High Frequency Conductivity of Hot Electrons in Carbon Nanotubes

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Abstract

High frequency conductivity of hot electrons in an undoped single walled achiral carbon nanotubes (CNTs) under the influence of ac-dc driven fields is considered. We investigated semiclassically by solving the Boltzmann's transport equation with and without the presence of the hot electrons source to derive the current densities. Plots of the normalized current density versus frequency of ac-field reveal an increase in both the minimum and maximum peaks of normalized current density at lower frequencies as a result of a strong enough injection of hot electrons . The applied ac-field plays twofold role of suppressing the space-charge instability in CNT and simultaneously pumping an energy for lower frequency generation and amplification of THz radiations which have enormous promising applications in very different areas of science and technology.

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Introduction

Carbon nanotubes (CNTs) [1, 2, 3] have been the subject of intense research [4]-[18], since discovery in 1991 by the Japanese scientist Sumio Iijima. Their unique structures, fascinating electronic, magnetic and transport properties have sparked the interest and imagination of researchers worldwide [19]. These quasi-one-dimensional monomolecular nanostructural materials have a wide variety of possible applications [20]-[22]. Research in hot electrons, like any field in semiconductor research, has received a great deal of attention since the arrival of the transistor in 1947 [23]. Recently, it has become possible to fabricate semiconductor devices with submicron dimensions. The miniaturization of devices has led to high field well outside the linear response region, where Ohm's law holds for any reasonable voltage signal [24]. The physical understanding of the microscopic processes which underlie the operations of such devices at high electric fields is provided by research into hot electron phenomena [25]. Whereas, there are several reports on hot electrons generation in CNTs [26, 27, 28, 29], the reports on high frequency conductivity of hot electrons in CNTs are limited. Thus, in this paper, we present a theoretical framework investigations of high frequency conductivity of hot electrons in $(3, 0)$ zigzag (zz) CNT and $(3, 3)$ armchair (ac) The Boltzmann transport equation is solved in the framework of momentum-independent relaxation time using the semiclassical approach to obtain current density for each achiral CNTs. We probe the behavior of the electric current density of the CNTs as a function of the frequency of ac field with and without the axial injection of the hot electrons.

Theory

If a dc field E_z is applied along a $z-$ axis of an undoped single-wall carbon nanotube, electrons begin to move in accordance with the semiclassical Newtons law (neglecting scattering) [30]

$$\frac{dP_z}{dt} = eE_z \tag{1}$$

where P_z and e are the axial component of the quasimomentum and the electronic charge of the propagating electrons respectively. For a CNT, If energy level spacing $\Delta\varepsilon$ ($\Delta\varepsilon = \pi\hbar V_F/L$, $\hbar = h/2\pi$, h is Planck constant, V_F is Fermi velocity and L is the length of the nanotube) is large enough and the scattering rate v is small such that $\Delta\varepsilon >> aeE_z$ and $hv < aeE_z$ $(zz-CNT)$, and $\Delta\varepsilon >> \frac{a}{\sqrt{3}}aE_z$ $hv < \frac{a}{\sqrt{3}}aE_z$, $(ac - CNT)$, then the electrons oscillate inside the lower level with so-called Bloch frequency Ω given by [31]:

$$\Omega_{zz} = \frac{aeE_z}{\hbar} \tag{2}$$

$$\Omega_{ac} = \frac{aeE_z}{\sqrt{3}\hbar} \tag{3}$$

for $zz-$CNT and $ac-$CNT respectively. Here, a is the lattice constant of the CNT. The investigation is done within the semiclassical approximation in which the motion of the π-electrons are considered as classical motion of free quasiparticles in the field of the crystalline lattice with dispersion law extracted from the quantum theory. Taking into account the hexagonal crystalline structure of a rolled graphene in a form of CNTs and using the tight binding approximation, the energies for $zz-$CNT and $ac-$CNT are expressed as in equations (4) and (5), respectively [31]

$$\begin{aligned}\varepsilon(s\Delta p_\phi, p_z) &\equiv \varepsilon_s(p_z) = \\ &\pm \gamma_0\sqrt{1+4cos(ap_z)cos(\frac{a}{\sqrt{3}}s\Delta p_\phi)+4cos^2(\frac{a}{\sqrt{3}}s\Delta p_\phi)}\end{aligned} \quad (4)$$

