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Energy loss spectra of doped graphene and armchair carbon nanotubes at finite temperature

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Abstract. We have studied the dynamical polarizability associated to energy loss spectra of armchair carbon nanotubes in the context of tight binding model Hamiltonian including long range electronic interaction. Linear response theory in the context of random phase approximation has been implemented to obtain charge response function via calculating correlation function of density operators. The effects of both temperature and electron doping as well as the diameter on the frequency behaviour of charge response have been investigated. The sharp peak in the energy loss spectra of the armchair carbon nanotube disappears upon raising the temperature. This is not the case of non interacting case where temperature has no considerable effect on the dynamical susceptibility. Also, the similar calculations have been performed for graphene sheet.

1. Introduction

Carbon nanotubes (CNTs) are newly realized two and one dimensional electronic systems that have attracted great deals in the scientific community because of new physics they exhibit and because of their potential as new materials for technology [1]. This compound, which is made of a single graphite layer rolled up into a hollow cylinder, is a one dimensional allotrope of carbon. In order to study electronic properties of electron gas in the nano structures and graphene, the dynamical polarizability, whereby the screening effects have been found, is required [2]. The dynamical polarizability renormalizing the phononic Green's function can explain the phonon softening and Kohn anomaly phenomenon [1] at the Γ point. So far, the extensive studies have been performed on the charge response function of monolayer Graphene in the Dirac cone approximation so that the energy dispersion of the hexagonal lattice is considered to be linear form in terms of electronic wave vector. In the case of undoped graphene at zero temperature, the polarizability has been found by Gonzalaz [3]. Their results demonstrated the fluctuations of density in a bilayer case can present either single-component massive-chiral character or standard two layer character, depending on energy and doping. But, the effect of full band dispersion over all the Brillouin zone on the charge response of nano structures, such as nanotubes, is expected to be significant. In the obtaining of high energy corrections to the charge response, both inter and intraband transitions contribute to the frequency behavior of dynamical polarizability. Recently, a theoretical work provides a study of both imaginary and real part of non-interacting polarizability of graphene within an analytical approach [4]. The authors found that there is no noticeable angle dependence for imaginary part of polarizability around the van hove singularity, i.e $\frac{\hbar\omega}{t} = 2$ where t implies nearest neighbor hopping integral. An extensive study on the behavior of dynamical polarizability in various wave vector regions

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for graphene sheet beyond Dirac approximation has been performed by Peres et al. [4]. In the present paper, we will extend the Stauber's work [4] so that the results of polarizability or energy loss spectra for the other nano structures, such as armchairs CNTs, are presented. Here, we apply a random phase approximation to find the analytical form of polarizability for CNTs at finite temperature. In addition, to derive the results of charge response for graphene monolayer performed by Stuber and co-workers [4], we also obtain the noninteracting charge response of CNTs for various amounts of temperature, concentration and diameter. Interacting charge response function can be readily derived by using RPA approximation in which quantum correlation effects on the charge response are neglected. Also we investigate the plasmon excitations of electrons of CNTs for both interacting and noninteracting cases at finite temperature. The effects of diameters and electronic concentrations on the frequency behavior of polarizability as criterion of electron energy loss function and the collective electronic excitation have been also addressed.

2. Materials and method

The dynamics of electrons on the honeycomb lattice as graphene or CNT is described by the following tight binding model including a long-range Coulomb interaction term:

$$H = \sum_{k,\sigma} \epsilon_k c^{+\sigma}_{k,A} c_{k,B} + h.c. + \sum_{q,\alpha,\beta} v_q \rho_\alpha(q)\rho_\beta(-q), \quad (1)$$

where $v_q = 2\pi e^2/q$ and $\rho_{q,\alpha}$ are the Fourier transformation of Coulomb potential in two dimensional and electronic density operator, respectively. N is the number of unit cells in honeycomb lattice. Furthermore, we have $\epsilon_k = 1 + \cos(k_x/2)e^{-ik_y\sqrt{3}/2}$ where k_x and k_y belong to the first Brillouin zone of honeycomb lattice.

The quantity of interest for studying many particle properties such as plasmon oscillations and phonon softening is the dynamical charge response function. Also this quantity determines the effective electron-electron interaction and the Friedel oscillations. Linear response theory gives us the noninteracting charge response based on the correlation function of density-density operators as:

$$\chi^{(0)}(q, i\omega_n) = -\frac{1}{A} \sum_{\alpha,\beta=a,b,\sigma} d\tau e^{i\omega_n \tau}$$

$$\langle T(\rho^\sigma_\alpha(q,\tau)\rho^\sigma_\beta(-q,\tau))\rangle = 2\sum_{\alpha,\beta} \int_0^\beta e^{i\omega_n\tau}\chi^{(0)}_{\alpha\beta}(q,\tau), \quad (2)$$

where σ denotes the spin degree of freedom of electron. Also electronic density operator $\rho^\sigma_\alpha(q)$ is defined as:

