Time Dependent Quantum Transport in Graphene

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Abstract—We present an efficient time-dependent NEGF method for simulation of dynamic through steady-state intra- and interband quasi-ballistic quantum transport in graphene using an atomistic-tight-binding Hamiltonian, novel alternating direction semi-implicit numerical time evolution schemes, and injecting and absorbing boundary conditions.

Graphene, quantum transport, time dependence, NEGF

I. INTRODUCTION

Graphene is being considered for many purposes because of its novel properties. Although the lack of a band gap continues to challenge CMOS-like logic applications, high carrier velocities, the ultimate ultrathin body and potential process compatibility with silicon technologies still make graphene a promising candidate for radio frequency (RF) applications [1,2] and perhaps for novel "beyond CMOS" applications [3]. For such devices, however, one must go beyond quasi-static to truly time-dependent analysis to fully understand their intrinsic frequency limitations and dynamic response. Towards this goal, we are developing a timedependent non-equilibrium Green's function (NEGF) for modeling dynamic quantum transport in graphene. Here we describe the essential elements of the method-Hamiltonian, time evolution scheme and open boundary conditions-and illustrate them via simple MATLAB-based [4] simulations for clarity. With these elements given, future, e.g., electrostatically self-consistent simulation of graphene MOSFETs with a thermal distribution of source-injected carriers will add only to the computational burden, not the technical one.

The challenges of simulating time-dependent transport in graphene include fast (1 nm/fs), quasi-non-dispersive, and multi-band transport—the latter because band-to-band tunneling is a critical physical consideration for essentially all proposed graphene devices. To address these challenges, a π -orbital-based nearest-neighbor atomistic-tight-binding Hamiltonian model of graphene is used. Novel variations of an alternating-direction semi-implicit scheme are employed to solve the time-dependent Schrödinger equation on the hexagonal lattice, to maintain stability, conserve probability, and achieve computational efficiency. Potentially time dependent source terms and stationary but non-local self-energy terms are added to allow transport *through* the simulation region.

I. SOLUTIONS OF TIME-DEPENDENT SCHRODINGER EQ.

Within the tight-binding formalism, the time-dependent



Figure 1. Illustration of the graphene crystal lattice, represented by the points where the colors distinguish the sublattice, and the nearest tightbinding coupling of the tight-binding Hamiltonian, represented by the lines (solid or dashed, black or gray). For one implementation of an alternating direction implicit scheme, ADI1 (see Section III), the associated four-atom unit cell is shown in the rectangle. In this case, coupling between atoms within the same unit cell are indicated by the black solid lines, coupling between unit cells in the *x* direction by black dashed lines and coupling between unit cells in *y* direction by dashed gray lines.

Schrödinger equation is of the form,

$$i\hbar\frac{d}{dt}\boldsymbol{\Psi} = \mathbf{H}\boldsymbol{\Psi}\,,\tag{1}$$

where ψ and **H** are the wave-function column matrix and Hamiltonian square matrix, respectively. Here we consider on-site and nearest-neighbor π -bonding only on the twodimensional (2D) hexagonal lattice of graphene (Fig. 1), with nearest-nearest neighbor matrix elements $H_{i,i} = \tau_0 = -3.03$ eV, on-site matrix elements $H_{i,i}$ —defined by an electrostatic potential, e.g.—and all other matrix elements set to zero.

To numerically solve (1), we first consider discretization of time via the well-proven semi-implicit Crank-Nicolson (CN) method [5]. This approach provides accurate, unitary and time reversible evolution and serves as a reference here. Assuming a time step of Δt for time integration, the CN is

$$i\hbar \frac{\Psi(t+\Delta t)-\Psi(t)}{\Delta t} = \mathbf{H}\left(t+\frac{\Delta t}{2}\right)\left(\frac{\Psi(t+\Delta t)+\Psi(t)}{2}\right).$$
 (2)

Denoting $\psi^n = \psi(n\Delta t)$ and $\mathbf{H}^{n+1/2} = \mathbf{H}(n\Delta t + \Delta t/2)$, Eq. (2) can be rewritten for each time step as,

$$\left(\mathbf{I}+i\Delta t\mathbf{H}^{n+1/2}\right)\boldsymbol{\psi}^{n+1}=\left(\mathbf{I}-i\Delta t\mathbf{H}^{n+1/2}\right)\boldsymbol{\psi}^{n}.$$
(3)

