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Two-color interference effects for ultrashort laser pulses propagating in a two-level medium

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Abstract

Using a combination of a laser pulse and its third harmonic (resonant with the medium), we investigate the two-color interference effects for ultrashort laser pulses propagating in a two-level medium. It is found that the interference effects of the two laser pulses can substantially modify the behavior of the spectra and propagation features: higher spectral components can be produced even for small pulse areas due to the interference effects of the two laser pulses. Moreover, the oscillatory structures around the resonant frequency and the propagation features of the laser pulses depend sensitively on the relative phase ϕ of the two pulses.

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Keywords: Two-color interference effects; Spectra; Ultrashort laser pulses

1. Introduction

The interaction of intense laser light with a collection of two-level atoms has been an area of active research for many years [1–3]. For the case of a single pulse tuned to a two-level resonance, the McCall–Hahn area theorem [4] shows that the temporal evolution of the pulse depends on the pulse area. It can predict and explain many fascinating effects, such as self-induced transparency and pulse compression. All these were derived within the standing slowly varying envelope approximation (SVEA) [5] and the rotating-wave approximation (RWA) [6]. However, these approximations clearly fail if the pulse duration approaches the duration of several optical cycles [7–10].

Self-induced transparency effects for 100 fs pulse in a two-level medium were investigated by Ziolkowski et al. [7]. It was found that the time-derivative behavior of the oscillatory electric field plays an essential role in the nonlinear evolution of the system, which sustains the nonlinear process through null-field points. Further consideration was made by Hughes [9], he proposed for large pulse areas, that the area under individual carriers might themselves cause Rabi flopping (RF), which can lead to carrier-wave reshaping and significantly higher spectral components and even

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soft-X-ray generation [11]. Whereas, for small pulse areas, the self-induced transparency (SIT) results are still recovered, i.e., no higher spectral components occur. Area evolution of a few-cycle pulse laser in a two-level medium was also studied by Xiao et al. [10], they found contrary to the long-pulse case, that the variation of the few-cycle pulse area is caused by the pulse splitting but not by the pulse broadening or the pulse compression. Moreover, if only a few optical cycles are contained within the pulse envelope, the carrier-envelope phase (the absolute phase) will dramatically affect the temporal shape of electric field [12,13]. Recently, Baltuška et al. [14] reported the generation of intense, few-cycle laser pulses with a stable carrier envelope phase, they also demonstrated that the few-cycle-driven coherent soft-X-ray emission from ionizing atoms depend crucially on the absolute phase, and these X-ray photons provide a sensitive and intuitive tool for determining the carrier-envelope phase.

In the case of two-color laser pulses, the pulses overlap while propagating, which can lead to interference effects if the pulses are mutually coherent. In addition, the relative phase of the two laser pulses adds an additional degree of freedom. The coherent control of intense laser–atom interactions by two strong laser fields with different frequencies has received considerable attention in recent years, such as, controlling ionization [15–20], spontaneous radiation [21], population inversion [22], high-order harmonic generation (HHG) [1,23–28], propagation of pulses in the medium [29–31], etc. However, for ultrashort laser pulse, only a few theoretical studies have investigated using this control scheme. Recently, Bandrauk and Shon [32] investigated photoionization and HHG in molecular systems under the action of combined infrared femtosecond (fsec) and ultraviolet attosecond (asec) laser pulses. It was shown that punctual "turn-on" of an asec pulse can trigger the ionization process, which in turn leads to significant enhancement of HHG. Moreover, Brown and Meath [33] proposed a two-color scenario for the excitation of dipolar molecules, and demonstrated that the state populations depend on probe laser's absolute phase and not on the relative phase difference between the two lasers. Furthermore, two-color 100 fs laser phase control in the total ion yield of ionization in a tunneling regime was investigated by Watanabe et al. [34]. They found that adding the third harmonic with an intensity of only 10% enhances the ion yield by a factor of 7, and the intensities of high-order harmonics in the plateau region are enhanced by an order of magnitude.