$$\rho^\sigma_\alpha(q) = \sum_k c^+_{k+q,\sigma} c_{k,\sigma}.$$

Since paramagnetic phase is considered for CNT and graphene sheet, the contributions of two different spin components to the charge response are the same. Therefore the summation over σ is readily performed and coefficient 2 comes from spin degeneracy. Also A is the area of the nanostructure sample. $\omega_n = 2n\pi/\beta$ denotes the Bosonic Matubara frequancy in which β is the inverse of equilibrium temperature. Wick's theorem has been applied to write the charge response in terms of matrix elements of non-interacting electronic Green's function. Using the Fourier transformation in Matsubara's representation [2], charge response function can be written in the following form:

$$\chi^{(0)}_{\alpha\beta} = \frac{1}{N\beta}\sum_{k,m} G^{(0)}_{\alpha\beta}(k+q, i\omega_m) G^{(0)}_{\beta\alpha}(k, i\omega_m + i\omega_m), \quad (3)$$

where $\omega_m = (2m+1)\pi/\beta$ denotes the fermionic Matsubara frequency. According to the model Hamiltonian introduced in Eq. (1), the elements of electronic Green's function in the Fourier space get the following forms [4]:

$$G^{(0)}_{AA}(k, i\omega_m) = G^{(0)}_{BB}(k, i\omega_m) = \sum_{\lambda=\pm} \frac{1}{2((i\omega_m) - \epsilon_\lambda(k))},$$

$$G^{(0)}_{AB}(k, i\omega_m) = \sum_{\lambda=\pm} \frac{\phi^*}{2\epsilon_+} \frac{\lambda}{2((i\omega_m) - \epsilon_\lambda(k))},$$

$$G^{(0)}_{AB}(k, i\omega_m) = \sum_{\lambda=\pm} \frac{\phi}{2\epsilon_+} \frac{\lambda}{2((i\omega_m) - \epsilon_\lambda(k))}, \quad (4)$$

where the factors in the above equations are given by:

$$\epsilon_\lambda = t\lambda|\phi(k)|,$$

$$\phi(k) = 1 + \cos(k_x/2)e^{-ik_y\sqrt{3}/2}.$$

ϵ_λ introduces each of electronic band energies of honeycomb lattice.

Substituting Eq. (4) into Eq. (3) and performing Matsubara frequency summation over fermionic Matsubara's energies gives us the following expressions for one of the elements of dynamical susceptibilty tensor:

$$\chi^{(0)}_{AB}(q, i\omega_n) = \frac{1}{4N}\sum_k \frac{n_F(\xi_{\lambda,k+q}) - n_F(\xi_{\lambda',k})}{i\omega_n + \xi_{\lambda,k+q} - \xi_{\lambda',k}}, \quad (5)$$

where $n_F(E) = 1/(e^{\beta E} + 1)$ is the Fermi-Dirac distribution function. The charge response of electrons on the honeycomb lattice has been found after summation over the charge response elements in Eq. (5). Random phase approximation [2] has been employed to correct charge response function of electronic system

in the presence of electron-electron interaction part of Hamiltonian in Eq. (1), and the interacting dynamical response function is given by:

$$\chi(q, i\omega_n) = \frac{\chi^{(0)}(q, i\omega_n)}{1 - v_q \chi^{(0)}(q, i\omega_n)}. \quad (6)$$

Under a simple analytical continuition as:

$$i\omega_n \rightarrow \omega + i0^+,$$

retarded charge response function has been obtained from its Matsubara form. In the next section, we present the results of interacting and noninteracting charge response function for armchair CNT with various diameters and graphene sheet.

3. Numerical results

In this section, we present the numerical results of energy loss spectra corresponding to the imaginary part of dynamical polarizability introduced in Eqs. (5) and (6) for both armchair CNT and graphene sheet. We address the effects of electronic interaction on the eneregy loss spectra within RPA approximation. The effects of electron doping associated to the increase of chemical potential, μ/t, on the noninteracting energy loss spectra at wave vector, q_0, in low energy region, $0 < \omega/t < 0.5$, have been presented in Figure 1. The position of the point q_0 with amplitude $|q_0| = 1/(10\sqrt{3})$ and polar angle $\varphi = \pi/6$ into the Brillouin zone has been shown in Figure 1. The behavior of noninteracting energy loss spectra is free of chemical potential effects at high normalized frequencies and we have not shown it here. As it is obvious in Figure 1, a collective mode associated with resonances in the charge response function is excited around normalized frequency $\omega/t \approx 0.16$. As it is known, the imaginary part of charge response function is proportional to the electronic energy loss spectra [2]. Therefore, the position of peaks in the energy loss spectra is associated with the energy amounts that cross section of scattering of particles from sample get its maximum value. A novel feature shown in Figure 1 is the vanishing of frequency gap in the energy loss with electronic concentration.

