Hot electrons injection in carbon nanotubes under the influence of quasi-static ac-field

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Abstract

Hot electrons injection in carbon nanotubes (CNTs) where in addition to applied dc field ($\mathbf{E}$), there exist simultaneously a quasi-static ac electric field (i.e. when the frequency ω of ac field is much less than the scattering frequency v ($\omega \ll v$ or $\omega\tau \ll 1$, $v = \tau^{-1}$, where τ is relaxation time) is considered. The investigation is done theoritically by solving semiclassical Boltzmann transport equation with and without the presence of the hot electrons source to derive the current densities. Plots of the normalized current density versus dc field ($\mathbf{E}$) applied along the axis of the CNTs in the presence and absence of hot electrons reveal ohmic conductivity initially and finally negative differential conductivity (NDC) provided $\omega\tau \ll 1$ (i.e. quasi- static case). With strong enough axial injection of the hot electrons , there is a switch from NDC to positive differential conductivity (PDC) about $\mathbf{E} \geq 75kV/cm$ and $\mathbf{E} \geq 140kV/cm$ for a zigzag CNT and an armchair

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CNT respectively. Thus, the most important tough problem for NDC region which is the space charge instabilities can be suppressed due to the switch from the NDC behaviour to the PDC behaviour predicting a potential generation of terahertz radiations whose applications are relevance in current-day technology, industry, and research.

Introduction

Carbon nanotubes (CNTs) [1] - [3] are subject of many theoretical [4] - [9], and experimental [10] - [18] studies. Their properties include a thermal conductivity higher than diamond, greater mechanical strength than steel and better electrical conductivity than copper [19] - [21]. These novel properties make them potentially useful in a variety of applications in nanotechnology, optics, electronics, and other fields of materials science [22] [24]. Rapid development of submicrometer semiconductor devices, which may be employed in high-speed computers and telecommunication systems, enhances the importance of hot-electron phenomena [25]. Hot electron phenomena have become important for the understanding of all modern semiconductor devices [26]- [27]. There are several reports on hot electrons generation in CNTs [28]- [30], but the reports on hot electrons injection in CNTs under the influence of quasi-static ac field to the best of our knowlege are limited. Thus, in this paper, we analyzed theoretically hot electrons injection in $(3,0)$ zigzag(zz) CNT and $(3,3)$ armchair (ac) CNT where in addition to dc field, a quasi-static ac electric field is applied. Adopting semiclassical approach , we obtained current density for each achiral CNTs after solving the Boltzmann transport equation in the framework of momentum-independent relaxation

time . We probe the behaviour of the electric current density of the CNTs as a function of the applied *dc* field **E** of $ac - dc$ driven fields when the frequency of ac field (ω) is much less than the scattering frequency (v) $(\omega \ll v$ or $\omega\tau \ll 1$ i.e quasi-static case [31], where $v = \tau^{-1})$ with and without the axial injection of the hot electrons.

Theory

Suppose an undoped single walled achiral carbon nanotubes (CNTs) $(n, 0)$ or (n, n) of length L is exposed to a homogeneous axial dc field **E** given by $\mathbf{E} = V/L$, where V is the voltage between the CNT ends. Under the influence of the applied dc field and assuming scattering is negligible, electrons with electronic charge (e) obey Newton's law of motion given by [32]

$$\frac{d\mathbf{P}}{dt} = e\mathbf{E} \tag{1}$$

where **P** is a component of quasimomentum along the axis of the tube. Adopting semiclassical approximation approach and considering the motion of $\pi-$ electrons as a classical motion of free quasi-particles with dispersion law extracted from the quantum theory while taking into account to the hexagonal crystalline structure of CNTs and applying the tight-binding approximation gives the energies for $zz-$CNT and ac-CNT respectively

$$
\begin{aligned}
\varepsilon(s\Delta p_\vartheta, p) \quad &\equiv \quad \varepsilon_s(p) = \\
&\pm \quad \gamma_0 \left[1 + 4\cos\left(ap\right)\cos\left(\frac{a}{\sqrt{3}}s\Delta p_\vartheta\right) + 4\cos^2\left(\frac{a}{\sqrt{3}}s\Delta p_\vartheta\right) \right]^{1/2}
\end{aligned} \tag{2}
$$