$$\begin{aligned}\varepsilon(s\Delta p_\phi, p_z) &\equiv \varepsilon_s(p_z) = \\ &\pm \gamma_0\sqrt{1+4cos(as\Delta p_\phi)cos(\frac{a}{\sqrt{3}}p_z)+4cos^2(\frac{a}{\sqrt{3}}p_z)}\end{aligned} \quad (5)$$

where $\gamma_0 \approx 3.0eV$ is the overlapping integral, p_z is the axial component of quasimomemtum. Δp_ϕ is transverse quasimomentum level spacing and s is an integer. The expression for lattice constant a in equations (4) and (5) is given by

$$a = \frac{3a_{c-c}}{2\hbar} \quad (6)$$

where $a_{(c-c)} = 0.142nm$ is the C-C bond length. The $-$ and $+$ signs correspond to the valence and conduction bands respectively. Due to the transverse quantization of the quasimomentum P, its transverse component p_ϕ can take n discrete values,

$$p_\phi = s\Delta p_\phi = \frac{\pi\sqrt{3}s}{an} \qquad (s = 1, ..., n) \quad (7)$$

Unlike transverse quasimomentum, p_ϕ, the axial quasimomentum ϕ_z is assumed to vary continuously within the range $0 \leq p_z \leq 2\pi/a$, which corresponds to the model of infinitely long CNT ($L = \infty$). This model is applicable to the case under consideration because we are restricted to temperatures and/or voltages well above the level spacing [32], i.e. $k_\beta T > \varepsilon_c, \Delta\varepsilon$, where k_β

is Boltzmann constant, T is the temperature, ε_c is the charging energy. The energy expression in eq (4) and (5) can be expressed in the Fourier series as

$$\varepsilon(p_z, s\Delta p_\phi) = \varepsilon(p_z) = \gamma_0 \sum_{r\neq 0} exp(iarp_z) \tag{8}$$

where ε_{rs} is given as

$$\varepsilon_{r,s} = \frac{a}{2\pi\gamma_0}\int_0^{\frac{2\pi}{a}} \varepsilon_s(p_z)exp(-irap_z)dp_z \tag{9}$$

the quasiclassical velocity of an electron moving along the CNTs axis is given by the expression $v_z(p_z, s\Delta p_\phi) = \partial\varepsilon_{rs}(p_z)/\partial p_z$. Substituting eqn (9) and expressing further gives

$$v_z(p_z, s\Delta p_z) = \gamma_0 \sum_{r\neq 0} \frac{\partial(\varepsilon_{rs}exp(iarp_z)}{\partial p_z} = \gamma_0 \sum_{r\neq 0} iar\varepsilon_{rs}exp(iarp_z) \tag{10}$$

Considering the presence of hot electrons source, the motion of quasiparticles in an external axial electric field is described by the Boltzmann kinetic equation in the form as shown below [30, 31]

$$\frac{\partial f(p)}{\partial t} + v_z\frac{\partial f(p)}{\partial x} + eE(t)\frac{\partial f(p)}{\partial p_z} = -\frac{f(p) - f_0(p)}{\tau} + S(p) \tag{11}$$

where $S(p)$ is the hot electron source function, $f_0(p)$ is equilibrium Fermi distribution function, $f(p,t)$ is the distribution function, v_z is the quasiparticle group velocity along the $z-$axis of carbon nanotube and τ is the relaxation time. The relaxation term of equation (11) describes the electron-phonon scattering, electron-electron collisions [31, 32] etc. Using the method originally developed in the theory of quantum semiconductor superlattices [31], an exact solution of equation (11) can be constructed without assuming a weak electric field. Expanding the distribution functions of interest in Fourier series as

$$f(p,t) = \Delta p_\phi \sum_{s=1}^{n} \delta(p_\phi - s\Delta p_\phi) \sum_{\tau \neq 0} f_{rs} exp(iarp_z)\psi_v(t) \tag{12}$$

$$f_0(p) = \Delta p_\phi \sum_{s=1}^{n} \delta(p_\phi - s\Delta p_\phi) \sum_{\tau \neq 0} f_{rs} exp(iarp_z) \tag{13}$$