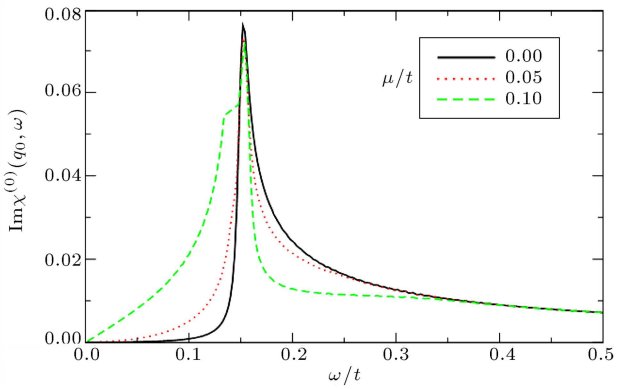

Figure 1. Noninteracting energy loss spectra of grapheme sheet as a function of normalized energy ω/t for different values of chemical potential $\frac{\mu}{t}$ at a particular wave vector q_0. The normalized temperature is assumed to be $\frac{kT}{t} = 0.05$.

The effect of electron doping on the behavior of energy loss spectra of armchair CNT (9,9) in the noninteracting regime for wave vector $\boldsymbol{q_1}$ has been presented in Figure 2. The wave vector $\boldsymbol{q_1}$ is one of the points in the Brillouin zone with high symmetry. By increasing the electronic concentration, the width of frequency gap in $\mathbf{Im\chi^{(0)}(q_1, \omega)}$ decreases. Energy loss spectra for half filling case gets a zero value in the energy region $\mathbf{0.0 < \frac{\omega}{t} < 1.0}$ while the energy gap range is in region $\mathbf{0.0 < \frac{\omega}{t} < 0.5}$ for electron doped case $\mathbf{\frac{\mu}{t} = 1.7}$. Figure 2 also implies plasmon modes of armchair CNT (9,9) shift to the lower frequency upon increase of chemical potential. We have also studied the effect of diameter on the noninteracting energy loss of armchair CNT for half filling case at $\mathbf{\frac{kT}{t} = 0.01}$.

In Figure 3, we plotted the frquency behavior

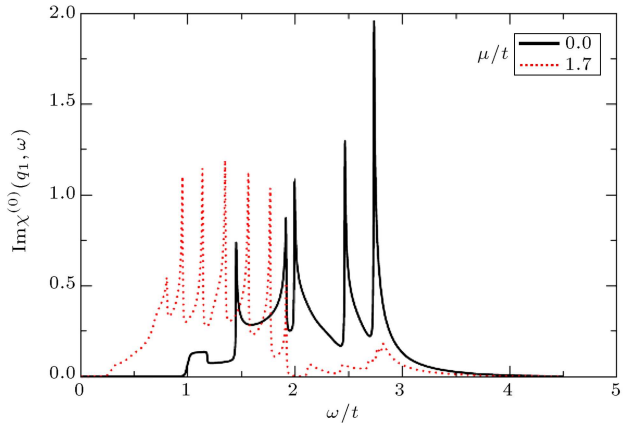

Figure 2. Energy loss spectra of CNT (9,9) for various chemical potentials.

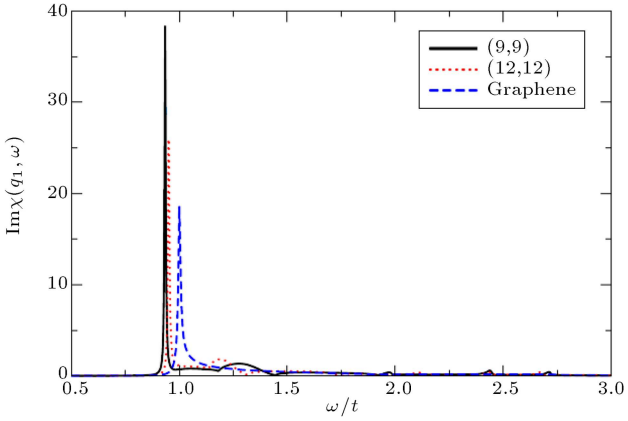

Figure 3. Interacting energy loss spectra of armchair CNT with different diameters.

of interacting energy loss spectra for three different diamters of armchair CNT at half filling case and $\frac{kT}{t} = 0.05$. The frequency gap in the energy loss spectra is approximately independent of the tube's diameter. Moreover, the intensity of energy loss spectra decreases with the radios of nanotube, and the peak position moves toward higher frequency.

4. Conclusions

In conclusion, we have presented the frequency dependence of the energy loss spectra of armchair CNT and monolayer grapheme. Using random phase approximation and a Green's function approach, the energy loss spectra has been studied. In particular, the effect of electron doping and temperature have been investigated. We have found that the electronic concentration has a major effect on the energy loss spectra. Also the results show that the increase of diameter leads to the intensity reduction of energy loss spectra of armchair CNT.

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