Ultrafast Fano Resonance between Optical Phonons and Electron-Hole Pairs at the Onset of Quasiparticle Generation in a Semiconductor

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Based on the many-body time-dependent approach applied to the ultrafast time region, we investigate the dynamics of creation of an optical phonon incorporating with the electron-hole continuum in a semiconductor. In the transient Fano resonance, due to an interference between those sharp (optical phonon) and continuum (electron-hole pair) quasiparticles, we find the robust destructive interference at birth of them, i.e., $\tau \approx 0$ if the created phonon is coherent under the irradiation of ultrashort optical pulses. The origin is found to be the potential scattering of the electron-hole pair by the $\mathbf{q} = \mathbf{0}$ coherent phonon. This finding agrees well with the recent experiment.

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Asymmetric Fano resonance in the linear optical spectra is one of the most dramatic phenomena of quantum interference in solid state physics [1]. It occurs when two quantum mechanically coupled transition paths from a given ground state to a localized state (sharp transition) and to a continuum of states (continuum transition) interfere with each other. According to the recent progress in the time-domain study by the generation of ultrashort laser pulses [2,3], time-resolved Fano resonance provides a new possibility to observe both build-up and decay of nonstationary coherent dynamics between competing degrees of freedom [4–6]. Fano profiles observed in a variety of phenomena [7,8] could be reinterpreted by the transient coherence in the time-resolved study.

The optical pumping of a semiconductor with a shorter pulsed femtosecond laser than a phonon period leads to the coherent oscillation in the optical properties like reflectivity or transmission [9]. This has been observed both in the direct gap semiconductor (GaAs) [10] and the indirect gap semiconductor (Ge) [11]. It signifies that the $\mathbf{q} = \mathbf{0}$ phonon mode is excited coherently—the coherent phonon is always the $\mathbf{q} = \mathbf{0}$ mode in those semiconductors—by the sudden screening of the surface space-charge field which couples to the polarization field of the longitudinal optical (LO) phonon in GaAs [12] and by the deformation potential interaction in Ge [13]. The coherent LO phonon with its frequency $\omega_{\rm LO}$ satisfies

$$\partial^2/\partial\tau^2 D_{\mathbf{q}=\mathbf{0}} + \omega_{\rm LO}^2 D_{\mathbf{q}=\mathbf{0}} = WN(\tau), \qquad (1)$$

where $D_{\mathbf{q}=\mathbf{0}}$ is the amplitude of the coherent $\mathbf{q} = \mathbf{0}$ phonon, $N(\tau)$ is the total number of photoexcited electron-hole pairs, and W is the constant depending on the electron-phonon interaction [9]. $WN(\tau)$ gives the displacivelike driving force acting on the ions. Ultrafast carrier dynamics in those systems is in fact governed by transient changes of the optical indexes [14]. Recently, Hase *et al.* [15] have explored the coherent response of Si to excitation with a 10-fs laser pulse, where the generation of the coherent phonon is driven with its subsequent dressing by electron-hole pairs. In the system, the Fano interference between responses of channels through the phonon and the electron-hole pair is observed in the frequency-time space. Interestingly, the Fano interference is always destructive at birth of the phonon and the electron-hole pair, i.e., $\tau \approx 0$.

It is a powerful idea in the ultrafast dynamics to solve the time-dependent Schrödinger equation in the many-body Hilbert space spanning the whole system, called many-body time-dependent diagonalization (MTD) [6,16]. In this Letter, we elucidate the carrier-lattice ultrafast dynamics induced by the ultrashort laser pulse in a semiconductor exploiting MTD. The transient Fano interference between those sharp (optical phonon) and electronic continuum excitations at the very early stage is examined. Especially, we address that, provided the corresponding optical phonon being coherent, the Fano interference always starts from the destructive one.