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$$\varepsilon(s\Delta p_\vartheta, p) \equiv \varepsilon_s(p_z) =$$

$$\pm \ \gamma_0 \left[1 + 4\cos(as\Delta p\}) \cos\left(\frac{a}{\sqrt{3}}p_\vartheta\right) + 4\cos^2\left(\frac{a}{\sqrt{3}}p_\vartheta\right)\right]^{1/2} \tag{3}$$

where $\gamma_0 \approx 3.0eV$ is the overlapping integral, Δp_ϑ is transverse quasimomentum level spacing and s is an integer. The lattice constant a in Eqn.(2) and (3) is expressed as [33]

$$a = \frac{3b}{2\hbar} \tag{4}$$

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

$$p_\vartheta = s\Delta p_\vartheta = \frac{\pi\sqrt{3}s}{an}(s = 1, ..., n) \tag{5}$$

As different from p_ϑ, we assume $\mathbf{p}$ continuously varying within the range $0 \leq \mathbf{p} \leq 2\pi/a$ which corresponds to the model of infinitely long CNT $(L = \infty)$. The model is applicable to the case under consideration because we are restricted to temperatures and/or voltages well above the level spacing [33], i.e. $k_BT > \varepsilon_c$, $\Delta\varepsilon$, where k_B is Boltzmann constant, T is the thermodyanamic temperature, ε_c is the charging energy. In the presence of hot electrons source, the motion of quasi-particles in an external axial electric field is described by the Boltzmann kinetic equation as [32]- [33]

$$\frac{\partial f(p)}{\partial t} + v\frac{\partial f(p)}{\partial x} + eE(t)\frac{\partial f(p)}{\partial p} = -\frac{f(p) - f_{eq}(p)}{\tau} + S(p) \tag{6}$$

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where $f_{eq}(p)$ is equilibrium Fermi distribution function, $f(p,t)$ is the distribution function, $S(p)$ is the hot electron source function, $\mathbf{v}$ is the quasiparticle group velocity along the axis of carbon nanotube and τ is the relaxation time. The relaxation term of Eqn.(6) above describes the electron-phonon scattering, electron-electron collisions [34] [35] etc.

Applying the method originally developed in the theory of quantum semiconductor superlattices [33], an exact solution of equation (6) can be constructed without assuming a weak electric field. By expanding the distribution functions of interest in Fourier series, we have:

$$f(p,t) = \Delta p_\vartheta \sum_{s=1}^{n} \delta(p_\vartheta - s\Delta p_\vartheta) \sum_{r \neq 0} f_{rs} exp(iarp)\psi_v(t) \qquad (7)$$

and

$$f_{eq}(p) = \Delta p_\vartheta \sum_{s=1}^{n} \delta(p_\vartheta - s\Delta p_\vartheta) \sum_{r \neq 0} f_{rs} exp(iarp) \qquad (8)$$

for zz-CNT and

$$f(p,t) = \Delta p_\vartheta \sum_{s=1}^{n} \delta(p_\vartheta - s\Delta p_\vartheta) \sum_{r \neq 0} f_{rs} \exp ir(a/\sqrt{3}p)\psi_v(t) \qquad (9)$$

and

$$f_{eq}(p) = \Delta p_\vartheta \sum_{s=1}^{n} \delta(p_\vartheta - s\Delta p_\vartheta) \sum_{r \neq 0} f_{rs} \exp\{ir(a/\sqrt{3}p)\} \qquad (10)$$

for ac-CNTs

where $\delta(p_\vartheta - s\Delta p_\vartheta)$ 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 nonequilibrium distribution function differs from its equilibrium distribution

