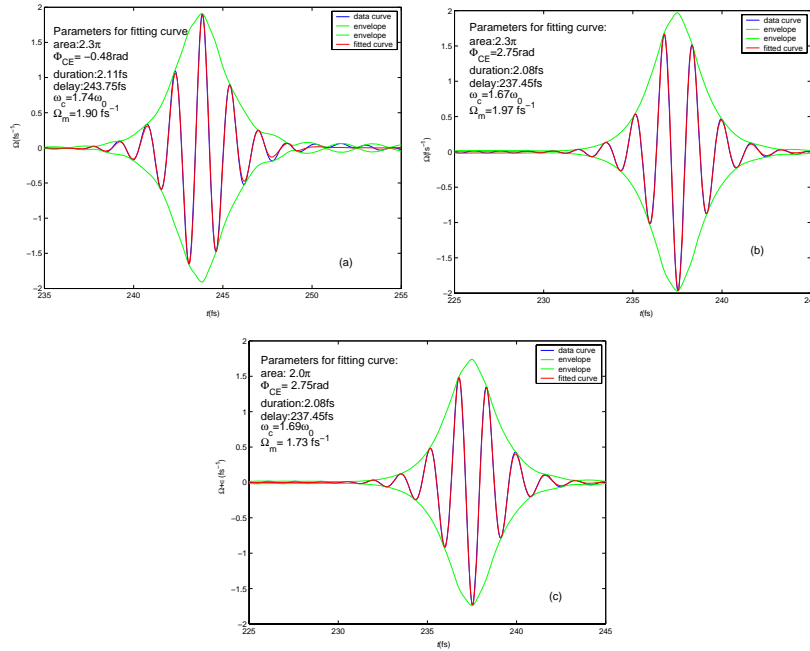


Fig. 1. Absolute phases and areas of three cases: (a) without LFC; (b) with LFC and (c)  $\Omega \in \mathcal{U}$  with LFC.

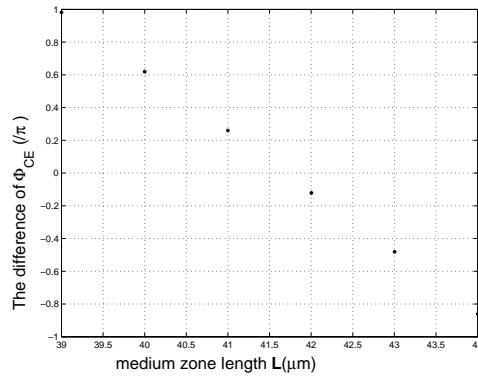
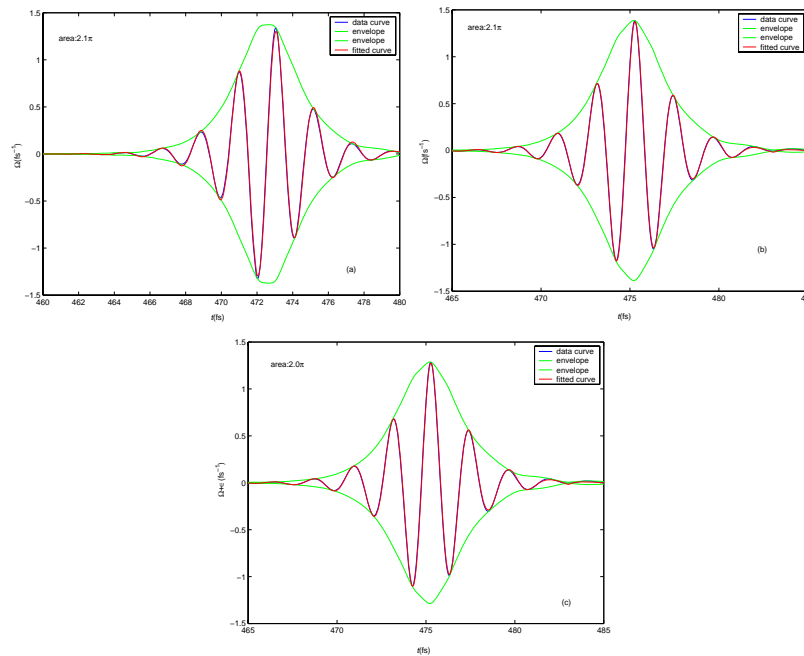


Fig. 2. Variation in the difference of  $\Phi_{CE}$  with medium zone length  $L$ .