Starting from the Fröhlich Hamiltonian describing the polar coupling in a semiconductor [17], we combine it with the light-matter interaction

$$\begin{aligned}\mathcal{H} = &\sum_{\mathbf{k}} \varepsilon_{\mathbf{k}}^{v} d_{\mathbf{k}}^{\dagger} d_{\mathbf{k}} + \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}}^{c} c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}} + \omega_{\rm LO} \sum_{\mathbf{q}} a_{\mathbf{q}}^{\dagger} a_{\mathbf{q}} \\ &+ \sum_{\mathbf{q}} \sum_{\mathbf{k}} M_{\mathbf{q}} c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}+\mathbf{q}} (a_{-\mathbf{q}} + a_{\mathbf{q}}^{\dagger}) \\ &+ \sum_{\mathbf{k}} \mathbf{j}(\mathbf{k}) \cdot \mathbf{A}_0 e^{i\omega\tau} c_{\mathbf{k}}^{\dagger} d_{-\mathbf{k}}^{\dagger} \Theta_0(\tau) + \text{H.c.} \\ &+ \sum_{\mathbf{k}} \mathbf{j}(\mathbf{k}) \cdot \mathbf{A}_1 e^{i\omega(\tau-\delta\tau)} c_{\mathbf{k}}^{\dagger} d_{-\mathbf{k}}^{\dagger} \Theta_1(\tau-\delta\tau) + \text{H.c.}, \qquad (2)\end{aligned}$$

where $d_{\mathbf{k}}^{\dagger}(d_{\mathbf{k}})$ is the hole operator for the valence band $\varepsilon_{\mathbf{k}}^{v}$ and $c_{\mathbf{k}}^{\dagger}(c_{\mathbf{k}})$ is the electron operator for the conduction band

$\varepsilon^c_{\mathbf{k}}$, while $a^\dagger_{\mathbf{q}}(a_{\mathbf{q}})$ is the phonon operator for the LO phonon $\omega_{\rm LO}$. The electron in the conduction band interacts with the optical phonon via the coupling matrix $M_{\mathbf{q}}$ ($\propto 1/|\mathbf{q}|$). The last two terms in $\mathcal{H}$ are from the light-matter interaction by two ultrashort pulses. ω is the energy of the laser. The length of pumping pulse is given by Δ_0 in $\Theta_0(\tau) = \Theta(\tau) - \Theta(\tau - \Delta_0)$ ($\Theta(\tau)$: Heaviside step function). In the same way, Δ_1 is the length of the probing pulse. $|\mathbf{A}_0|$ and $|\mathbf{A}_1|$ are intensities of two pulses, respectively, and $\mathbf{j}(\mathbf{k})$ is the current. $\delta\tau$ is the time delay between two pulses.

By turning on the optical pumping in a semiconductor at $\tau = 0$, the electron-hole pair would be excited and the electron in the conduction band undergoes scattering. Such dynamics can be described by the state of the whole system $|\Psi(\tau)\rangle$ with $|\mathbf{A}_0| \to 0$ and $|\mathbf{A}_1| \to 0$,

$$|\Psi(\tau)\rangle = C(\tau)|0\rangle_c|0\rangle_v|0\rangle_{\rm ph} + \sum_{\mathbf{q}} C_{\mathbf{q}}(\tau)|0\rangle_c|0\rangle_v|\mathbf{q}\rangle_{\rm ph} + \sum_{\mathbf{k}\mathbf{k}'} C_{\mathbf{k};\mathbf{k}'}(\tau)|\mathbf{k}\rangle_c|\mathbf{k}'\rangle_v|0\rangle_{\rm ph} + \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} C_{\mathbf{k};\mathbf{k}';\mathbf{q}}(\tau)|\mathbf{k}\rangle_c|\mathbf{k}'\rangle_v|\mathbf{q}\rangle_{\rm ph}. \quad (3)$$