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counterpart. For simplicity, we consider a hot electron source of the simplest form given by the expression,

$$S(p) = \frac{Qa}{\hbar}\delta(\varphi - \varphi') - \frac{aQ}{n_0}f_s(\varphi) \tag{11}$$

where $f_s(p)$ is the static and homogeneous (stationary) solution of Eqn.(6), 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 $\varphi = a\mathbf{p}/\hbar$ and $\varphi' = a\mathbf{p}'/\hbar$ for zz-CNTs and $\varphi = a\mathbf{p}/\sqrt{3}\hbar$ and $\varphi' = a\mathbf{p}'/\sqrt{3}\hbar$ for ac-CNTs,

We now obtain the current density in the nonequilibrium state for zz-CNT where in addition to applied dc field, there exist simultaneously a quasi-static ac electric field by considering perturbations with frequency ω and wave-vector κ of the form [32].

$$E(t) = \mathbf{E} + E_{\omega,k}exp(-i\omega t + ikx) \tag{12}$$

$$f = f_s(\varphi) + f_{\omega,k}exp(-i\omega t + ikx) \tag{13}$$

where $\mathbf{E}$ is dc field along the axis of the tube, $E_{\omega,\kappa}e^{-i\omega t + i\kappa x}$ is ac-field, $E_{\omega,\kappa}$ is peak ac field and $f_s(\varphi)$ is the static and homogeneous (stationary) solution of Eqn.(6). Substituting Eqn.(12) and (13) into Eqn.(6) and rearranging yields,

$$\frac{\partial f_{\omega,k}}{\partial\varphi} + i[\alpha + k\mathbf{v}_z\hbar/ae\mathbf{E}]f_{\omega,k} = -\frac{E_{\omega,\kappa}}{\mathbf{E}}\frac{\partial f_s(\varphi)}{\partial\varphi} \tag{14}$$

where $\alpha = -\hbar(\omega + iv)/aeE$ and $f_{\omega,k}$ is the solution of Eqn.(14). Solving the homogeneous differential Eqn.(14)and and then introducing the Jacobi-Anger expansion, we obtain the normalized current density in the presence

of hot electrons (j_{HE}^{zz}) as

$$j_{HE}^{zz} = i\frac{4\sqrt{3}e^2\gamma_0}{n\hbar^2}\sum_{r=1}r\sum_{s=1}f_{rs}\varepsilon_{rs}\sum_{m,l=-\infty}\frac{i^l m l j_m(\beta)j_{m-l}(\beta)I_{m-l}(\beta)aeE}{([\omega+iv]\hbar - m(ae)\mathbf{E})}$$

$$\times\left\{\eta\frac{n_o}{2\pi}\sum_r\frac{ae\mathbf{E}exp(ir\varphi)}{(ir(ae\mathbf{E})+v\hbar+\eta(ae\mathbf{E}))}\left(exp(-ir\varphi') - \frac{v\hbar}{(v\hbar+ir(ae\mathbf{E}))}\right)\right.$$

$$\left. + \frac{v\hbar}{(v\hbar+ir(ae\mathbf{E}))}\right\} \quad (15)$$

where

$$f_{rs} = \frac{a}{2\pi\Delta p_\vartheta}\int_0^{2\pi/a}\frac{\exp(-iarp)}{1+\exp\{\varepsilon_s(p)/k_BT\}}dp$$

$$\varepsilon_{rs} = \frac{a}{2\pi\gamma_0}\int_0^{2\pi/a}\varepsilon_s(\mathbf{p})\exp(-iarp)dp$$

$\beta = \kappa\gamma_0 a/\Omega\hbar$, $\eta = Q/\Omega n_0$ and $\Omega = ea\mathbf{E}/\hbar$, $j_m(\beta)$ is the mth order Bessel function of the first kind, $J_{(m-1)}(\beta)$ is the $(m-1)$th order Bessel function of the first kind, $I_{(m-l)}(\beta)$ is $(m-1)th$ order modified Bessel function of the first kind, Q is rate of hot electrons injection, n_0 is the particle density Ω is the Bloch frequency and η is the non-equilibrium parameter.

