Effect of time-dependent ionization on the propagation of a few-cycle laser pulse in a two-level medium

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We investigate the influence of ionization on the propagation and spectral effects of a few-cycle ultrashort laser pulse in a two-level medium. It is found that when the fractional ionization is weak, the production of higher spectral components makes no difference. However, when the two states are essentially depleted before the peak of the laser pulse, the impact of ionization on the higher spectral components is very significant.

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I. INTRODUCTION

The propagation of laser pulses in various nonlinear media has been a topic of research for many years. The resonant interaction of a laser pulse with a collection of two-level atoms has been actively researched since the milestone work of McCall and Hahn [1]. Under the rotating-wave approximation [2] and slowly-varying-envelope approximation [3], eminent effects such as self-induced transparency [4], Rabi flopping [5], etc., have been exhibited, while for propagation in other nonlinear media such as condensed media and air, several important phenomena, e.g., pulse splitting [6,7], beam filamentation [8], and white-light continuum generation [9,10], have been predicted.

Recently, with tremendous developments in ultrashort and ultraintense laser pulses, there has been increasing interest in a variety of new effects involving the interaction of intense ultrashort laser pulses with matter [11–16]. For example, Rothenberg found that the usually invoked slowly-varying-envelope approximation breaks down long before the temporal structure reaches the time scale of an optical cycle. This breakdown leads to a dramatic departure from the predicted symmetric development of the self-focusing long pulse. For short-pulse focusing, an asymmetric development of the self-focusing pulse into a temporal structure with a greatly enhanced trailing portion occurs [11]. In a two-level-atom system, Ziolkowski *et al.* have found that the time-derivative-driven nonlinearities have a significant impact on the time evolution of ultrashort laser pulse [12]. For large pulse areas, Hughes has demonstrated that the carrier-wave effects become predominant, and a different type of carrier-wave Rabi flopping occurs, which can lead to the production of significantly higher spectral components on the propagating pulse [14]. In particular, on further increasing the pulse area, frequency components up to 100 eV and transients well into the x-ray regime can be generated [15]. Carrier-wave Rabi flopping has been experimentally demonstrated in GaAs [16]. Usually, the field strengths of these few-cycle laser pulses are very high, indeed reaching laser intensities in excess of $10^{12}\ \mathrm{W/cm^2}$. For such high intensities, Tritschler *et al.* have demonstrated that the simple two-level system can still serve as a reference point [17–19]. However, for some atom mediums or much higher laser intensity, it becomes questionable whether the ionization of atoms can affect the evolution of few-cycle laser pulses in a two-level medium. Indeed, in Ref. [15] the author also writes: "It remains to be seen whether such effects will occur in real atoms where bound-bound transitions and ionization become important." In this paper, we would like to clarify this question.

Investigations have shown that ionization has significant effects on many nonlinear interaction processes [20–25]. The influence of photoionization on a stimulated Raman adiabatic passage (STIRAP) scheme in a dense medium of three-level atoms has been investigated by Buffa *et al.* [20]. They found that photoionization depletion and electron impact dephasing will be significant in maximal coherence wave mixing and STIRAP population transfer schemes operated at high density. Moreover, the effect of time-dependent ionization on the harmonics generated by bound-bound transitions was investigated by Jarque *et al.* [21]; it was found that the introduction of tunneling and above-barrier ionization rates in a simple two-level atom can induce an unexpected richness in

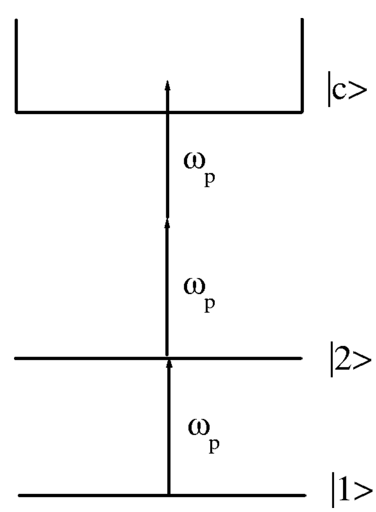

FIG. 1. Level scheme considered in this paper.