In $|\Psi(\tau)\rangle$, we note $|\mathbf{k}\rangle_c = c^\dagger_{\mathbf{k}}|0\rangle_c$, $|\mathbf{k}\rangle_v = d^\dagger_{\mathbf{k}}|0\rangle_v$, and $|\mathbf{q}\rangle_{\rm ph} = a^\dagger_{\mathbf{q}}|0\rangle_{\rm ph}$ for a single electron, hole, and phonon state, respectively. The many-body Hilbert space spanned by $|\Psi(\tau)\rangle$ makes us envisage the one-phonon Raman process. The initial condition $|\Psi(0)\rangle$ should be the ground state of the system before irradiation, therefore we have $|\Psi(0)\rangle = |0\rangle_c|0\rangle_v|0\rangle_{\rm ph}$. The time-dependent Schrödinger equation $i\partial/\partial\tau|\Psi(\tau)\rangle = \mathcal{H}|\Psi(\tau)\rangle$ now gives infinitely many coupled differential equations for $C(\tau)$, $C_{\mathbf{q}}(\tau)$, $C_{\mathbf{k};\mathbf{k}'}(\tau)$, and $C_{\mathbf{k};\mathbf{k}';\mathbf{q}}(\tau)$. It is the basic scheme of MTD to solve those coupled differential equations. From the solution, we can calculate the time-dependent Raman scattering cross section $P(\tau)$ with $\bar{P} = \sum_{\mathbf{q}}[a_{\mathbf{q}} + a^\dagger_{\mathbf{q}}]$,

$$P(\tau) = \langle\Psi(\tau)|\bar{P}|\Psi(\tau)\rangle.$$

$P(\tau)$ is equivalent to a usual correlation function for the Raman scattering [18]. $P(\tau)$ is given by $P_0(\tau) + P_1(\tau)$,

$$P_0(\tau) = \sum_{\mathbf{q}}[C^*(\tau)C_{\mathbf{q}}(\tau) + C(\tau)C^*_{\mathbf{q}}(\tau)],$$
$$P_1(\tau) = \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}}[C^*_{\mathbf{k};\mathbf{k}'}(\tau)C_{\mathbf{k};\mathbf{k}';\mathbf{q}}(\tau) + C_{\mathbf{k};\mathbf{k}'}(\tau)C^*_{\mathbf{k};\mathbf{k}';\mathbf{q}}(\tau)]. \quad (4)$$

An observation of the coherent LO phonon by the reflectivity is made by the first-order Raman tensor $\partial\chi/\partial Q$, where χ is the linear susceptibility and Q is the phonon coordinate, and therefore, the Raman scattering cross section gives the valid observable [19].

As manifested in $\mathcal{H}$, the photocarrier in the system is assumed to be the electron rather than the hole. Thus, the hole would not affect much the relevant dynamics at least in the n-type semiconductor in our assumption. In addition, we impose an approximation of an infinitely massive hole, corresponding to neglecting the $\mathbf{k}$-dependency of the hole [20]. As an example, we take the necessary material parameters from GaAs and thus we have $\varepsilon^c_{\mathbf{k}} = \mathbf{k}^2/2m_e$ ($m_e \approx 0.1$) and the energy gap $\varepsilon_G \approx 1.4$ eV. We also have $\omega_{\rm LO} = 36$ meV ($= 8.7$ THz) from GaAs. In addition, for the laser parameters, we take $\omega = 3.81$, 4.08, and 4.35 eV larger than ε_G and $\Delta_0 = 10$ fs and $\Delta_1 = 2.5$ fs with $|\mathbf{A}_0| = 10|\mathbf{A}_1|$ for the pump and the probe lights. The choice of much higher energy ω than the direct band gap in GaAs guarantees the photoexcitation of carriers above the band gap, which predominates the screening of the surface space-charge field to induce the coherent LO phonon [12], not associated with underlying band structures, e.g., resonance effects.

It is worth noting that our formulation is nonperturbative, so $P_0(\tau)$ and $P_1(\tau)$ in principle include all the high order responses related to combination of pumping and probing action. Nevertheless, perturbative diagrams illustrated in Fig. 1 are useful to figure out which physical processes dominantly occur in $P_0(\tau)$ and $P_1(\tau)$. Figure 2 shows the time-dependent Raman cross section $P_1(\tau)$ and $P_0(\tau) + P_1(\tau)$. $P_0(\tau)$ gives a monotonous oscillation with the frequency $\omega_{\rm LO}$ by $|0\rangle_c|0\rangle_{\rm ph} \to |0\rangle_c|\mathbf{q}\rangle_{\rm ph}$, as implied by Fig. 1(a) (we hereafter suppress the hole degree of freedom unless mentioned otherwise). On the other hand, $P_1(\tau)$ contains the electronic continuum excitation (the intraband transition) through the virtual processes $|\mathbf{k}\rangle_c|0\rangle_{\rm ph} \to |\mathbf{k}'\rangle_c|0\rangle_{\rm ph}$, where $|\mathbf{k}'\rangle_c|0\rangle_{\rm ph}$ ends in $|\mathbf{k}\rangle_c|\mathbf{q}\rangle_{\rm ph}$ coupled by the phonon final state interaction, as implied by Fig. 1(b). $P_0(\tau)$ and $P_1(\tau)$ interfere with each other to form the one-phonon Raman process, which should be noted to be the Fano interference.