The change of  $\Phi_{CE}$  can be interpreted as a combination result of three factors: the decreasing in the center frequency, the increasing in the group velocity and the self-phase modulation. For the case without LFC, the center frequency of the pulse is 1.74 times the resonant frequency, i.e.  $1.74\omega_0$ , but it decreases to  $1.67\omega_0$  owing to the dynamic redshift in the resonance frequency when the LFC is considered, which means that the center frequency decreases by about  $0.16\text{fs}^{-1}$ , as shown in Fig. 1. Numerical results show that the group velocity increases slightly and the self-phase modulations of  $\Omega$  for both cases are negligible. The change in the absolute phase mainly attributes to the change in the center frequency and the group velocity.

According to the area theorem [3-5], the area of stable h.s. pulses should be  $2\pi$ . However, for the few-cycle pulses propagating in a dense medium, if we follow Miklaszewski and define a generalized pulse area as Ref. [40], the area of ultrashort solitary pulses is evidently larger than  $2\pi$  whether the LFC is considered or not. Combining with the polarization, i.e.  $\Omega + \epsilon u$ , the pulse interacts with dense medium as a pulse whose area is  $2.0\pi$ , see Fig. 1. So we define an effective Rabi frequency:  $\Omega_{\text{eff}} = \Omega + \epsilon u$ , i.e. an effective coupling of light-matter. As shown in Figs. 3(a-f), for smaller density of medium  $\epsilon = 0.1 \text{ fs}^{-1}$  and weaker incident pulse  $\Omega_0 = 1.4 \text{ fs}^{-1}$  in comparison with the case in Fig. 1, the area of  $\Omega$ , no matter the LFC is considered or not, is about  $2.1\pi$ , but that of  $\Omega_{\text{eff}}$  is  $2.0\pi$ . For larger density of medium  $\epsilon = 0.4 \text{ fs}^{-1}$  and incident pulse  $\Omega_0 = 1.8 \text{ fs}^{-1}$ , the area of  $\Omega$  in both cases is about  $2.4\pi$ , while that of  $\Omega_{\text{eff}}$  is still  $2.0\pi$ . In dense medium, the area of Rabi frequency  $\Omega$  is obviously larger than the value predicted by the standard area theorem. However, numerical experiments show that, in spite the density of medium and the peak of envelope of Rabi frequency vary in a relative large range, the area of  $\Omega_{\text{eff}}$  keeps invariant area  $2\pi$ .



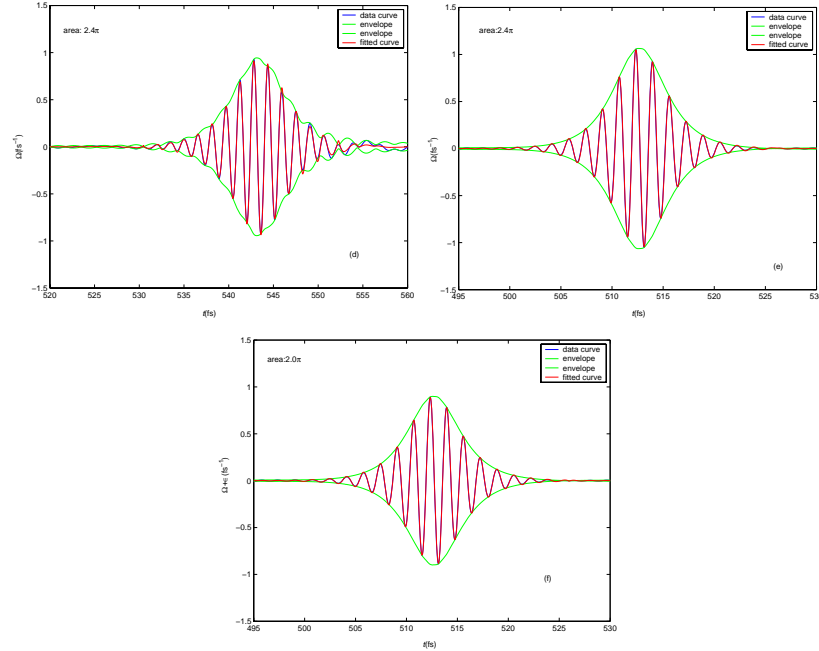
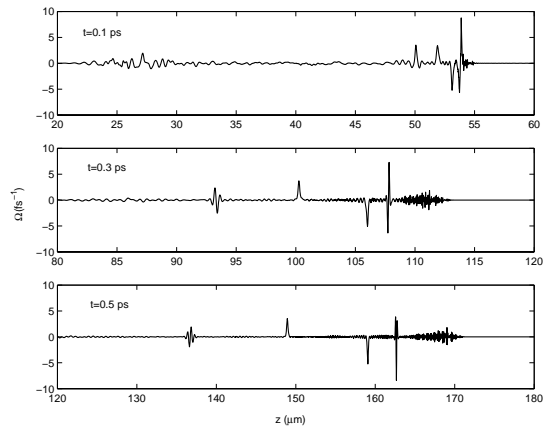