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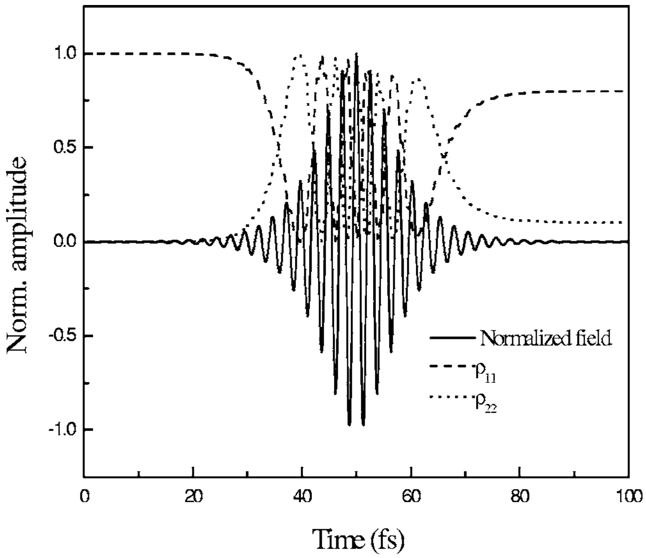

FIG. 2. The normalized electric field (solid line) and the population changes (dashed and dotted lines) near the input face of the two-level medium with $\sigma^{(2)}=1\times10^{50}$ cm^4 s.

the harmonic spectra, leading to the production of radiation at high frequencies. Such frequencies are not reached when no ionization is considered. Ionization is also believed to play an important role in the propagation of femtosecond laser pulses in a transparent medium. It can lead to a reduction of the refractive index and consequently the propagation is deeply modified [22]. Moreover, it has been demonstrated that the generation of free electrons due to multiphoton ionization is an important mechanism that limits self-focusing in condensed media [10].

Here we study qualitatively the influence of ionization on the propagation of a few-cycle laser pulse in a dense two-level medium. By using an iterative predictor-corrector finite-difference time-domain method to solve numerically the full semiclassical Maxwell-Bloch (MB) equations, which avoids the limitations of the slowly-varying-envelope and rotating-wave approximations as ultrashort laser pulses are considered [12], we found that suboptical-carrier effects can still occur when the fractional ionization is weak. However, when the states are essentially depleted before the peak of the laser pulse, the production of higher spectral components is greatly suppressed.

This paper is organized as follows. In Sec. II, we present a theoretical model of the interaction of a few-cycle laser pulse with an open two-level medium. In Sec. III, we investigate the evolution and the corresponding spectral effects of a few-cycle laser pulse for various ionization cross sections and input pulse areas in this model. Finally, we offer some conclusions in Sec. IV.

II. THEORETICAL MODEL

The system we consider is shown in Fig. 1. It has three components: two discrete levels and a continuum. The continuum can be incorporated into the two-level model through an incoherent decay (ionization) of the excited state. We ignore any possible mechanism of recombination of the ionized population to the bound state, and neglect relaxation effects such as spontaneous emission and collisional broadening because they have characteristic times much higher than those involved in our problem. This simple two-level-plus-continuum model is widely used in previous studies [21].