for zz-CNTs

$$f(p,t) = \Delta p_\phi \sum_{s=1}^{n} \delta(p_\phi - s\Delta p_\phi) \sum_{\tau \neq 0} f_{rs} exp(ibrp_z)\psi_v(t) \tag{14}$$

$$f_0(p) = \Delta p_\phi \sum_{s=1}^{n} \delta(p_\phi - s\Delta p_\phi) \sum_{\tau \neq 0} f_{rs} exp(ibrp_z) \tag{15}$$

for ac-CNTs where $b = a/\sqrt{3}$ or $a = b/\sqrt{3}$, $\delta(p_\phi - s\Delta p_\phi)$ is the Dirac-delta function, f_{rs} is the coefficients of the Fourier series and $\psi_v(t)$ is the factor by which the Fourier transform of the nonequilibruim distribution function differs from its equilibrium distribution counterpart. The expression for f_{rs} can be expanded in the analogous form as

$$f_{rs} = \frac{a}{2\pi} \int_0^{\frac{2\pi}{a}} \frac{exp(-iarp_z)}{1 + exp(\varepsilon_s(p_z))/k_\beta T)} dp_z \tag{16}$$

The electron surface current density j_z along the CNTs axis is also given by the expression

$$j_z = \frac{2e}{(2\pi\hbar)^2} \int\int f(p,t) v_z(p) d^2p \tag{17}$$

the integration is carried over the fist Brillouin zone. For simplicity, we consider a hot electron source of the simplest form given by the expression,

$$S(p) = \frac{Qa}{\hbar}\delta(\phi - \phi') - \frac{aQ}{n_0} f_s(\phi) \tag{18}$$

where $f_s(p)$ is the stationary (static and homogeneous) solution of equation (19), Q is the injection rate of hot electron , n_0 is the equilibrium particle density, ϕ and ϕ' are the dimensionless momenta of electrons and hot electrons respectively which are expressed as $\phi_{zz} = ap_z/\hbar$ and $\phi'_{zz} = ap'_z/\hbar$ for zz-CNTs and $\phi_{ac} = ap_z/\sqrt{3}\hbar$ and $\phi'_{ac} = ap'_z/\sqrt{3}\hbar$ for ac-CNTs, We now find the high frequency conductivity of hot electrons in the nonequilibrium state for zz-CNT by considering perturbations with frequency ω and wave-vector k of the form

$$E(t) = E_z + E_{\omega,k} exp(-i\omega t + ikx) \tag{19}$$

$$f = f_s(\phi) + f_{\omega,k} exp(-i\omega t + ikx) \tag{20}$$

Substituting equations (19) and (20) into equation (11) and rearranging yields,

$$\frac{\partial f_{\omega,k}}{\partial \phi} + i[\alpha + kv_z] f_{\omega,k} = -\frac{E_{\omega,k}}{E_z} \frac{\partial f_s(\phi)}{\partial \phi} \tag{21}$$

where $\alpha = -(\omega + iv_z)\,\Omega_{zz}$. Solving the homogeneous differential equation (21) and then introducing the Jacobi-Anger expansion and averaging the current over time, we obtain the current density for the zz-CNTs in the presence of hot electrons (j^{zz}_{zHE}) as

$$j^{zz}_{zHE} = i\frac{4\sqrt{3}e^2\gamma_0}{n\hbar^2} \sum_{l=1} r \sum_{s=1} f_{rs}\varepsilon_{rs} \sum_{m,l=-\infty} \frac{i^l m l j_m(\beta) j_{m-l}(\beta) I_{m-l}(\beta) \Omega_{zz}}{\omega + iv - m\Omega_{zz}} \times$$
$$\eta_{zz}\frac{n_0}{2\pi} \sum_r \frac{\Omega_{zz} exp(ir\phi)}{(ir\Omega_{zz} + v + \eta\Omega_{zz})} (exp(-ir\phi' - \frac{v}{(v + ir\Omega_{zz})}) + \frac{v}{(v + ir\Omega_{zz})}) \tag{22}$$

where β is the normalized amplitude of the ac-field, $j_m(\beta)$ is the bessel function order m and $I_m(\beta)$ is the modified bessel function order m. In the absence of hot electrons, the nonequalibrium parameter for zz-CNT $\eta_{zz} = 0$, hence the current density for zz-CNTs without hot electron source $j_(z)^{zz}$ could be obtained from equation (22) by setting $\eta_{zz} = 0$. Therefore, the current density of zz-CNTs in the absence of hot electrons j_z^{zz} is given by