More insight into the early-time dynamics of the transient Fano interference can be obtained by the continuous wavelet transformation (CWT). In Fig. 3, we provide the intensity distribution in the frequency-time domain, obtained by CWT. As shown in the left panels of Fig. 3, the CWT decomposes the signal into the broad band response ($0 \sim 30$ THz) near $\tau = 0$ and the LO phonon response at ≈ 8.7 THz. The broad band response near $\tau = 0$ is mainly from the response of the electron-hole pair and is rapidly suppressed by the pair dephasing in the time scale of hundreds of femtoseconds. The LO phonon response is from $P_0(\tau)$. Interestingly, the total response in the left

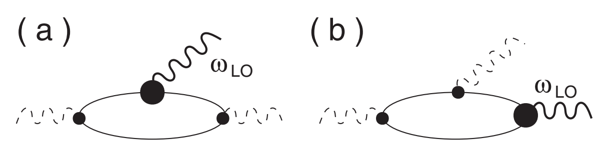

FIG. 1. Diagram for the one-phonon Raman process. The solid wavy line is the phonon $\omega_{\rm LO}$ and the dashed wavy line the photon. The small circle and the large circle represent the light-matter vertex and the electron-phonon vertex, respectively.

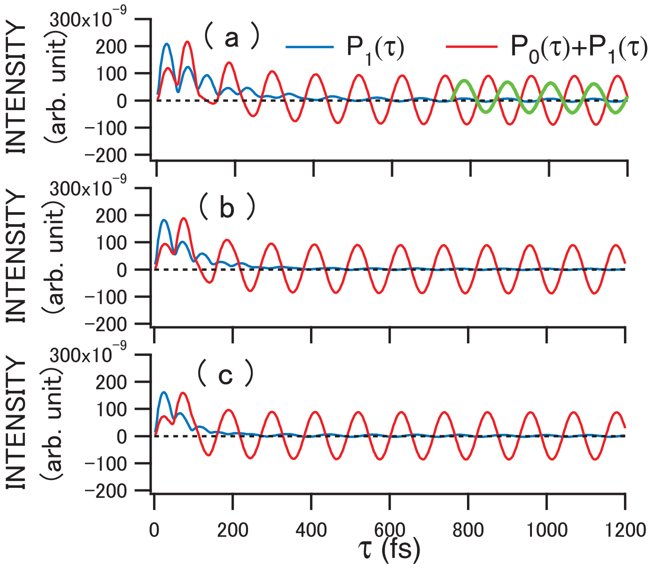

FIG. 2 (color). Time-dependent Raman cross section $P_1(\tau)$ and $P_0(\tau) + P_1(\tau)$ by Eq. (4). (a) $\omega = 3.81$ eV, (b) $\omega = 4.08$ eV, and (c) $\omega = 4.35$ eV for the energy of the laser. The thick green line in (a) is the tail of $P_1(\tau)$ magnified by 10 times.

panels of Fig. 3 is found to have the obvious dip (or antiresonance) near $\tau = 0$, slightly above the phonon mode frequency. This feature can be understood from the destructive Fano interference between responses of the electron-hole pair and the phonon. The dip at birth of the electron-hole pair and the phonon (i.e., at $\tau \approx 0$) is found robust as shown in Fig. 3(a)–3(c). It is to be noted that the