The propagation property of a laser pulse in a two-level-plus-continuum medium with an atomic density N can be modeled using the MB equations [21]:

$$\partial_t H_y = -\frac{1}{\mu_0}\partial_z E_x,$$

$$\partial_t E_x = -\frac{1}{\varepsilon_0}\partial_z H_y + \frac{2N\mu_{12}}{\varepsilon_0}\left(-\omega_0 u_4 - \frac{\gamma(t)}{2}u_3\right),$$

$$\dot{u}_1 = -2\frac{\mu_{12}}{\hbar}E_x u_4,$$

$$\dot{u}_2 = 2\frac{\mu_{12}}{\hbar}E_x u_4 - \gamma(t)u_2,$$

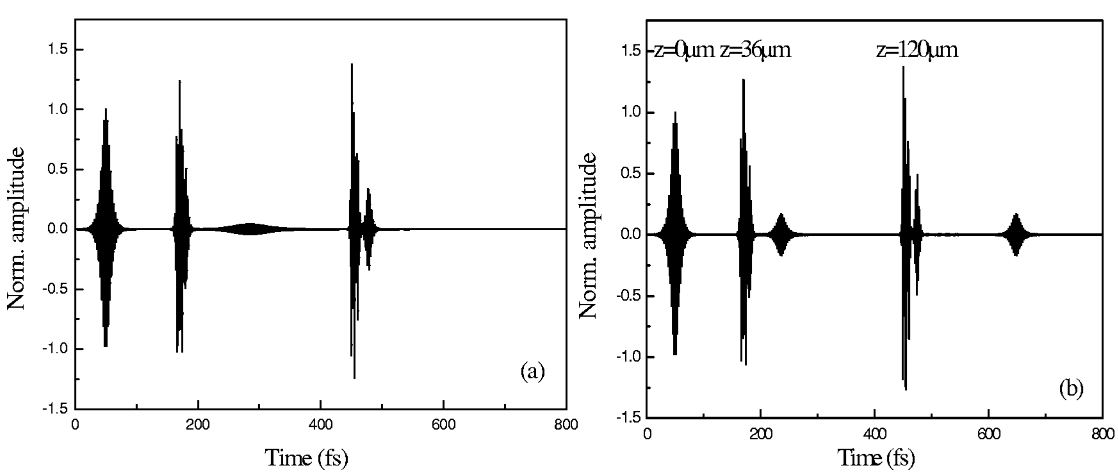

FIG. 3. 10π-pulse evolution (a) in our two-level-plus-continuum medium with $\sigma^{(2)}=1\times10^{50}$ cm^4 s; (b) in a conventional two-level medium without ionization.

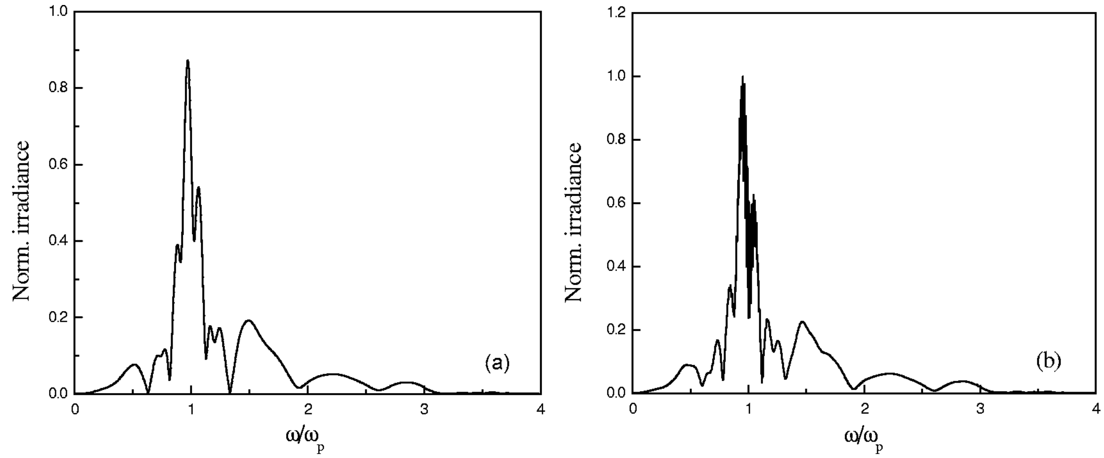

FIG. 4. The corresponding spectra at $z=120$ μm (a) for our two-level-plus-continuum model with $\sigma^{(2)}=1\times10^{50}$ cm^4 s; (b) for a conventional two-level medium without ionization.