$$j_z^{zz} = i\frac{4\sqrt{3}e^2\gamma_0}{n\hbar^2}\sum_{l=1} r \sum_{s=1} f_{rs}\varepsilon_{rs} \sum_{m,l=-\infty} \frac{i^l m l j_m(\beta) j_{m-l}(\beta) I_{m-l}(\beta)\Omega_{zz}}{\omega + iv - m\Omega_{zz}} \quad (23)$$

Using similar argument like one for zz-CNT, the current density for an ac-CNT with and without the injection of hot electrons are expressed respectively as:

$$j_{zHE}^{ac} = i\frac{4e^2\gamma_0}{\sqrt{3}n\hbar^2}\sum_{l=1} r \sum_{s=1} f_{rs}\varepsilon_{rs} \sum_{m,l=-\infty} \frac{i^l m l j_m(\beta) j_{m-l}(\beta) I_{m-l}(\beta)\Omega_{ac}}{\omega + iv - m\Omega_{ac}} \times$$
$$\eta_{ac}\frac{n_0}{2\pi}\sum_r \frac{\Omega_{ac} exp(ir\phi)}{(ir\Omega_{zz} + v + \eta\Omega_{ac})}(exp(-ir\phi' - \frac{v}{(v + ir\Omega_{ac})}) + \frac{v}{(v + ir\Omega_{ac})}) \quad (24)$$

$$j_z^{ac} = i\frac{4e^2\gamma_0}{\sqrt{3}n\hbar^2}\sum_{l=1} r \sum_{s=1} f_{rs}\varepsilon_{rs} \sum_{m,l=-\infty} \frac{i^l m l j_m(\beta) j_{m-l}(\beta) I_{m-l}(\beta)\Omega_{zz}}{\omega + iv - m\Omega_{zz}} \quad (25)$$

Results and discussion

We now present a semiclassical theory of electron transport in a CNT under conditions where, in addition to the dc field causing a Negative Differential Conductivity (NDC), a similarly strong ac field is present,. Here the ac field plays a two fold role: It suppresses the space-charge instability in CNT and simultaneously pumps an energy for generation and amplification of THz radiation at higher frequency [33]. Figure 1 displays the behaviour of the normalized current density ($J_z = \frac{J_{zHE}^{zz}}{j_0}$, where $j_0 = \frac{4e^2\gamma_0}{\sqrt{3}n\hbar^2}$) as a function of the frequency (ω) of ac field for the CNTs stimulated axially with the hot electrons, represented by the nonequilibrium parameter η. In the absence

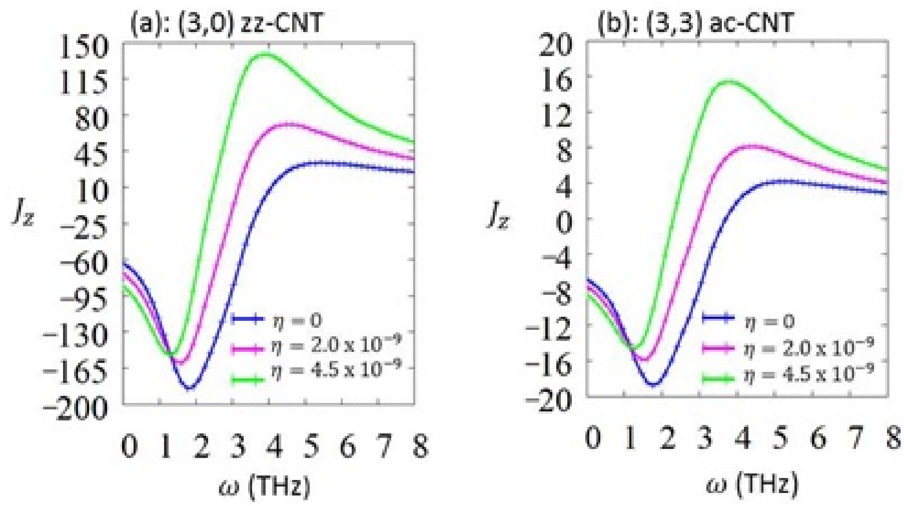

Figure 1: A plot of normalized current density (J_z) versus frequency of ac field (ω) as the nonequilibrium parameter (η) increases from 0 to 4.5×10^{-9} for (a) $(3,0)$ zz-CNT and (b) $(3,3)$ ac-CNT , $T = 287.5K$ and $v = 1THz$