In this Letter, we apply a laser pulse with frequency ω_L and its third harmonic with frequency $3\omega_L$ (resonant with the medium) to investigate the two-color interference effects for ultrashort laser pulses propagating in a two-level medium. By solving the full Maxwell–Bloch (M–B) equations which avoid the limitations of SVEA and RWA as ultrashort laser pulses are considered [5,7,8,35], we demonstrate that higher spectral components can be produced even for small pulse areas due to the interference effects of the two laser pulses. Moreover, the oscillatory structures around the resonant frequency and the propagation features of the laser pulses depend sensitively on the relative phase ϕ of the two pulses.

This Letter is organized as follows: in Section 2, we present a theoretical model of the interaction of few-cycle pulse with a two-level medium. In Section 3, we investigate the coherent control of spectra using a combination of the fundamental field and its third harmonic field. Finally, we offer some conclusions in Section 4.

2. Theoretical model

We consider the system shown in Fig. 1. A pulse laser with frequency ω_L and its third harmonic with frequency $3\omega_L$ interact with a two-level medium. The propagation property of a laser pulse in a two-level medium with an atomic density N can be modeled

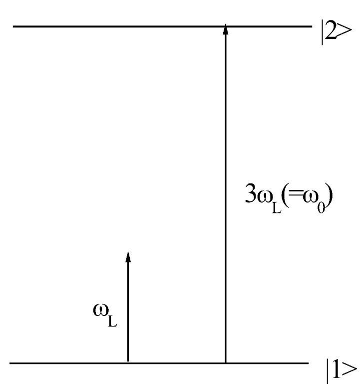

Fig. 1. Level scheme considered in this Letter.

using the M–B equations [2,10]

$$\partial_t H_y = -\frac{1}{\mu_0}\partial_z E_x, \qquad \partial_t E_x = -\frac{1}{\varepsilon_0}\partial_z H_y - \frac{1}{\varepsilon_0}\partial_t P_x,$$
$$\partial_t u = -\gamma_2 u - \omega_0 v,$$
$$\partial_t v = -\gamma_2 v + \omega_0 u + 2\frac{d}{\hbar}E_x w,$$
$$\partial_t w = -\gamma_1(w - w_0) - 2\frac{d}{\hbar}E_x v, \tag{1}$$

where E_x and H_y are the electric and magnetic fields, γ_1 and γ_2 are the population and polarization relaxation constants, respectively. ω_0 is the transition frequency of the two-level medium. The macroscopic nonlinear polarization $P_x = N\,du$ is related to the off-diagonal density matrix element $\rho_{12} = (u + iv)/2$ and the population difference $w = \rho_{22} - \rho_{11}$ between the upper and lower states, d 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 [7] for solving the full-wave Maxwell equations, and predictor-corrector method to solve the Bloch equations. The time and space increments Δt and Δz are chosen to ensure $c\Delta t \leqslant \Delta z$ [36]. For the external laser pulse field, the initial condition is

$$\begin{aligned} E_x(t=0,z) = E_{01}\,&\mathrm{sech}\big[(z/c + z_0/c)/\tau_{01}\big] \\ &\times \cos\big[\omega_L(z+z_0)/c\big] \\ &+ E_{02}\,\mathrm{sech}\big[(z/c + z_0/c)/\tau_{02}\big] \\ &\times \cos\big[3\omega_L(z+z_0)/c + \phi\big], \end{aligned}$$

where ω_L is the fundamental laser pulse frequency, E_{01} and E_{02} are the amplitudes of the fundamental laser pulse and the third harmonic pulse, respectively. $\tau_{p1} = 2ar\cosh(1/\sqrt{0.5})\tau_{01}$ and $\tau_{p2} = 2ar \times \cosh(1/\sqrt{0.5})\tau_{02}$ are the full width at half maximum (FWHM) of the two laser pulses intensity envelopes, and ϕ is the relative phase of the two laser pulses. In the numerical analysis, the medium is initialized with $u = v = 0$, $w_0 = -1$ at $t = 0$. The choice of z_0 ensures that the laser pulse penetrates negligibly into the medium at $t = 0$. Considering the conditions of SIT, we choose that the relaxation times are much longer than the input laser pulse duration and adopt the following pulse and material parameters: $\tau_{p1} = \tau_{p2} = 10$ fs, $\omega_0 = 1.8\ \mathrm{fs}^{-1}$, $\omega_L = 0.6\ \mathrm{fs}^{-1}$, $z_0 = 15\ \mu\mathrm{m}$, $d = 2.65$ eÅ, $N = 2\times 10^{18}\ \mathrm{cm}^{-3}$, $\gamma_1^{-1} = \gamma_2^{-1} = 1$ ns, $w_0 = -1$. The two pulse areas are $A_1 = dE_{01}\tau_{p1}\pi/\hbar$ and $A_2 = dE_{02}\tau_{p2}\pi/\hbar$, respectively. The result to follow can be scaled to various laser and material parameters.