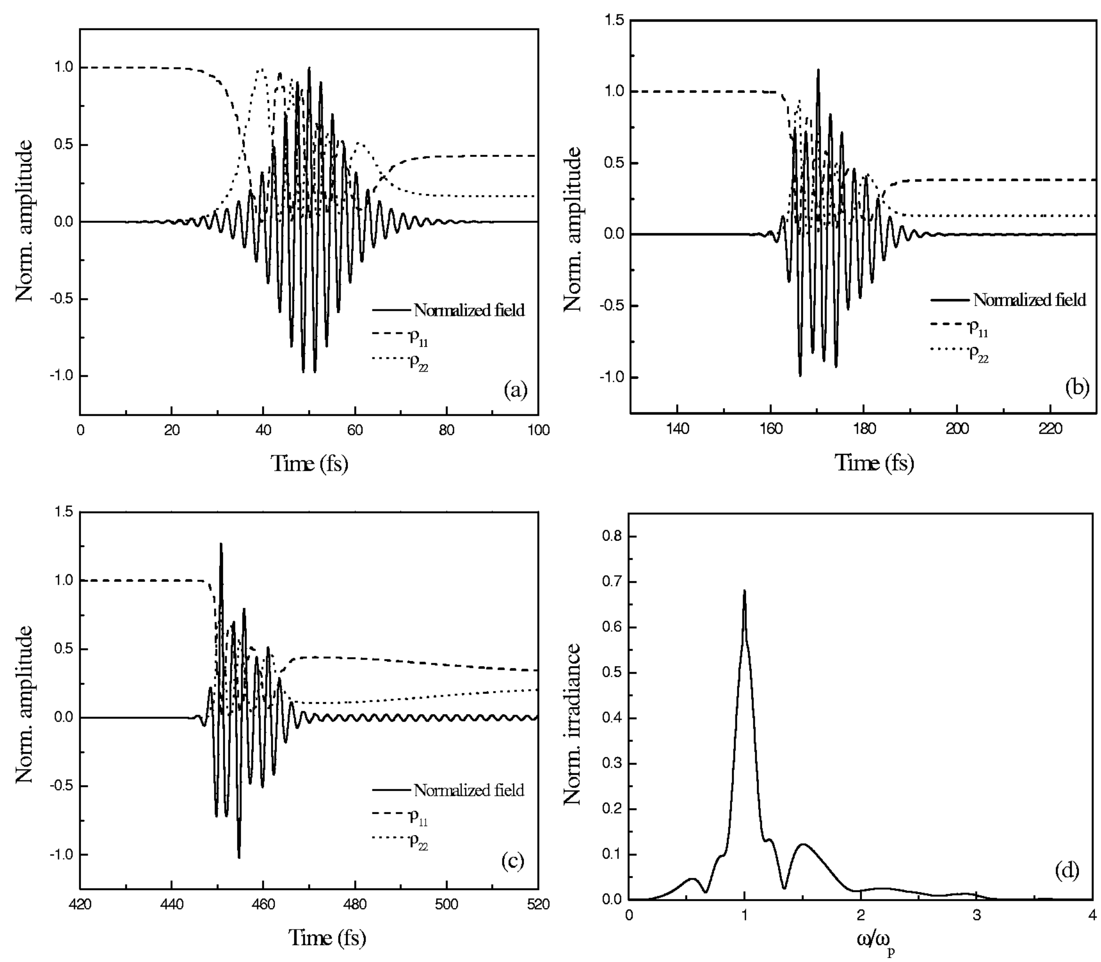

FIG. 5. (a)–(c) 10π-pulse evolution at the respective distances of 0, 36, 120 μm in our two-level-plus-continuum medium with $\sigma^{(2)}=5\times10^{50}$ cm^4 s; (d) the corresponding irradiance spectra at $z=120$ μm.