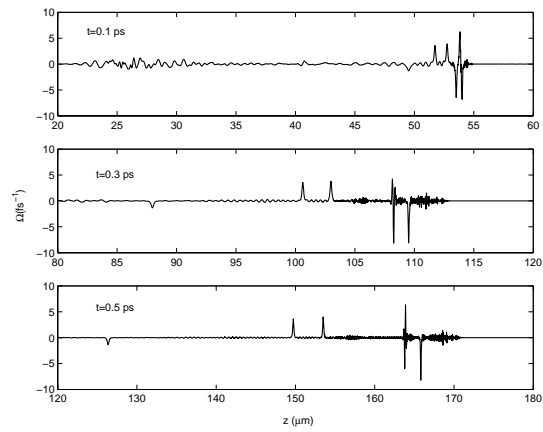
Fig. 3. Areas of  $\Omega$  and  $\Omega_{\text{eff}}$  at  $z=130\mu$  m. (a)-(c) for  $\epsilon = 0.1 \text{ fs}^{-1}$ ,  $\Omega_0=1.4 \text{ fs}^{-1}$ ; (d-f) for  $\epsilon = 0.4 \text{ fs}^{-1}$ ,  $\Omega_0=1.8 \text{ fs}^{-1}$ . (a) and (d) for  $\Omega$  in the case without LFC; (b) and (e) for  $\Omega$  with LFC; (c) and (f) for  $\Omega_{\text{eff}}$  with LFC.

Now, we focus on the influence of NDD interaction on the number of solitary pulses. It can be seen in Eq. (3) that, when the LFC is considered, the polarization  $u$  modifies the light-matter interaction: the polarization enhances the light-matter interaction when it follows the field with identical direction, and suppresses the interaction when it oscillates in opposite direction to the field. We find that there are some area thresholds of input pulse, where the NDD effect causes the number of solitary pulses, which are produced by the penetrating part, to change. As an example in Fig. 4, for larger medium density,  $\epsilon = 2/3 \text{ fs}^{-1}$ , and higher input pulse,  $\Omega_0=5 \text{ fs}^{-1}$  corresponding to  $14\pi$ , in the case without LFC, the penetrating part produces four solitary pulses: two unipolar half-cycle solitons and two high-frequency oscillating solitary pulses. However, when the LFC is considered, the effect of NDD causes that the slowest oscillating pulse changes into a unipolar half-cycle pulse, which is well investigated in Refs.[12, 41, 42], and the penetrating part produces five pulses: one oscillating solitary pulses, two positive polarity unipolar half-cycle solitons and two negative polarity unipolar solitons. For each unipolar half-cycle pulse, our numerical results and analysis expressions [12, 41] both show that the polarization follows the field quite accurately in time with identical direction. So, the polarization enhances the interaction of unipolar pulses with matter. Although the polarization follows the oscillating pulses with opposite direction and then depresses the light-matter interaction, for all pulses, the total effect enhances the interaction. Thus the same area of pulse near certain thresholds can produce more solitary pulses as compared with the case without LFC. Fixing the medium density, i.e.  $\epsilon = 2/3 \text{ fs}^{-1}$ , for weaker input pulse,  $\Omega_0=4.3 \text{ fs}^{-1}$ , as shown in Fig. 5, when the LFC is not considered, the penetrating field produces three oscillating solitary pulses. Numerical investigation shows that the polarization  $u$  well follows the pulse in time, but with opposite direction. So, the effect of NDD depresses the light-matter interaction and arouses that the penetrating part only produces two oscillating pulses. As shown by above analysis, when the area of input pulse is

close to a certain threshold, the effect of NDD could cause the number of solitary pulses to change.