3. Numerical results and analysis

In this section we present representative numerical solutions of the coupled M–B equations given by Eq. (1). We first model the propagation of laser pulse for the case in which only one-color pulse is injected to the medium. Fig. 2(a) depicts the electric field profile (solid line) of the resonant third harmonic with pulse area equals to 4π at the input surface of the nonlinear medium. The driven density shows two symmetric transversals between the ground and excited states, and drives two complete RF (dash line). As a consequence, the propagating laser pulse evolves into two separate pulses with different profiles (see Fig. 2(b)), this is consistent with that of Hughes's [9], i.e., standard SIT 4π pulse is essentially reproduced with minor modifications due to local carrier effects. The corresponding spectrum is presented in Fig. 2(c). Higher spectra components can hardly be seen for this small pulse area, and the spectrum shows an oscillatory feature at around $3\omega_L$, which becomes more evident with further propagations. As for the oscillatory feature, theoretical [37,38] as well as experimental [39] studies have been made. It was demonstrated that this feature arises as a result of the pulse shaping that occurs as the pulse propagation through the system. Moreover, it is related to the time difference τ for the two splitting pulses to arrive at the observation point: the larger the τ is, the more obvious the oscillatory features are, and our numerical results are in accord with theirs.

When only the nonresonant fundamental pulse is considered (see Fig. 3), the pulse laser is difficult to split because of a large detuning. Slightly carrier-wave reshaping can be found during the course of propagating, which result in a much weaker third harmonic in the corresponding spectrum (see Ref. [40]).

Now we turn on both laser pulses. The interference effect between two laser pulses can lead to a larger modification of the temporal shape of electric field. Moreover, this modification depends sensitively on the relative phase of the two laser pulses (see Fig. 4). Be-