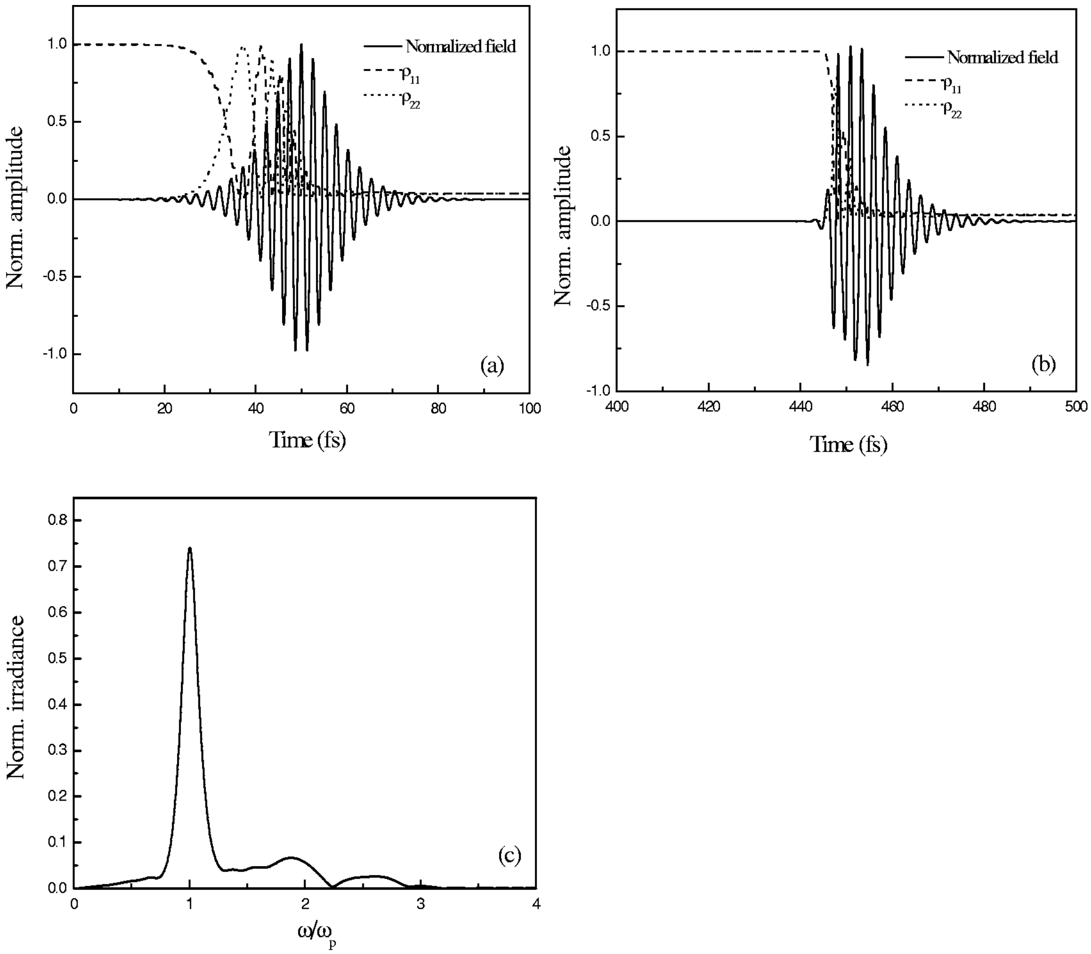

FIG. 6. (a), (b) As in Figs. 5(a) and 5(c) but for 15π-pulse propagation. (c) As in Fig. 5(d) but for 15π-pulse propagation.

$$\dot{u}_3 = -\omega_0 u_4 - \frac{\gamma(t)}{2} u_3,$$

$$\dot{u}_4 = \omega_0 u_3 - \frac{\mu_{12}}{\hbar} E_x (u_2 - u_1) - \frac{\gamma(t)}{2} u_4. \qquad (1)$$