In the absence of hot electrons, the nonequalibrium parameter for zz-CNT, $\eta = 0$, hence the current density for zz-CNTs without hot electron source j^{zz} could be obtained from Eqn.(15) by setting $\eta = 0$. Therefore, the electric current density of zz-CNTs in the absence of hot j^{zz} is given by

$$j_z^{zz} = i\frac{4\sqrt{3}e^2\gamma_0}{n\hbar^2}\sum_{l=1}r\sum_{s=1}f_{rs}\varepsilon_{rs}$$

$$\times\sum_{m,l=-\infty}\frac{i^l m l j_m(\beta)j_{m-l}(\beta)I_{m-l}(\beta)(ae)\mathbf{E}}{([\omega+iv]\hbar - m(ae)\mathbf{E})}\left\{\sum_r\frac{v\hbar}{v\hbar+ir(ae\mathbf{E})}\right\} \quad (16)$$

Applying 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:

$$j_{HE}^{ac} = i\frac{4e^2\gamma_0}{\sqrt{3}n\hbar^2}\sum_{r=1}\sum_{s=1}f_{rs}\varepsilon_{rs}\sum_{m,l=-\infty}\frac{i^l ml j_m(\beta)j_{m-l}(\beta)I_{m-l}(\beta)(ae)\mathbf{E}}{(\sqrt{3}[\omega+iv]\hbar - m(ae)\mathbf{E})}$$

$$\times\left\{\eta\frac{n_o}{2\pi}\sum_r\frac{(ae)\mathbf{E}exp(ir\varphi)}{(ir(ae\mathbf{E})+\sqrt{3}v\hbar+\eta(ae\mathbf{E}))}\left(exp(-ir\vartheta') - \frac{\sqrt{3}v\hbar}{(\sqrt{3}v\hbar + ir(ae\mathbf{E}))}\right)\right.$$

$$\left. + \frac{\sqrt{3}v\hbar}{(\sqrt{3}v\hbar + ir(ae\mathbf{E}))}\right\}\,(17)$$

and

$$j^{ac} = i\frac{4e^2\gamma_0}{\sqrt{3}n\hbar^2}\sum_{r=1}r\sum_{s=1}f_{rs}\varepsilon_{rs}\sum_{m,l=-\infty}\frac{i^l ml j_m(\beta)j_{m-l}(\beta)I_{m-l}(\beta)(ae)\mathbf{E}}{(\sqrt{3}[\omega+iv]\hbar - m(ae)\mathbf{E})}$$

$$\times\sum_r\frac{\sqrt{3}v\hbar}{\sqrt{3}v\hbar + ir(ae\mathbf{E})}\quad(18)$$

where

$$f_{rs} = \frac{a}{2\pi\Delta p_\vartheta}\int_0^{2\pi/a}\frac{\exp(-iarp/\sqrt{3})}{1+\exp\{\varepsilon_s(p)/k_BT\}}dp$$

$$\varepsilon_{rs} = \frac{a}{2\pi\gamma_0}\int_0^{2\pi/a}\varepsilon_s(\mathbf{p})\exp(iarp/\sqrt{3})dp$$

$\beta = \kappa\gamma_0 a/\Omega\sqrt{3}\hbar$, $\eta = Q/\Omega n_0$ and $\Omega = ea\mathbf{E}/\sqrt{3}\hbar$

Results and discussion

We display the behaviour of the normalized current density ($J = j/jos$) and $jos = (4\sqrt{3}e^2\gamma_0)/n\hbar$ (zz-CNT) or $4e^2\gamma_0/\sqrt{3}n\hbar^2(ac-CNT)$ as a function of the applied dc field $\mathbf{E}$ when frequency of ac field ω is much less than

