Chapter 5

Chapter 5: Summary And Conclusions

The development in the field of nanoscience is fuelled by observed unusual properties of nanometre size matter, which may be exploited to improve the performance in various applications such as optics, microelectronics, thermo electrics and magnetics. To study the low dimensional system in detail it is appropriate to model it in terms of electron gas/liquid picture. Graphene because of its extremely thin structure is compared with electron gas or electron liquid model as per its application. Graphene has spawned tremendous interest and activity in studying the properties of this unique system of 2DEG. Because of the different energy band dispersions, screening properties in graphene exhibit significantly different behavior from the conventional two dimensional systems.

Chapter 1 of thesis presents a brief review of Graphene and graphene based systems such as Bilayer graphene, Gapped Graphene and Graphene Superlattices. Graphene is a single layer of carbon atoms covalently bonded together in free state. Its real-space lattice structure is a honeycomb shaped structure lying in two dimensional plane. The primitive unit cell of a Bravais lattice is a parallelogram. The most unique feature, that makes it different from conventional other two dimensional electron gas, is its linear dispersion relation and thus the band structure is linear with zero gap between valence and conduction band. Experimentally the first evidence of existence linear dispersion was given by S.Y.Zhou *et.al.*, using ARPES. Its unique properties makes it a potential substance for developing efficient devices for scientific and technological applications. Some of its unique properties include high mobility, linear energy dispersion relation and many more to its attribute. Single layer graphene being a gapless semiconductor, its Fermi level lies at

Dirac point in intrinsic case. This level can be shifted by applying an external gate voltage, which introduces 2D free carriers, electrons or holes, producing extrinsic graphene with gate-voltage-induced tuneable carrier density.

The simplest method to obtain graphene is peeling of the graphite using a Scotch Tape, as was developed by Novoselov and Geim in 2004 leading to groundbreaking development in Science and Technology. Since then various groups have worked upon to obtain graphene on large scale implementing various experimental methods using top-bottom or bottom-top approach such as Exfoliation and cleavage, Plasma enhanced chemical vapor Deposition techniques, Various chemical methods, Thermal Decomposition of SiC ,Thermal Decomposition on other substrates, unzipping CNTs etc.

There have been various attempts made to modify graphene, based on the requirements in industries for its useful applications, such as Bilayer graphene, gapped graphene, graphene superlattice, Graphene Nanoribbons, Graphene arrays, etc. This chapter also reports, briefly, unique features of Bilayer graphene, gapped graphene and graphene superlattice. Bilayer graphene consists of two single layer graphene placed above one another in Bernal stacking arrangements. It consists of four nondegenerate bands, while gapped graphene has a gap opening at its Dirac point changing the shape of band structure from linear to parabola. The $E \rightarrow \vec{k}$ relation of gapped graphene differs from that of single layer graphene (SLG) by a value of Δ i.e. $E(\vec{k}) = \sqrt{\hbar^2 v_f^2 k^2 + \Delta^2}$. This gap can be introduced by Spin-Orbit Interaction, Breaking of Sublattice symmetry, External magnetic field, finite geometry effect and various other instabilities. The density of states, interaction parameter for single layer graphene, Bilayer graphene and gapped graphene differs drastically from each other, making them unique.

The modelled superlattice consists of repetitive structure of single layer graphene on SiO_2 substrate. This type of modified structures can be of great scientific and technological application, because of the enhancement in properties and work efficiency as compared to original crystal structures. Superlattice structures can be experimentally realized with the help of molecular beam epitaxy (MBE), sputtering techniques etc. The study of GBS helps to analyze the experimental study of Raman spectroscopy which characterizes the superlattice systems. Change in the properties can be attributed to the change in band structure of the superlattice.

A review of Many particle aspects and collective excitations has been presented. The Many-body theory provides the framework for understanding the collective behavior of vast assemblies of interacting particles and the changes induced by these effects on a given system. These effects are relevant only in systems containing large numbers of constituents and its study can be extremely complex because the motions of particles are intricate. Many particle theory paves way to understand various properties such as collective excitations, exchange and correlation, scattering, screening, energy levels, susceptibility, superconductivity, ground state properties etc. One of the most successful and widely accepted approaches to solve the problem of screening has been the random-phase approximation (RPA). The RPA is a many body theoretic method by which quantitative predictions beyond the Hartree–Fock model can be made. In this approximation it is assumed that only single-particle excitations of the same wave vector as the Coulomb interaction plays an effective role in the screening process while the effects of others having different wave vectors cancel out. Use of the RPA is justified when the electron-electron interaction is strong enough that quantum coherence does not dominate.

