Formation and Transport of Correlated Electronic States at Room Temperature in Graphene Bilayers

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We propose a combination of methods to investigate the characteristics of electron-hole bilayer graphene at spontaneous coherence using novel path integral Monte Carlo and Green's function based methods at room temperature. Using the presented numerical techniques at room temperature, we find a sharply peaked differential tunneling conductance and quasiparticle paths which wind around our solution domain both of which are indicative of the formation of a strongly correlated electronic state at room temperature.

Introduction

In normal semiconducting systems, strongly correlated effects are seen only at low temperatures. For example, the ν = 1 quantum Hall state in semiconductor bilayers is only observed at large magnetic fields and low temperatures (1). Such a state is often described as type of electron-hole condensate. Roughly speaking, one can identify electrons in the half-filled top layer as being strongly correlated with holes in the half-filled bottom layer (1). As is the case for other quantum Hall phenomena, the strong magnetic field plays an important role of quenching the electrons kinetic energy, allowing Coulomb interactions to play a dominant role.

Recently, there has been interest in whether correlated hole states can form in bilayer graphene: two sheets of graphene separated by a thin insulator. In electron-hole bilayer graphene, the Kosterlitz-Thouless transition for small interlayer separations has been predicted to exceed room-temperature (2). This theoretical proposal is quite remarkable, as no magnetic field is invoked to drive this strongly correlated system, rather, the correlation is predicted to emerge from intrinsic properties of the bilayer. Two features make bilayer graphene an attractive candidate for electron-hole condensation: (i) its small physical dimensions, since each layer is only one monolayer (ML) thick, and (ii) its identical dispersion of electrons and hole. Similar excitonic condensates have recently been predicted to occur in thin films made from topological insulators (3).

Here we will discuss the calculation of room-temperature interlayer transport in bilayer graphene heterojunction devices using novel path integral Monte Carlo (PIMC) algorithms and a variant of Non-Equilibrium Greens Functions (NEGF). Our preliminary simulations of bilayer graphene show the same sharp peak in the interlayer differential conductance as was found in semiconducting bilayers in the FQHE regime.

Bilayer Device Structure

The device structure is illustrated in Fig. 1. Two sheets of graphene, each one monolayer thick, 40 nm wide, and 100 nm in length, separated by 1 nm of SiO₂. The oxide barrier isolates the two layers from one another while still allowing the quasiparticles within one layers to interact with the particles in the other layer. Above and
below each monolayers of graphene is a 20 nm buffer of SiO$_2$ and then a metal gate. The pair of gates control the quasi-particle density in each of the layers. We apply a bias from the left hand side of the top layer to the right hand side of the bottom layer to drive an interlayer current.

![Diagram of a bilayer graphene tunneling device](image)

**Figure 1.** Bilayer graphene tunneling device. Two sheets of graphene are separated by a one-nanometer thick insulating oxide layer. Electron and hole densities in the two layers are set by the top and bottom gates and currents are driven by the four leads.

### Calculation Methodology

Single electron and hole quasiparticles in a single graphene sheet are often described by a Dirac Hamiltonian,

$$H^0 = \left(-i \hbar v_f \vec{\alpha} \cdot \vec{V}\right)$$  \[1]\n
where $v_f \approx 10^8$ cm/s and $\vec{\alpha} = (\sigma_x, \sigma_y)$ is a two-dimensional vector of Pauli matrices that act on a spin-like degree of freedom arising from the A and B sublattices of hexagonal graphene lattice. This Hamiltonian captures the low-energy physics of electrons and holes on Dirac cones near the Fermi points of a neutral graphene layer. The electrons and holes also have a true spin degree of freedom that feels a very weak spin-orbit coupling, which is ignored in Equation [1].