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scattering frequency $v(\omega \ll v$ or $\omega\tau \ll 1$ i.e. quasi-static case) for the CNTs stimulated axially with the hot electrons, represented by the nonequilibrium parameter η in figure 1. As we increase the nonequilibrium parameter η
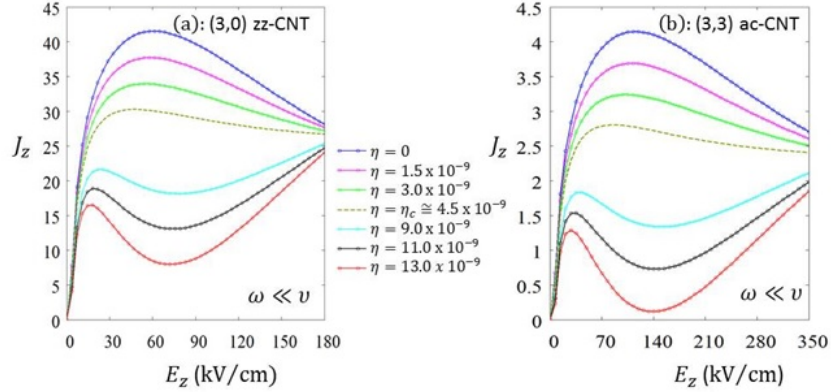

Figure 1: A plot of normalized current density (J_z) versus applied dc field (E_z) as the onequilibrium parameter η increases from 0 to 13.0×10^{-9} when $\omega << v$ or $\omega\tau << 1$ (i.e.quasi-static case), for (a) $(3,0)$ zz-CNT and (b) $(3,3)$ ac-CNT, $T = 287.5K$, $\omega = 10^{-4}THz$, $v = 1THz$ or $\tau = 1ps$ and $\omega\tau = 10^{-4}$

from 0 to 13.0×10^{-9}, we noticed that the normalized current density has the highest peak for η (no hot electrons). As the hot electrons injection rate increases, the peak of the current density decreases and shifts to the left (i.e., low dc fields). This is caused by the scattering effects due to electron-phonon interactions as well as the increase in the direct hot electrons injection rate [36] [37]. The normalized current density (J) of the CNTs exhibits a linear monotonic dependence on the applied dc field $(\mathbf{E})$ at weak field (i.e.,the region of ohmic conductivity) when frequency of ac field ω is much less than scattering frequency v ($\omega \ll v$ or $\omega\tau \ll 1$ i.e. quasi-static case, where $v = \tau^{-1}$). As the applied dc field $(\mathbf{E})$ increases, the normalized current density (J) increases and reaches a maximum, and drops off, experiencing

a negative differential conductivity (NDC) for both the *zz*- CNT and the *ac*-CNT as shown in figures 1*a* and 1*b*, respectively. The NDC is due to the increase in the collision rate of the energetic electrons with the lattice that induces large amplitude of oscillation in the lattice, which in-turn increases the electrons scattering rate that leads to the decrease in the current density at high dc field [37]. Similar effect was observed by Mensah, et. al. [40] in su-perlattice. As the injection rate of the hot electrons becomes strong enough , the current density up-turned, exhibiting a positive differential conductiv-ity (PDC) near $75kV/cm$ and $140kV/cm$ for the *zz*-CNT and the *ac*-CNT, respectively. In this region, the hot electrons become the dominant deter-mining factor [36]. The physical mechanism behind the switch from NDC to PDC is due to the interplay between the hot electrons pumping frequency (Q/n_0), which is a function of rate of hot electrons injection (Q), and the Bloch frequency (Ω), which depends on the *dc* field (**E**) [37]. At stronger *dc* field, the rate of scattering of the electrons by phonons is well pronounced re-sulting in the gradual decrease in the current density with increasing dc field (NDC region). However, as the rate of hot electrons injection increases, the corresponding rise in the current density due to hot electrons injection now far exceeds the reduction in the current density due to scattering of electrons by phonons. Thus, the net effect on the current density from the two oppos-ing sources (with the hot electrons being dominant) gives rise to the PDC characteristics as shown in figure 1 for $\eta \geq 9.0 \times 10^{-9}$. The desirable effect of a switch from NDC to PDC takes place when η is larger than a critical value $\eta_c \approx 4.5 \times 10^{-9}$. When axial injection of hot electrons into achiral CNTs is strong enough, the nonequilibrium parameter η exceeds the critical value