A brief review of density-density response function and RPA bubble diagram is reported to derive the expression for dynamic polarization function, which has been a fundamental quantity applied to obtain various ground state properties and collective excitations of Graphene and graphene based system throughout the thesis. When a system with certain density is perturbed with onset of some external density, it leads to density fluctuation due to disturbances created. It is important to conceptualize this response of the system to understand basic character of material. A physical quantity of crucial utility in the understanding of the many body properties of condensed matter is the dynamic electron density-density response function χ. Importance of Dielectric function and quantities based on this function has been explained and the literature survey has been presented. The important ground state properties studied are Structure factor, Pair correlation function, screening charge density, self-energy and screened potential, compressibility, energy loss, wake effects, while the plasmon-phonon coupling of graphene superlattices has been reviewed.

Chapter 2 reports study on structure factor, pair correlation functions, self energy, density of screening charge and screened potential within and beyond the RPA by incorporating local field corrections in the Hubbard approximation using the density-density and spin density response functions. We find that the computed structure factor diverges with increasing k due to the continuum model of the relativistic Dirac equation. After the regularisation of the response functions the static structure factor shows a broad hump and then saturates, but for increasing values of coupling strengths the structure factor begins to grow slightly with wave vector. The regularised magnetic structure factor also shows a broader hump that saturates for higher values of x for all coupling constants unlike the case of static structure factor.

A striking feature that comes fore in our study of magnetic structure factor is the observation of sharp peaks at large coupling strengths, $\alpha = 3\ \&\ 4$, which is a signature of paramagnetic instability and is due the electronic exchange interaction. However, in contrast to the regularised static structure factor the regularised magnetic structure factor is seen to saturate for increasing values of wave vector at large coupling constants too. The symmetric spin pair correlation function worked out in this Hubbard scheme at low densities shows negative values at zero separation but at very high densities this discrepancy is removed. However, the Hubbard approximations fails in the estimation of parallel spin and anti parallel spin pair correlation functions which suggests that other competing local field corrections should be tested and also be verified through quantum Monte Carlo simulations. The results obtained on the static structure factor or the pair correlation function can be used to calculate the exchange energy per electron of the graphene in the electric potential created by its own exchange hole and the electron-electron correlation energy. And by a familiar strategy of integration of the structure factor by combining with another coupling constant our results can be used to obtain the interaction energy contribution to Helmholtz free energy or the thermo dynamical potential of the 2D Dirac electron gas. Besides the results can be of interest in building beyond linear density approximation exchange-correlation energy density functional and also the magnetic response of the Dirac electron gas. Self energy, density of screening charge and screened potential of MLG are found to behave similar to that of 2DEG since the dielectric function is almost similar upto $q < 2k_f$. Freidel oscillations have been observed in screened potential and density of screening charge. With increase in the value of α for a fixed value of carrier density, Friedel oscillations enhance, whereas on keeping α fixed and increasing carrier density reduces the amplitude of

the oscillations. The observation of Friedel oscillations in screened potential and screening charge density can be seen as a signature of Fermi liquid state in graphene. Pair distribution function is calculated as a function of carrier density suggests that exchange and correlation terms make negligible contribution to compressibility of raphene. Incorporation of LFC reduces the magnitude of self energy, screening charge density and screened potential. It is envisaged that the present work will further stimulate experimental and theoretical work on structure factor, pair correlation function and spin polarised phases in graphene.

Chapter 3 presents analytical and numerical results for energy loss and wake effects in gapped graphene. Gapped graphene is modelled to be a single layer graphene sheet lying on SiO_2 surface. The gap between valence and conduction band can be introduced using various techniques such as radiating it with circularly polarized light, substrate induced techniques, spin orbit interactions, doping, absorption of water molecules, applying gate voltage, confinement of dimensions (dot, nanoribbons), hybrids, Kekule lattice distortion etc. All these techniques can create a gap of approximately 10^{-3}eV upto 0.5 eV.

Numerical calculations on structure factor reveals that, since $\chi(q,\omega)$ is linearly proportional to q at large q and decreases only like ω^{-1} at large ω in both gapped and gapless graphene, accordingly the structure factor obtained is divergent. In order to improve divergence, we need to add and subtract vacuum polarization inside frequency integral and regularize the structure factor for gapped graphene as well.

Our model consists of gapped graphene lying on SiO_2 substrate, where a particle is travelling above gapped sheet at a distance. The two mechanisms studied in this chapter are Energy loss and wake effects. As the particle travels above the sheet it will interact with the sheet losing some energy to the system as well as

creating wake effects travelling behind the particle as it moves in forward direction. These two quantities are important to be investigated as it reflects the characteristics of the system under consideration, when being interfered by some external objects. The energy loss of the gapped graphene has a solution in terms of Bessel's function, similar to SLG, but differs in magnitude by a value of $1/1 + b^2$. Thus the magnitude energy loss in gapped graphene is less than that in single layer graphene. Thus with a gap being introduced, gapped graphene can be seen as an alternative to semiconductor based devices. Numerically the energy loss function can be obtained from structure factor of the system.