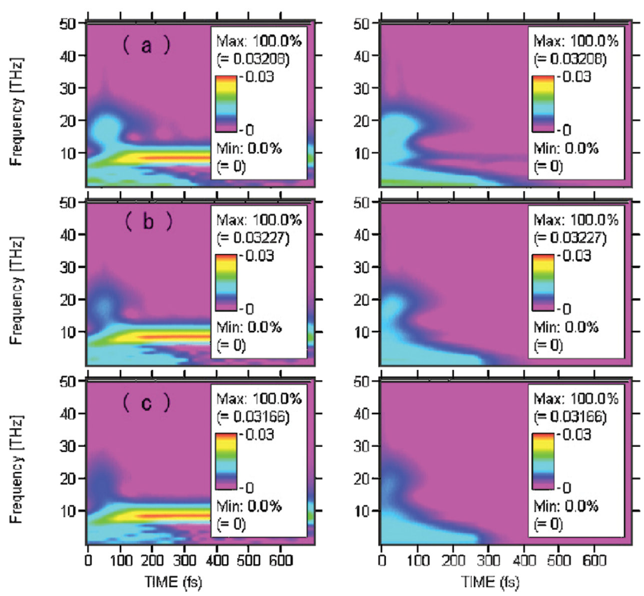

FIG. 3 (color). Intensity distribution by CWT in the frequency-time domain. Left panels are from the total response $P_0(\tau) + P_1(\tau)$, while right panels are from the response of only the electron-hole channel $P_1(\tau)$. We have (a) $\omega = 3.81$ eV, (b) $\omega = 4.08$ eV, and (c) $\omega = 4.35$ eV.

destructive interference (the dip) at ≈ 10 THz actually reflects the case of the negative asymmetry parameter q in the Fano function $|\mu_{Fg}|^2 \propto (q + \varepsilon)^2/(1 + \varepsilon^2)$ (ε: normalized energy) [1,7], as evident in Fig. 4.

The time-dependent cross sections by MTD numerical calculations (Figs. 2–4) can be scrutinized on the analytic bases. We take the linear susceptibility $\chi(\tau) \propto [\alpha\delta(\tau) + \Theta(\tau)]e^{-\gamma_{\rm ph}\tau}e^{i\omega_{\rm LO}\tau}$, which was introduced for the Fano function in time domain [21]. Here, $\gamma_{\rm ph}$ is the dephasing due to the phonon population relaxation but in the present study, $\gamma_{\rm ph} = 0$. Regarding the coefficient α, it is proportional to the asymmetry parameter q in the steady Fano resonance by $|\mu_{Fg}|^2$ [21]. However, we are dealing with the highly transient Fano resonance, and thus the same argument on α and q will not be applicable in our case. Instead, one may derive α in terms of the perturbation theory for an electron accompanying a virtual optical phonon as the phonon cloud. The electron-phonon wave function $|\psi\rangle$ can then be written down as

$$|\psi\rangle = |\mathbf{k}\rangle_c|0\rangle_{\rm ph} + \sum_{\mathbf{q}} M_{\mathbf{q}} \frac{|\mathbf{k}+\mathbf{q}\rangle_c|\mathbf{q}\rangle_{\rm ph}}{\varepsilon^c_{\mathbf{k}} - \varepsilon^c_{\mathbf{k}+\mathbf{q}} - \omega_{\rm LO}}. \quad (5)$$

The $\mathbf{q} = \mathbf{0}$ optical phonon, the only relevant mode resulting from the (almost) zero momentum transfer in the Raman scattering, reduces $|\psi\rangle$ to

$$|\psi\rangle = |\mathbf{k}\rangle_c|0\rangle_{\rm ph} - \lim_{\bar{q}\to 0} \sum_{|\mathbf{q}|<\bar{q}} M_{\mathbf{q}} \frac{|\mathbf{k}\rangle_c|\mathbf{q}\rangle_{\rm ph}}{\omega_{\rm LO}}. \quad (6)$$

The factor of (-1) occurring between $|\mathbf{k}\rangle_c|0\rangle_{\rm ph}$ and $|\mathbf{k}\rangle_c|\mathbf{q} = 0\rangle_{\rm ph}$ ($|\mathbf{q} = \mathbf{0}\rangle_{\rm ph} \equiv a^\dagger_{\mathbf{q}=\mathbf{0}}|0\rangle_{\rm ph}$) is due to the potential scattering of the electron-hole pair by the phonon final state interaction. This leads to the out-of-phase (π-phase shift) oscillation of $P_1(\tau)$ by Eq. (4), as demonstrated in Fig. 2(a), which indicates $\alpha \approx -1$ [22]. It follows that $\chi(\tau) \propto P_0(\tau) + P_1(\tau)$ with $P_0(\tau) = e^{i\omega_{\rm LO}\tau}$ and $P_1(\tau) = -e^{-\gamma_{cv}\tau}e^{i\omega_{\rm LO}\tau} + \mathcal{P}^*_1(\tau)$. Here, γ_{cv} is the dephasing rate of the electron-hole pair and $\mathcal{P}^*_1(\tau)$ is the transient contin-