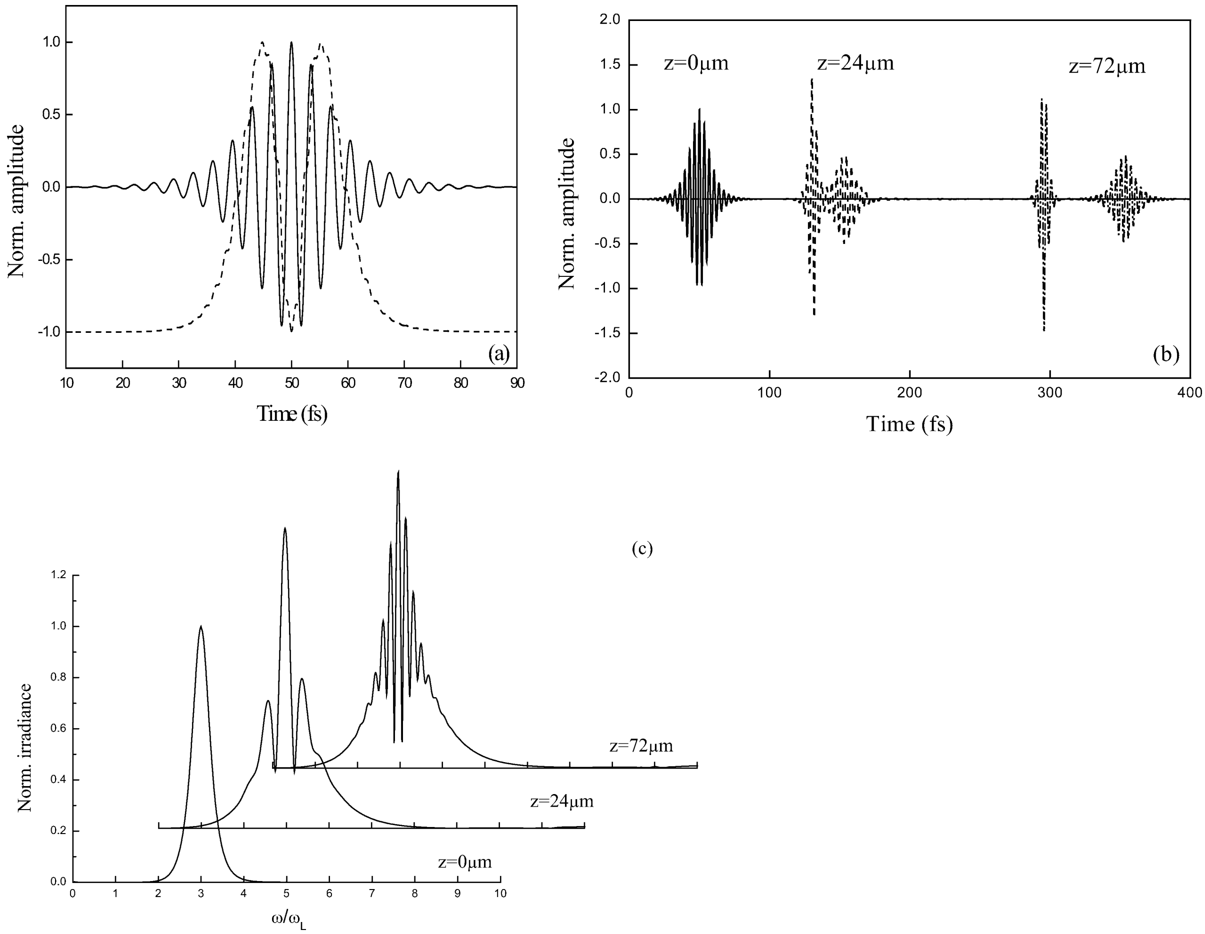

Fig. 2. (a) The normalized electric field (solid line) and population difference (dash line) of 4π pulse at the input surface of the nonlinear medium. (b) Normalized electric field at the respective distances of 0, 24 and 72 μm. (c) The corresponding spectrum of 4π pulse at the same propagation distance.

cause intense laser–matter interactions depend on the electric field of the pulse, the interference effect will dramatically affect the spectral and propagation features during the course of propagation. Fig. 5(a)–(d) presents the spectra produced by the two-color laser pulses ($A_1 = A_2 = 4\pi$) at the propagation distance of $z = 72$ μm for different initial relative phases of the pulses $\phi = 0$ and $\pi/2$, π, respectively. The interference effects of two-color laser pulses on the spectra are clearly seen. Higher spectral components (from $4\omega_L$ to $12\omega_L$) are generated due to the ω_L–$3\omega_L$ mixing. Moreover, the oscillatory features at around $3\omega_L$ (resonant frequency) dependent crucially on the relative phase: the number of oscillations decreases with ϕ increased from 0 to π.

Fig. 6 depicts the electric profiles of the two-color pulses ($A_1 = A_2 = 4\pi$) at the propagation distance of $z = 72$ μm with the relative phase equals to 0 and π. It is known that a 4π pulse with resonant frequency can evolve into two 2π pulses with long propagation distance. Hence, there is a weaker 2π pulse splitted from the main pulse in all these three figures. Moreover, strong carrier-wave reshaping is indeed found in the main pulses. However, there are discrepancies among them: for $\phi = 0$, the weak 2π pulse's propagation velocity is much slower than for $\phi = \pi$, i.e., the time of the pulse to arrive at the observation point is much longer for $\phi = 0$ than $\phi = \pi$. Similar results can be obtained for other combination of pulse areas.