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$\eta_c \approx 4.5 \times 10^{-9}$ and the NDC characteristics change to the PDC characteristics. Thus, the most important tough problem for NDC region which is the space charge instabilities that inevitably lead to electric field domains formation resulting in non uniform electric field distribution which usually destroys THz Bloch gain can be suppressed due to the switch from the NDC behaviour to the PDC behaviour [38]. This is mainly due to the fact that PDC is considered as one of the conditions for electric stability of the system necessary for suppressing electric field domains [38]. Hence a critical challenge for the successful observation of THz Bloch gain is the suppression of electric field domains by switching from NDC region to PDC region. This is similar to that observed by Mensah, et. al. [40] in effect of ionization of impurity centers in superlattice. To put the above observations in perspective, we display in figure 2, a 3-dimensional behaviour of the normalized current density (J) as a function of the applied dc field ($\mathbf{E}$) and nonequilibrium parameter (η) when frequency of ac field ω is much less than scattering frequency v ($\omega \ll v$ or $\omega\tau \ll 1$ i.e. quasi-static case, where $v = \tau^{-1}$) for the CNTs. The dc differential conductivity and the peak of the current density are at the highest when the nonequilibruim parameter η is zero. For both zz-CNT and ac-CNT, as the nonequilibrium parameter η gradually increases the dc differential conductivity and the peak normalized current density decrease until the critical nonequilibrium parameter value $\eta_c \approx 4.5 \times 10^{-9}$ is reached, beyond which the NDC characteristics slowly changes to PDC characteristics as shown in figure 2. We further display the behaviour of the normalized current density (J) as a function of the applied dc field ($\mathbf{E}$) of $ac - dc$ driven fields as $\omega\tau$ incrreasing from 0.01 to 0.15 when the nonequilibrium parameter
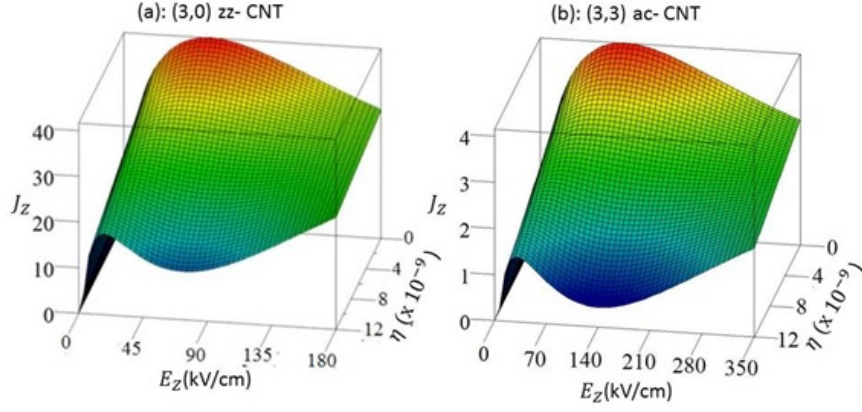
Figure 2: A $3D$ plot of normalized current density (J_z) versus applied dc field (E_z) as the onequilibrium parameter η increases for (a) $(3,0)$ zz-CNT and (b) $(3,3)$ ac-CNT, when $\omega << v$ or $\omega\tau << 1$ (i.e. quasi-static case), $v = \tau^{-1}$, $T = 287.5K$, $\omega = 10^{-4}THz$, $v = 1THz$ or $\tau = 1ps$ and $\omega\tau = 10^{-4}$