of hot electrons ($\eta = 0$), we observed that the differential conductivity is initially negative at zero frequency. With increasing frequency of ac electric field ω from zero, the differential conductivity becomes more negative until a minimum peak is reached at a frequency ω about $1.8THz$ for both zz-CNT and ac-CNT . Then after the differential conductivity turns positive when $\omega > 1.8THz$ until the maximum peak is attained at $\omega \approx 4.5THz$ and then decrease when $\omega > 4.5THz$ for both zz-CNT and ac-CNT. The Positive Differential Conductivity (PDC) is considered as one of the conditions for electric stability of the system [34] and indicative for terahertz gain without the small spike or fluctuations of electrons associated with NDC that amplifies, induces space charge accumulation and finally develops into electric field domain [35]. The electrical domain development and transporting induce unstable non uniform electric field distribution, which in turn prevents the operation of the Bloch oscillations. Thus suppressing domain formation is a prerequisite to observe Bloch oscillations necessary for terahertz gain [35]. As we increase the nonequilibrium parameter η which increases as the rate of hot electrons injection increases from 0 (no hot electrons) to 4.5×10^{-9} (presence of hot electrons), we observed that the minimum peak decreases and shifts to the left (i.e., low frequency). In the contrary, the maximum peak increases and also shifts to the left (i.e., low frequency) as shown in figure 1. In figure 2, we display the behaviour of normalized current density (J_z) as a function of frequency of ac field (ω) as the nonequilibrium parameter (η) is further increased to 23.0×10^{-9}. As we further increase the nonequilibrium

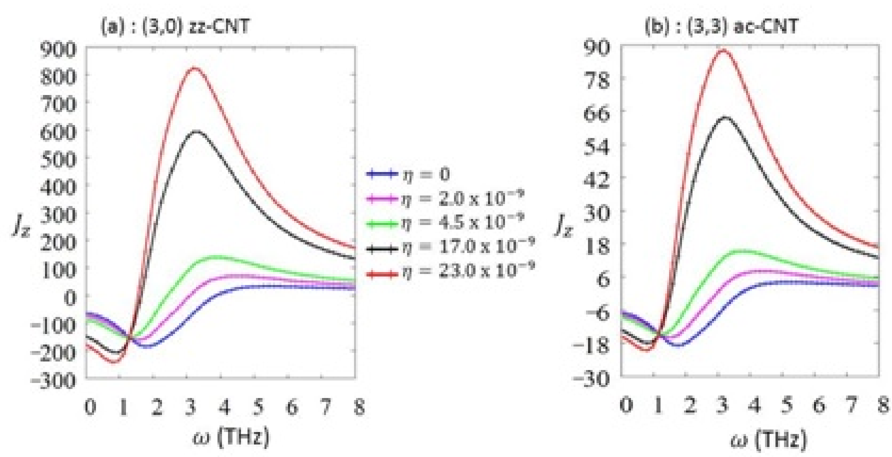

Figure 2: A plot of normalized current density (J_z) versus frequency of ac field (ω) as the nonequilibrium parameter (η) increases from to 23×10^{-9} for (a) $(3,0)$ zz-CNT and (b) $(3,3)$ ac-CNT , $T = 287.5K$ and $v = 1THz$

parameter η to 23.0×10^{-9} (i.e strong enough injection rate), we now noticed that both the minimum and maximum peaks increase and shift to the left (i.e., low frequency) for $\eta \geq 17.0 \times 10^{-9}$. Hence high frequency conductivity of strong enough hot electrons in CNTs leads to increase in both the minimum and maximum peaks of normalized current density at lower frequencies as shown in figure 2. To put the above observations in perspective, we display in figures 3 and 4, a 3-dimensional behavior of the normalized current density (J_z) as a function of the frequency of ac field (ω) and nonequilibrium parameter (η) In figure 3, we observed that when nonequilibrium parameter