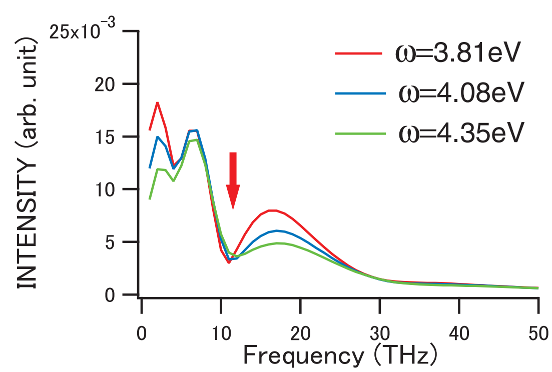

FIG. 4 (color). Slices at $\tau = 0$ of intensity distributions of the total response $P_0(\tau) + P_1(\tau)$ by CWT of Fig. 3. The red arrow designates the dip by the destructive interference.

uum in the broad band. That is, the Dirac delta function $\delta(\tau)$ has been replaced by another short-lived transient governed by the pair dephasing. From $\chi(\tau)$, it is then found that the phonon mode vanishes by the destructive interference at $\tau \approx 0$. But as time goes on, the destructive interference becomes incomplete.

It is important to have deeper insight of the $\mathbf{q} = \mathbf{0}$ mode of the created optical phonon. For an actual observation of the transient destructive Fano interference, we need the macroscopic number of the π-phase shifted $\mathbf{q} = \mathbf{0}$ phonon induced by that number of the electron-hole pair. This requirement can be automatically satisfied by realizing that the created phonon is the coherent phonon. In fact, the $\mathbf{q} = \mathbf{0}$ phonon mode coherently couples with the total number (macroscopic number) of photoexcited electron-hole pairs, as described in Eq. (1). It can then be understood that the final state interaction with the $\mathbf{q} = \mathbf{0}$ coherent phonon makes each interference event pure destructive at $\tau \approx 0$. Incorporating the birth of the $\mathbf{q} = \mathbf{0}$ coherent phonon, now one can note the consilience and the reliability of our argument, relating to the recent experimental results for Si by Hase *et al.* [15]. In the experiment, they have observed the transient reflectivity giving the coherent oscillation of the $\mathbf{q} = \mathbf{0}$ optical phonon and found the antiresonance due to the strong destructive interference between the optical phonon and the continuous electron-hole pair excitation near $\tau = 0$. Through Figs. 2–4, the agreement is found good enough to motivate the common fundamental origin of the destructive Fano interference near $\tau = 0$ even if Si is the indirect gap semiconductor where hole dominates the carrier dynamics [7]. An interesting difference is that the experiment shows the 22 fs delayed appearance of the dip, while the present study predicts an instantaneous appearance, i.e., $\tau \approx 0$. This would be explained by the small phase shift ($\Delta\phi \approx 23°$) of the coherent optical phonon from the cosine function [15] and energy-time uncertainty during the quantum mechanical Fano interference. The destructive Fano interference could now be generalized to the case with the coherent phonon and the continuum quasiparticles, i.e., to the metallic system as well as the (n- or p-type) semiconducting system with the direct or indirect gap [23].

In summary, we have examined the ultrafast dynamics of creation of an optical phonon and the electron-hole pair continuum in a semiconductor. In the transient Fano resonance between channels through the optical phonon and the continuum electronic quasiparticles, the robust destructive Fano interference (antiresonance) at the birth of them ($\tau \approx 0$) is found if the optical phonon is coherent under the irradiation of the femtosecond laser. It is argued that the origin is the potential scattering of the electron-hole pair continuum by the $\mathbf{q} = \mathbf{0}$ coherent phonon.

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