We begin our analysis of the interacting bilayer system by defining the system Hamiltonian in pseudospin notation where the top layer is $\uparrow$ and the bottom layer is $\downarrow$. The Hamiltonian can then be written as,

$$H = H^0_\uparrow + H^0_\downarrow + \Delta_i \left( \left| \uparrow \rangle \langle \downarrow \right| + \left| \downarrow \rangle \langle \uparrow \right| \right) + \hat{V}$$  \[2]\n
where $H^0_\uparrow$, $H^0_\downarrow$ are the single particle Hamiltonians, Equation [1], for the top and bottom layers, $\Delta_i$ is the splitting between symmetric and anti-symmetric states due to tunneling between layers, and $\hat{V}$ represents all interactions between quasiparticles. We model the 1 nm of insulating oxide by setting $\Delta_i = 1$ µeV.
In References, (2) and (3), all effects of Coulomb interactions are ignored except for an enhancement of interlayer tunneling that emerges from a mean-field treatment of Bose condensation. Roughly speaking, one neglects interactions except for an “onsite” Coulomb interaction for an electron and hole adjacent to each other, but in opposite layers. Assuming a condensation of excitons that drives a non-zero expectation value for the operator, \( \langle a_i a_i^\dagger \rangle \), they find an effective mean-field Hamiltonian,

\[
H = H_0^0 + H_0^0 + \Delta_r \left( \langle \uparrow \downarrow \rangle + \langle \downarrow \uparrow \rangle \right) + U \left( \langle \uparrow \downarrow \rangle \right) e^{i\phi} + \langle \downarrow \uparrow \rangle \langle \uparrow \downarrow \rangle e^{-i\phi}
\]  

where the last term is a many-body enhancement to tunneling created by the electron-hole condensate. While the first three terms are simply a function of the graphene and oxide insulator parameters, the third term contains the on-site Coulomb interaction, \( U \), and the effective pseudospin field, \( \phi \). The effective pseudospin field can be found simply by taking expectation values of the resulting density matrix, however, the on-site Coulomb term must be calculated before we may examine the transport properties of the system.

Path integral Monte Carlo for estimation of quantum coherent tunneling enhancement

To obtain the Coulomb tunneling enhancement term, \( U \), we need a many-body method capable of accurately treating electron-hole correlations and possible Bose condensation of electron-hole pairs. We are attempting new PIMC simulations of interacting electrons and holes to extract the tunneling enhancement for realistic device parameters. Here, we present our simulation strategy and some preliminary results.

In PIMC simulations, we work with the full many-body thermal density matrix and recognize that it can be calculated as an imaginary-time path integral as,

\[
\rho(R, R'; \beta) = \frac{1}{Z} \int_{R(\beta)\rightarrow R} D\tau \exp\left(-\frac{1}{\hbar} S_E[R(\tau)]\right)
\]  

In Equation [4], \( Z \) represents the partition function, \( \beta = (k_B T)^{-1} \), and \( S_E \) is the Euclidean action. In our simulations, \( S_E \) includes the kinetic action, as well as the confining potential and Coulomb actions. We use the fixed node approximation (4) to handle the “fermionic sign problem” and use periodic boundary condition to simulate condensation on an infinite sheet of graphene.

We are pursuing two strategies to predict the tunneling enhancement:

**Strategy 1: Atomistic PIMC:** In order to capture the Dirac physics of the graphene layers, Eq. (1), within quantum Monte Carlo, we find it useful to start from a tight-binding model that retains one \( \text{pz} \) orbital from each carbon atom. For the PIMC simulations, it is convenient to map this system onto a hexagonal lattice of hydrogen atoms, which preserves the underlying physics of Eq. (1) while being more manageable for computation. In Figure 2 we show charge densities and typical imaginary-time paths from a PIMC simulation of bilayer graphene. For these initial calculations, we limited the periodic simulation cell to 120 electrons, and artificially decreased the interlayer separation to scale with the small simulation cell.
In the path-integral formalism, tunneling appears as instantons: rapid crossing of paths between potential minima. In Fig. 2(d), one can see several paths crossing between layers. Such instanton paths correspond to the bare tunneling term ($\Delta t$, which is considerably larger than 1 neV in these preliminary calculations) and the tunneling enhancement due to correlated electron-hole interactions ($U$) in Equation [3]. Quantitative extraction of $U$ from these simulation will require solution to several unresolved problems: (i) this tight-binding graphene model will have to calibrated with physical graphene, (ii) the effects of bare tunneling and correlation enhanced tunneling may have to be separated in the analysis of the PIMC data, and (3) finite size-scaling will have to be considered, due to the small dimensions of the simulation cell in Figure 2.