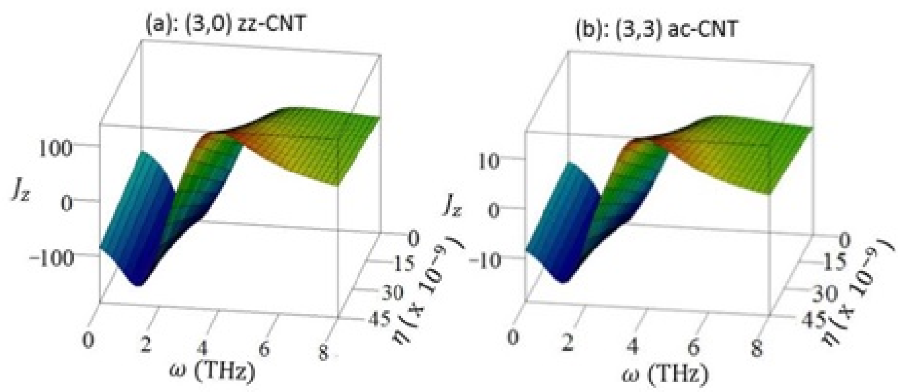

Figure 3: A $3D$ plot of normalized current density (J_z) versus frequency of ac field (ω) as the nonequilibrium parameter (η) increases from 0 to 4.5×10^{-9} for (a) $(3,0)$ zz-CNT and (b) $(3,3)$ ac-CNT , $T = 287.5K$ and $v = 1THz$

η is zero, the minimum peak is the greatest at a relative high frequency while the maximum peak is the least also at high frequency. As nonequilibrium parameter η increases from 0 to 4.5×10^{-9}, the minimum peak gradually decreases and shifts towards left (i.e. low frequency) while the maximum peak slowly increases and also shift towards left (i.e. low frequency) In figure 4, as nonequilibrium parameter η further increases from 0 to 23.0×10^{-9}, the minimum peak initially decreases and shifts towards left (i.e. low frequency) and then finally increases and shifts towards left (i.e low frequency) until the

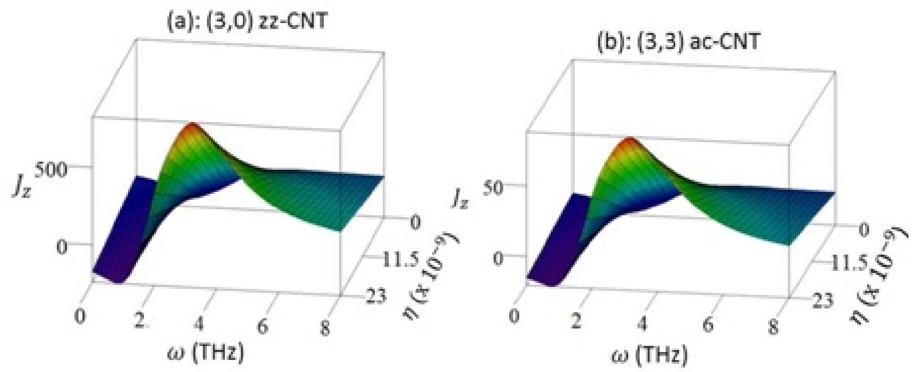

Figure 4: A $3D$ plot of normalized current density (J_z) versus frequency of ac field (ω) as the nonequilibrium parameter (η) increases to 23×10^{-9} for (a) $(3, 0)$ zz-CNT and (b) $(3, 3)$ ac-CNT , $T = 287.5K$ and $v = 1THz$

highest minimum peak is attained at the lowest frequency. The trend of the maximum peak as nonequilibrium parameter further increases to 23.0×10^{-9} remain unchanged.

Conclusion

In summary, we have shown theoretically a high frequency conductivity of hot electrons in a CNT under conditions where, in addition to the dc field causing NDC, a similarly strong ac field is applied . The applied ac-field plays twofold role of suppressing the space-charge instability in CNT and simultaneously pumping an energy for generation and amplification of THz radiation which have enormous promising applications in very different areas of science and technology. The generation of this radiation occurs at lower frequency . This is mainly because of increase in both the minimum and maximum peaks of normalized current density at lower frequencies as a result of the presence of hot electrons .

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