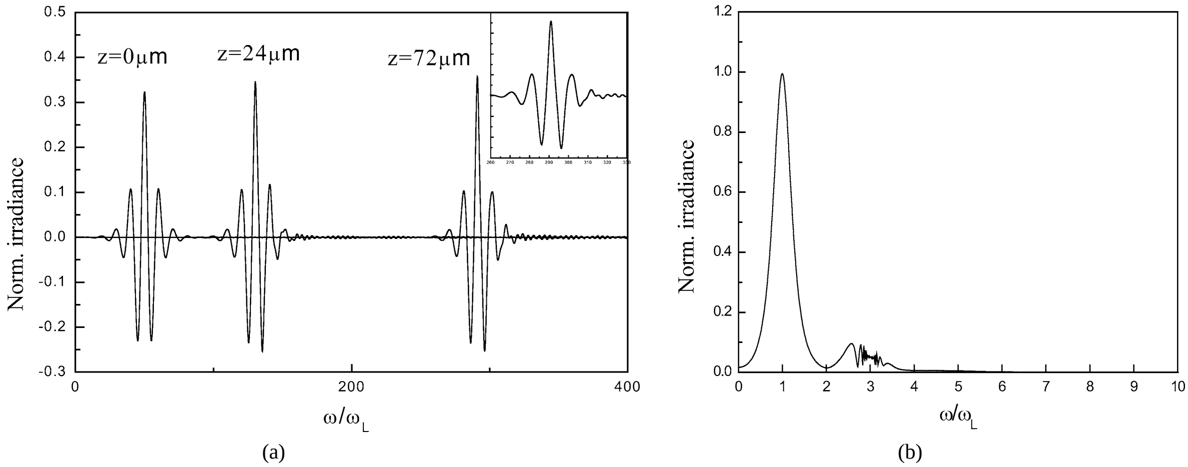

Fig. 3. (a) As in Fig. 1(a) but for fundamental nonresonant laser pulse. (b) The corresponding spectrum of 4π pulse at $z = 72$ μm. The enlargement of laser pulse at $z = 72$ μm is shown in the inset to (a).

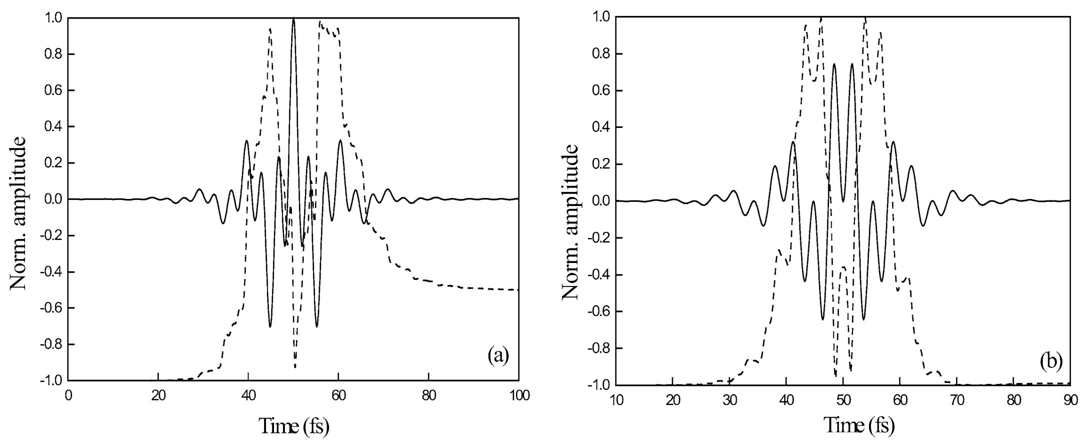

Fig. 4. As in Fig. 1(a) but for two-color 4π few-cycle laser pulses: (a) for $\phi = 0$, (b) for $\phi = \pi$.

These phenomena can be qualitatively interpreted by the electric profiles of these two pulses at the input surface of the nonlinear medium. In Fig. 4, we can see that incomplete Rabi flops occur instead of the anticipated complete ones due to the interference effect between the two laser fields (dash line), i.e., the local carrier-wave RF occurs, which result in carrier reshaping and subsequently to the production of significantly higher spectral components on the propagating laser pulses. Moreover, for $\phi = 0$, more energy is concentrated in the central line, while for $\phi = \pi$, the peaks are symmetrically located around the central line. Hence, the dispersion effect with $\phi = 0$ is stronger than that for $\phi = \pi$ (see Ref. [41]). As a result, the propagation velocity of the weak 2π pulse splitted from the main pulse with $\phi = 0$ is much slower than that for $\phi = \pi$. This is agreed with that in Fig. 5: the oscillatory features of the spectra at around $3\omega_L$ (resonant frequency) is more intense for $\phi = 0$ than that for $\phi = \pi$.