$\eta = 0.9 \times 10^{-9}$ (presence of hot electrons) and $\eta = 0$(absence of hot electrons) for $(3,0)$ zz- CNT and $(3,3)$ ac-CNT in figure 3. As we increase $\omega\tau$
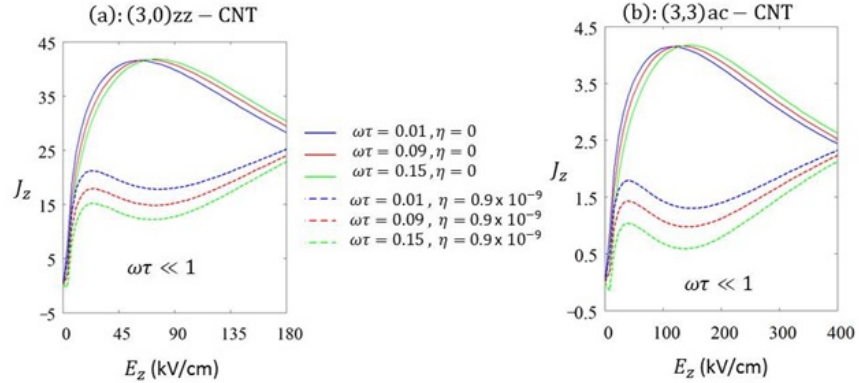

Figure 3: A plot of normalized current density (J_z) versus applied dc field (E_z) as $\omega\tau << 1$ increases from 0.01 to 0.17 for (a) $(3,0)$ zz-CNT and (b) $(3,3)$ ac-CNT when $\eta = 0$ and $\eta = 0.9 \times 10^{-9}$, $v = 1THz$ or $\tau = 1ps$

from 0.01 to 0.15 , we observed that the normalized current density has the highest peak at $\omega\tau = 0.01$. Upon increasing the $\omega\tau$, the peak current density

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decreases until the least peak is attained when $\omega\tau = 0.15$. Furthermore, we observed a switch from NDC to PDC near $75kV/cm$ and $140kV/cm$ for zz-CNT and ac- CNT respectively so far as $\omega\tau \ll 1$(i.e 0.01 to 0.15). Also the differential conductivity ($\partial J/\partial \mathbf{E}$) in NDC region is fairly constant as $\omega\tau$ increases from 0.01 to 0.15. However in PDC region after the switch from NDC, differential conductivity($\partial J/\partial \mathbf{E}$)fairly increases as $\omega\tau$ increases from 0.01 to 0.15 as shown in figure $3a$ and $3b$ for zz-CNT and ac-CNT respectively. In the absence of hot electrons ($\eta = 0$), we observed a shift of peak current density towards right (i.e high dc-field) as $\omega\tau$ increases from 0.01 to 0.15 for each achiral CNT. Hence, the current density dc field ($J - \mathbf{E}$)characteristics for CNTs show a negative differential conductivity at stronger electric field without hot electrons and with strong enough axial injection of hot electrons (i.e. $\eta \geq 0.9 \times 10^{-9}$), there is a switch from NDC to PDC leading to high electric field domain suppression necessary for generation of THz radiations provided $\omega\tau \ll 1$(i.e quasi-static ac field).

Conclusion

In summary, we have analyzed theoretically that strong enough injection of hot electrons in a CNT under conditions where, in addition to the dc field causing NDC, a similarly ac field is applied with a frequency ω much less than that of the scattering frequency v (i.e. $\omega \ll v$ or $\omega\tau \ll 1$, quasi-static case, $v = \tau^{-1}$), NDC switches to PDC. Hence, strong enough axial injection of hot electrons in CNT under the influence of quasi-static ac field results in a switch from NDC to PDC leading to the suppression of the destructive electric domain instability, predicting a potential generation of terahertz radiations whose applications are relevance in current-day technology, industry, and

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research. Although similarly effect has been observed in the absence of quasi-static ac field [37], the differential conductivity ($\partial J/\partial \mathbf{E}$) is higher and also hot electrons injection rate beyond which there is a switch from NDC to PDC represented by critical noneqilibrium parameter η_c) is lower in the presence of quasi-static *ac*- field than in the absence.

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