Wake effects is an important physical quantity to be studied to gain knowledge about the response of the system to any external perturbation. A simple analytical calculation yields the solution in terms of Whittaker function. The induced number density exhibits oscillations on both sides of the particle position, similar in the case of carbon nanotubes.

Chapter 4 deals with Plasmon-phonon coupling and energy loss in superlattice. Unique properties of graphene, inspired to think that graphene based multilayer and superlattice structures stand a better chance of application in various industries. The interlayer interactions are strong enough to make it a better component that can be utilized in developing efficient electronic devices.

The plasmon frequency without the effect of superlattice and without phonon coupling has a $\sqrt{q}$ dependence. Inclusion of structure factor for superlattice changes the q-dependence as well as the magnitude of plasma frequency. At lower boundary the dispersion is linear while the upper boundary has dispersion less limit.

The solution of equation $\epsilon(q, \omega, q_z) = 0$ has four roots, two modes for each value of $\cos(q_z d)$, one of which is plasmon like while other is phonon like. For $\cos(\mathrm{q_z d}) =$

-1, one of the two energies corresponds to $\omega_0 qd/\sqrt{2}$, while other is close to ω_{Lo} for $q \to 0$. When $\cos(\mathrm{q_z d}) = 1$ and $q \to 0$, plasmon like mode approximately approaches to $\omega_o\sqrt{2}$ and phonon like mode is close to $\left(\omega_{TO}^2 + (\frac{\omega_{LO} q}{q_s})^2\right)^{1/2}$. We find that Plasmon-Phonon coupling is stronger at higher electron densities. It is to be noted that CPPM observed in GBS are very different than the coupled plasmon-phonon modes found in SLG. Optical bulk like modes that has frequency $\omega_o\sqrt{2}$ and acoustic modes like $\omega_0 qd/\sqrt{2}$ cannot be observed in SLG. For weak coupling case the behaviour of GBS is similar to SLG. The coupling of CPPM is stronger for decreasing value of interlayer distance. The decay of CPPM is dominated by Dirac delta function i.e. $\delta(\omega^2 - {\omega_{LO}}^2)$. The decay in low frequency depends on interlayer distance and charge density. Using real and imaginary parts of dielectric function susceptibility dependent refractive index can be obtained.

The attainable values in our results are compatible in Near Infrared (NIR) to visible spectra, and hence making it feasible to develop such high-frequency based devices. The devices working in this range are image sensors, optical fiber cables, photo detectors, modulators, opto-electronic switches etc. The electromagnetic radiations in this regime can resonate with the CPPM of GBS making it possible to realize devices working in this frequency range. Though there have been GNRs developed which can work in this same frequency regime but GBS can also be viewed as an equivalent efficient device. Plasmon-Phonon coupling mode helps to gain insight in heat capacity, thermal conductivity and various other important quantities which should be considered during device fabrication.

When an external particle perturbs the system, it results into collective excitations and single-particle excitation regimes. These interactions results in energy loss. As the charge density is increased the intensity of energy loss is increased. With

increase in distance between the layers (i.e. almost SLG behavior) the zero energy loss regions is more compared to more compact GBS. Thus GBS can be considered as better device making material with lesser energy loss than SLG. This low energy loss in GBS can have better technological applications as energy storage devices.

Glossary Term	Meaning
CNT	*Carbon Nanotube*
SLG	*Single Layer Graphene*
BLG	*Bilayer Graphene*
GBM	*Graphene Based Multilayer*
GBS	*Graphene Based Superlattice*
2DEG	*Two Dimensional Electron Gas*
2DQEG	*Two dimensional quantum electron gas*
DOS	*Density of States*
RPA	*Random Phase Approximation*
IRPA	*Improved Random Phase Approximation*
QHT	*Quantum hydrodynamic Theory*

MDF	*Massless Dirac Fermions*
LFC	*Local field Correction*
EELS	*Electron Energy loss Spectroscopy*
AREELS	*Angle-resolved reflection electron-energy-loss spectroscopy*
ARPES	*Angular Resolved Photoemission Spectroscopy*
STEM	*Scanning Tunneling Electron Microscopy*
HREELS	*High resolution electron energy loss spectroscopy*
SiO_2	*Silicon Dioxide*
SiC	*Silicon dioxide*
PECVD	*Plasma enhanced chemical vapour deposition*
HOPG	*Highly Oriented Pyrolytic Graphite*
CPPM	*Coupled Plasmon-Phonon modes*

GNR	*Graphene Nano-ribbons*
GNRA	*Graphene Nano-ribbon Arrays*
MIR	*Mid-Infrared*
NIR	*Near Infrared*
THz	*TeraHertz*