To calculate the on-site Coulomb term, we examine the interacting interlayer conductivity versus the non-interacting interlayer conductivity. The ratio of these values will give us the interaction enhancement of the tunneling which will become part of the on-site term we desire. To calculate the interlayer conductivity, we work with the imaginary time current-current correlation functions. These correlations functions then may be used in the Kubo formula for the electrical conductivity. The current-current correlation function is collected by watching the paths fluctuate between the two layers, as seen in Figure 2. Here we see paths fluctuating from the top and bottom indicating particle transfer between the layers. When the system is at spontaneous coherence, we expect typical values of the exchange coupling between the layers to be \(~10\%\) of the system Fermi energy.

**Figure 2.** Charge densities (a,c) and typical paths (b,d) from atomistic PIMC simulations of interacting electrons in bilayer graphene. Here graphene is modeled with one orbital per atom, corresponding to the electronically active out-of-plane carbon 2p$_z$ orbitals. In this figure, the red paths represent spin up electrons and the blue paths represent spin down electrons. Charge densities are collected as a Monte Carlo sum over many such paths. An electric field in the -z direction (applied with the top and bottom gates in Figure 1) has pushed 0.6 electrons from the bottom layer to top layer, corresponding to a carrier density (electrons quasiparticles in the top layer and hole quasiparticles in the bottom layer) of $4\times10^{11}$ cm$^{-2}$. From Reference (10).
Strategy 2: Quasi-particle PIMC: In the atomistic PIMC approach, the simulations are challenged by two different length scales. The atoms are separated by a very short carbon-carbon bond length, while the average separation between quasiparticles may be much lower. When considering an electron-hole condensate, it would be desirable to consider dozens or even hundreds of electron-hole pairs. Such a simulation would likely involve thousands of atoms, and may not be tractable in an atomistic simulation. As an alternative approach, we are conducting quasiparticle PIMC simulations, illustrated in Figure 3.

In quasiparticle PIMC simulations, we simulate the electron and hole quasiparticles, allowing much larger simulation cells. Such calculations have been used to study three-dimensional electron-hole condensates in model semiconductors [5]. The ability to simulate larger cells comes at a price: (i) simulation of the Dirac Hamiltonian, Eq. (1), becomes much more difficult, so we instead model graphene as a parabolic semiconductor with identical electron and hole masses, and (ii) in our constant-number simulations we are unable to include electron-hole recombination, so tunneling between layers is not explicitly included in the simulations. Instead, the Coulomb enhancement of tunneling in quasiparticle PIMC must be inferred from analysis of interlayer correlation of electrons and holes—a subject of future research.

One advantage of the quasiparticle PIMC technique is that Bose-Einstein condensation of excitons can be directly simulated. An excitonic superfluid appears as long permuting paths that wind around the periodic boundaries of the simulation cell (5,6). As an example, in Figure 3 we show electron and hole paths for a GaAs/AlGaAs bilayer, where we have artificially set \( m_h = m_e \). In Figure 3(a), the temperature is 300K and we see excitons, but no superfluid. In Figure 3(b), the temperature is 30K and we see excitonic superfluidity.

![Figure 3](image_url)

Figure 3. Typical paths from a PIMC simulation of quasiparticle excitons in a GaAs/AlGaAs bilayer, illustrating (a) excitons above the condensation transition temperature, and (b) room-temperature excitonic Bose condensation.