Here E_x and H_y are the electric and magnetic fields, ω_0 is the transition frequency of the two-level medium, and $\gamma(t)$ is the time-dependent ionization rate of the excited state; $u_1 = \rho_{11}$ and $u_2 = \rho_{22}$ are, respectively, the populations of the ground and the excited levels. The macroscopic nonlinear polarization $P_x = N\mu_{12} u_3$ is related to the off-diagonal density matrix element $\rho_{12} = (u_3 + iu_4)/2$ and μ_{12} is the dipole moment. The refractive index is determined by the real part of ρ_{12} and the gain coefficient is proportional to the imaginary part of ρ_{12}.

We employ a standard finite-difference time-domain approach [12] for solving the full-wave Maxwell equations, and the predictor-corrector method to solve the Bloch equations. For the external laser pulse field, the initial condition is $E_x(t=0,z) = E_0 \operatorname{sech}[(z/c + z_0/c)/\tau]\cos[\omega(z+z_0)/c]$, where ω is the laser pulse frequency and E_0 is the amplitude of the laser pulse. $\tau_p = 2ar\cosh(1/\sqrt{0.5})\tau_0$ is the full width at half maximum of the laser pulse intensity envelope. In the numerical analysis, the medium is initialized with $u_2 = u_3 = u_4 = 0$, $u_1 = 1$ at $t = 0$. The choice of z_0 ensures that the laser pulse penetrates negligibly into the medium at $t = 0$. The parameters that we adopted are the following: $\tau_p = 10$ fs, $\omega = \omega_0 = 2.45$ fs^{-1}, $z_0 = 15$ μm, $\mu_{12} = 1.44 \times 10^{-29}$ C m, $N = 4 \times 10^{19}$ cm^{-3}. The corresponding pulse area is $A = dE_0 \tau_p \pi/\hbar \times 1.76$ and $\gamma(t) = \sigma^{(2)} I(t)^2/(\hbar\omega)^2$ (where $\sigma^{(2)}$ is the photoionization cross section of the excited state).

III. NUMERICAL RESULTS AND ANALYSIS

First we model the propagation of a 10π few-cycle laser pulse in the ionization of a two-level medium; the ionization cross section is $\sigma^{(2)} = 1 \times 10^{-50}$ cm^4 s. Figure 2 shows the electric field and the population changes near the input face of the two-level medium. In this case, the two states are not significantly depleted during the interaction, only around the peak, ionization can occur, and the fractional ionization is roughly 10%. Carrier-wave Rabi flopping is still clearly discerned. The evolution of the electric field through the medium is depicted in Fig. 3(a). For the same parameters, Fig. 3(b) shows the case for a conventional two-level medium without ionization. It can be seen that the impact of ionization on the electric field is mainly manifest in the absorption of the small splitting pulse. In the two-level-plus-continuum model, though the 10π few-cycle laser pulse can still split,

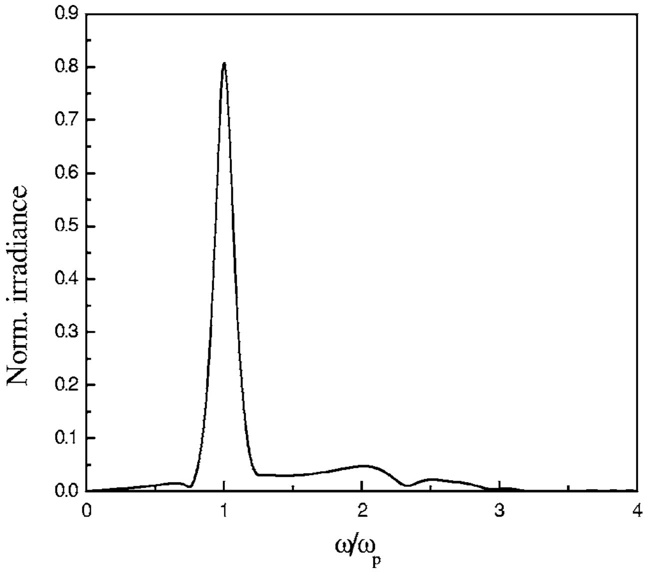

FIG. 7. As in Fig. 5(d) but for 20π-pulse propagation.