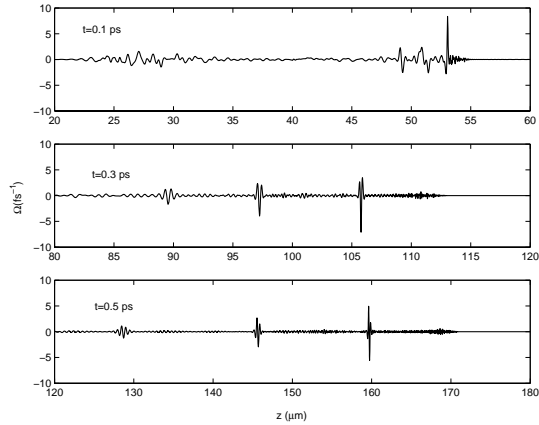


(a)

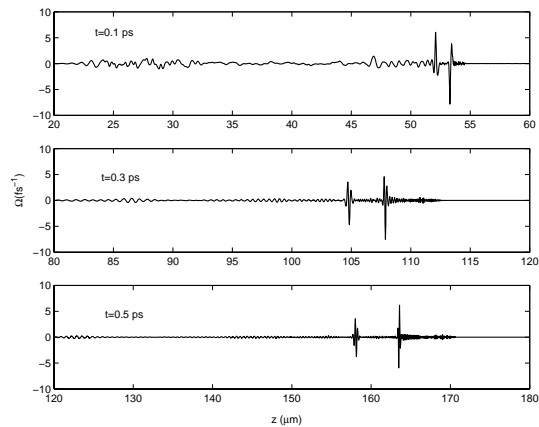


(b)

Fig. 4. Change in the number of solitary pulses in the two cases: (a) without LFC and (b) with LFC for  $\epsilon = 2/3 \text{ fs}^{-1}$ ,  $\Omega_0 = 5 \text{ fs}^{-1}$ .



(a)



(b)

Fig. 5. Change in the number of solitary pulses in the two cases: (a) without LFC and (b) with LFC for  $\epsilon = 2/3 \text{ fs}^{-1}$ ,  $\Omega_0 = 4.3 \text{ fs}^{-1}$ .

#### 4. Conclusion

With a Yee's FDTD discretization scheme and the predictor-corrector method, we numerically investigated the extreme nonlinear processes caused by a few-cycle pulse normally inputting into the surface of a dense two-level medium from free space and found that the effect of NDD arises several essential differences in the penetrating and transmitting solitary pulses' properties. Firstly, the carrier-envelope phase  $\Phi_{\text{CE}}$  of solitary pulse drastically changes as comparison with the case without LFC. At some location, the difference of  $\Phi_{\text{CE}}$  between two cases with and without LFC can go up near to  $\pi$ , i.e. the Rabi frequency oscillates in opposite direction. The absolute phases of pulses exiting into free space are constant for both cases, but the difference varies linearly with the medium zone length. The absolute phase  $\Phi_{\text{CE}}$  has essential influence on a large number of extreme nonlinear interactions owing to the extremely short duration of pulse, and then the NDD effect plays an

important role. Secondly, in a dense medium, the area of the stable state pulse, which deviates from the value predicted by the standard area theorem, is larger than  $2\pi$ . However, if we consider the effective coupling of light-matter, i.e.  $\Omega + \epsilon u$ , the area keeps  $2\pi$  in spite the medium density varies in a relative large range. Furthermore, the polarization  $u$  can enhance or depress the strength of light-matter interaction. This effect accounts for the change in the number of solitary pulses at certain input pulse area.

### **Acknowledgments**

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