Non-Equilibrium Transport at Spontaneous Coherence

With the Hamiltonian elucidated, we now look to find the non-equilibrium transport properties of the bilayer system. For this section, we assume the Hartree-Fock value of
the tunneling enhancement $U$ to be $\sim 10\%$ of the system Fermi energy, for the system is at spontaneous coherence. To explore interaction physics in bilayer transport qualitatively we use a local-density approximation in which the interaction contribution to the quasiparticle Hamiltonian is proportional to the pseudospin-magnetization at each point in space. In this representation we may define the resulting pseudospin fields that appear in Eq. (3), and comprise the $\phi$ coherence term in the electron-electron interaction as,

$$\Delta_x = \rho_{\uparrow\uparrow} + \rho_{\downarrow\downarrow}, \quad \Delta_y = -i\rho_{\uparrow\downarrow} + i\rho_{\downarrow\uparrow}$$ [4]

However, we must also include a pseudospin field in the $z$ direction which represents the difference between the electrostatic and disorder potentials in the top and bottom layers.

The calculation strategy follows a standard self-consistent field procedure. Given the density-matrix of the 2D system, we can evaluate the mean-field quasiparticle Hamiltonian. For the inter-layer tunneling part of the Hamiltonian we use the local-density-approximation outlined above. The electrostatic potential in each layer is calculated from the charge density in each of the 2D layers by solving the full 3D Poisson equation for the entire structure using an alternating direction implicit method with appropriate boundary conditions. The boundary conditions employed in this situation were hard wall boundaries on the top and sides of the simulation domain and Neumann boundaries at the points where current is injected and extracted. Given the mean field quasiparticle Hamiltonian and voltages in the leads, we can solve for the steady state density matrix of the two dimensional bilayer using a variant of the popular NEGF with a real-space basis (7) which correctly accounts for mode to mode transitions. In our simulations we set contacts at the left hand side of the top layer to $V_{TL} = -V$ and the contact on the right hand side of the top layer to be $V_{TR} = V$. The other two edges on the bottom are contacted; however, the potential in these contacts is allowed to float to their self-consistent field value. The density matrix obtained from the quantum transport kernel is updated at each state in the interaction process using the Broyden method (8) to accelerate self-consistency. The updated density is then fed back into the Poisson solver. The effective interlayer tunneling amplitudes are also updated and the loop proceeds until a desired level of self-consistency is achieved. The transport properties are calculated after self-consistency is achieved via the Landauer formula at finite temperature, i.e. using,

$$I(V_{sd}) = \frac{2e}{h} \int T(E) [f_s(E) - f_d(E)] dE$$ [4]

Path Integral Monte Carlo Results

We have run PIMC simulations on the quasiparticle model for bilayer graphene, with results and detailed discussion given in Reference (10). Here we focus on the calculated superfluid order parameter and an estimate of the transition temperature, $T_c$. For a system of $N$ bosons in a two dimensional box, the superfluid fraction is given by (4,10),

$$\frac{\rho_s}{\rho} = \frac{2mT}{n} \left\langle W^2 \right\rangle,$$ [5]
where $W$ is the topological winding number for the closed paths. For a system of two-dimensional excitons, there are four distinct winding numbers—$W_{e,x}$, $W_{e,y}$, $W_{h,x}$, $W_{h,y}$—for electrons and holes and for winding in the x and y directions. Since excitons are composite bosons made from a coupled electron and hole, the bosonic winding for Equation [5] is given by (10),

$$\langle \tilde{W}^2 \rangle = \frac{1}{4} \left( \langle W_{e,x} + W_{e,y} \rangle + \langle W_{h,x} + W_{h,y} \rangle \right).$$  \[6\]