the small splitting pulse cannot drive a complete Rabi flopping due to the decrease of the population at the back edge of the laser pulse. So the small splitting pulse is much weaker than that in the nonionization case, and during the course of propagation, this small splitting pulse is gradually absorbed. The corresponding spectra are shown in Fig. 4. The influence of the introduction of the ionization on the oscillatory feature is quite apparent, while the impact on the production of higher spectral components is seen to be negligible.

As for the oscillatory feature, theoretical [26,27] as well as experimental [28] studies have been made. It was demonstrated that this feature arises as a result of the interference between the splitting pulses. It is related to the time difference t for the two splitting pulses to arrive at the observation point: the larger t is, the more obvious the oscillatory features are. When ionization is considered, the absorption of the small splitting pulse reduces the effect of the interference; hence, the oscillatory feature is much weaker.

In Fig. 5, we see how the electric field and the spectra evolve with larger ionization cross section ($\sigma^{(2)}=5\times 10^{-50}\ \mathrm{cm}^4\ \mathrm{s}$). In this case, a significant fraction of the population is ionized in the front edge of the laser pulse which induces much energy loss. Hence, during the course of propagation, the front edge of the laser pulse becomes steep, and the number of Rabi floppings becomes less and less, which directly affect the production of the higher spectral components. From Fig. 5(d) we can see that higher spectral components become much weaker than in Fig. 3(b). Moreover, due to the enhanced ionization (ionization probability is 40.6%), the splitting of the pulse becomes difficult, and hence the oscillatory features around the resonant frequency can hardly be seen. This is consistent with our analysis.

With this ionization cross section, next we increase the pulse area. Figures 6(a) and 6(b) depict the electric profile with area equal to 15π at the respective propagation distances of 0 and 120 μm. It can be seen that the states are essentially depleted around the peak of the laser pulse. Similar to Fig. 5, the front edge of the laser pulse still becomes steep during the course of propagation due to the ionization loss, while the back edge remains unchanged because the residual populations cannot induce any excitation under this situation. Figure 6(c) is the corresponding spectrum at the propagation distance $z=120\ \mu$m. The oscillatory features around the resonant frequency disappear. Comparing with Fig. 5(d), higher spectral components are slightly enhanced in this case. Moreover, due to the enhanced ionization, these higher spectral components no longer increase even if we further increase the pulse area (see Fig. 7).

IV. CONCLUSIONS

In conclusion, we carried out a numerical investigation of the influence of ionization, causing neutral atom depletion, on the propagation and spectral effects of a few-cycle laser pulse with a fairly general time-dependent two-level-plus-continuum model of multiphoton ionization. It was found that suboptical-carrier effects can still occur and the production of higher spectral components is not different if the fractional ionization is weak during the interaction. Only when the states are essentially depleted before the peak of the laser pulse is the impact of ionization on the spectra significant; the higher spectral components no longer increase even as the pulse area is increased. Hence, to achieve the soft-x-ray spectrum using this method, one must choose a special material that can sustain much higher laser intensity, or adopt some control scheme to avoid the ionization. Recently, Kuznetsova *et al.* have proposed a method which can greatly reduce the excited-state absorption by applying an additional driving laser field [29]. In our previous paper, we demonstrated that higher spectral components can be produced even for small-area pulses using two-color ultrashort laser pulses [30]. These methods greatly increase the possibility of experimental production of a soft-x-ray spectrum using suboptical-carrier transients.

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