4. Conclusions

In conclusion, we investigated the two-color interference effects for ultrashort laser pulses propagating in the two-level medium by solving the full M–B equations using the ω–3ω combination scenario. It was

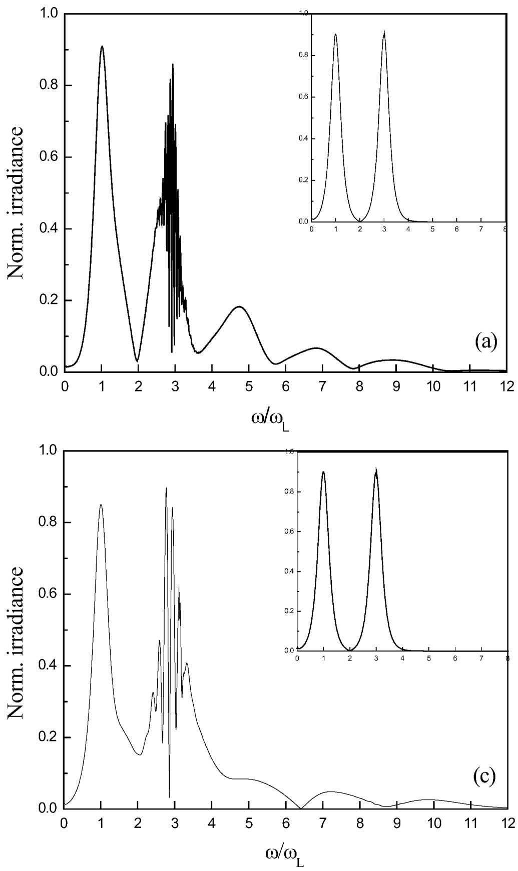

Fig. 5. Irradiance spectra of two-color 4π few-cycle pulses at $z = 72$ µm for different relative phases: (a) for $\phi = 0$, (b) $\phi = \pi/2$, (c) $\phi = \pi$. The initial spectra of them are shown in the inset, respectively.

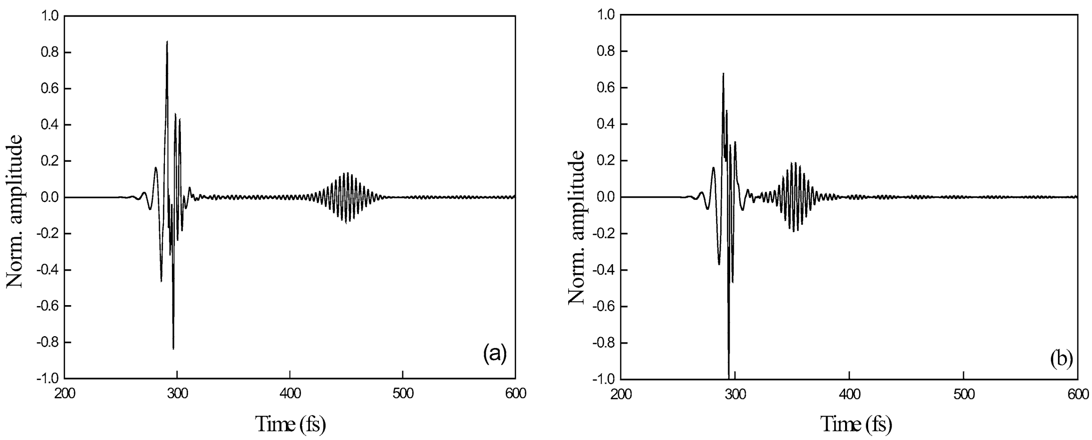

Fig. 6. The normalized electric field of 4π pulses at the propagation distances of 72 µm: (a) for $\phi = 0$, (b) $\phi = \pi$.

demonstrated that the interference effects of the two laser pulses can substantially modify the behavior of the spectra and propagation features: higher spectral components can be produced even for small pulse areas due to the interference effects of the two laser pulses. Moreover, the oscillatory structures around the resonant frequency and the propagation features of the laser pulses depend sensitively on the relative phase ϕ of the two pulses.

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