We have run several quasiparticle PIMC simulations for the device structure shown in Figure 1 and calculated the superfluid densities shown in Figure 4. For analysis, we have fit this data to the functional form,

$$\frac{\rho_s}{\rho} = 1 - \left( \frac{T}{T_c} \right)^\alpha,$$  \[5\]

as shown by a solid line in Figure 4. From the fit, we find $T_c=850$ K and $\alpha=2.2$. While this result is encouraging, it is only suggests a room temperature condensate is possible in bilayer graphene. Our calculations have included all quantum and thermal fluctuations for a many-body system of interacting electrons and holes, but there are still a number of approximations that must be considered, which may reduce $T_c$. Specifically: (i) This result is for a finite size system. We expect a small decrease in $T_c$ with proper consideration of finite size scaling. (ii) We have neglected spin and valley degeneracy. By considering the effects of spin degeneracy on free bosons, we estimate a decrease in $T_c$ by a factor of two. Given our rather high estimate (>800K) from this PIMC calculation, we would still expect to see $T_c$ above room temperature. (iii) Our quasiparticle model neglects the chiral structure and linear dispersion of ideal graphene. This is biggest uncertainty in interpreting these PIMC calculations. (iv) We use a fixed-node approximation with excitonic nodes to handle the sign problem.

To summarize the PIMC results to-date, we have seen clear evidence for excitonic superfluidity in a model system that captures the dimensionality and geometry of the device, proper many-body interactions, full treatment of quantum and thermal fluctuations, and accounting for the quasiparticle velocities and carrier densities in experimental conditions. While a number of issues remain to be checked, our calculations suggest that condensation should be robust above room temperature, due to strong Coulomb correlations and excitonic pairing between the closely-spaced graphene layers.
Figure 4. Superfluid fraction, as predicted by PIMC simulations for an excitonic superfluid in bilayer graphene indicating room temperature superfluidity. PIMC data points are from Reference (10) and solid line fit is to Equation [7] with $T_c=850$ K and $\alpha=2.2$.

**Non-Equilibrium Results**

In Fig. 5, we now bias the gates at $V_{TG} = 0.4$ V and $V_{BG} = -0.4$ V and plot the differential interlayer conductance at room temperature. There are nevertheless pseudospin-dependent interaction contributions to the quasiparticle Hamiltonian. When a current flows, the pseudospin orientation of transport electrons is altered, just as in metal spintronics, and some of the same phenomena can occur. The resulting change in the quasiparticle Hamiltonian is responsible for a current-induced pseudospin-torque which alters the state of non-transport electrons well away from the Fermi energy. Indeed although the spin-splitting field appears spontaneously in ferromagnetic metals, spintronic phenomena usually depend on interplay with external fields due to magnetic-dipole interactions, spin-orbit coupling, and external magnetic fields. In graphene bilayers the pseudospin external field is due to the single-particle interlayer tunneling amplitude and is typically of the same order as the interaction contribution to the pseudospin-splitting field. At low interlayer bias, we find the differential conductance to be greater than 4 orders of magnitude greater than that at large interlayer. As we have a balanced situation where the magnitude of the total voltage drop between the two layers of $V_{TOT} = 0.8$ V, we are past the phase boundary and enter examine the transport of the interlayer indirectly bound excitons. Under these conditions, a gap opens up in the bandstructure around the Fermi energy and the injected electrons undergo reflections very similar to Andreev reflections seen in superconductors assuming that the pairing is of an s-wave nature. Nevertheless, while the transport across the layers in identically zero; there is still a very large interlayer current which decays as we increase the bias between the two layers. The reason for the decay of the differential tunneling conductance can be attributed to the pseudospin torque effect (11).

**Summary**

Here we have presented quantum transport calculations on electron-hole bilayer graphene systems using PIMC and recursive scattering matrices. We see the differential tunneling conductance has peaked behavior under low interlayer bias at room
temperature in bilayer graphene in stark contrast to the semiconducting case. Finally, we explain the decay in differential tunneling conductance as a manifestation of the pseudospin torque effect.

![Graph showing differential tunneling conductance versus interlayer voltage](image)

Figure 5. Our prediction of experimentally measurable differential tunneling conductance versus the applied interlayer voltage in the electron-hole bilayer.

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