As an illustration of time evolution, we consider a Gaussian wave-packet centered around one of the Dirac points \mathbf{k}_D in wave-vector space and initially well-localized in real-space, as per Fig. 2(a),

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Figure 2. Snapshots of time evolution of initially Gaussian wave-packet at (a) t = 0 and (b) t=14 fs.

$$\psi(\mathbf{r},t=0) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\left|\mathbf{r}-\mathbf{r}_o\right|^2 / 2\sigma^2 + i\mathbf{k}_D \begin{pmatrix} 1\\ i \end{pmatrix}\right). \tag{4}$$

The two-element column matrix is the so-called "pseudo-spin" matrix describing the relation between the complex amplitude coefficients of the two sub-lattices of graphene. While fixed initially here, in general it can be a function of **r** as well. This wave-packet contains equal contributions from the conduction and valence band, and extends well into both. Due to the predominately linear dispersion/constant carrier speed in graphene within the relevant energy range, the wave-packet spreads from the center in a asymmetric ring-like shape with little change in the ring thickness with time, as shown in Fig. 2(b) at t = 14 fs. The principle direction of motion is associated with the given initial pseudo-spin phase. This behavior, confirmed by analytic results for this simple system [6], contrasts markedly to the broadening-Gaussian-evolution characteristic of particles in a single band of well-defined mass.

Accurate simulation, however, requires a quite small time step. For an explicit time-evolution scheme, the fixed carrier speed of 1 nm/fs and nearest-neighbor inter-atomic spacing on the order of 0.1 nm would suggest the use of a time step less than or equal to about 0.1 fs simply to track a wave front. In practice, we have also found this estimate to be appropriate for our half-implicit scheme(s). As shown in Fig. 3, the use of a 1.0 fs time step results in slowed and qualitatively inaccurate motion of the wave-packet by comparison to the result for a 0.1 fs time step. Note that in the latter case (Fig. 3(a)) the leading edge of the wave-packet travels about 3 nm in 3 fs as expected.

With the basic CN method, the computational cost per time step increases super-linearly with size of the considered simulation region due to the far-from-diagonal nonzero elements of the Hamiltonian required in (3). To reduce the computational effort, we introduce two split operator alternating-direction implicit (ADI) methods.



Figure 3. Snapshot of a time evolved initially Gaussian wave-packet on Graphene at 3 fs for a time step of (a) 0.1 fs and (b) 1 fs.

In the first approach, ADI1, we split the Hamiltonian operator **H** into quasi-rectangular coordinates with \mathbf{H}_x and \mathbf{H}_y , and the time step in half such that (2) becomes,

$$\begin{pmatrix} \mathbf{I} + i\frac{\Delta t}{2}\mathbf{H}_{x}^{n+1/2} \end{pmatrix} \mathbf{\psi}^{n+1/2} = \begin{pmatrix} \mathbf{I} - i\frac{\Delta t}{2}\mathbf{H}_{y}^{n+1/2} \end{pmatrix} \mathbf{\psi}^{n}$$

$$\begin{pmatrix} \mathbf{I} + i\frac{\Delta t}{2}\mathbf{H}_{y}^{n+1/2} \end{pmatrix} \mathbf{\psi}^{n+1} = \begin{pmatrix} \mathbf{I} - i\frac{\Delta t}{2}\mathbf{H}_{x}^{n+1/2} \end{pmatrix} \mathbf{\psi}^{n+1/2} .$$

$$(5)$$

Hopping in the nominal v direction is fully explicit and hopping in nominal x direction is fully implicit during the first half of the time-step, and vice-versa during the second half. However, there is more than one way to perform such a quasirectangular splitting of the Hamiltonian. We obtain \mathbf{H}_{x} and \mathbf{H}_{y} by considering the rectangular unit cell shown in Fig. 1. Coupling between atoms within the unit cell (solid black lines) is considered in both \mathbf{H}_x and \mathbf{H}_y but at half-strength $\frac{1}{2}t_o$. Coupling between unit cells in the x direction (dashed black lines) is considered only in \mathbf{H}_x but at full strength t_0 . And coupling between unit cells in the y direction (dashed gray lines) is considered only in \mathbf{H}_{v} but again at full strength t_{o} . Any on-site contributions to \mathbf{H} are split evenly between \mathbf{H}_x and \mathbf{H}_{v} . This operator splitting leads to pentadiagonal and tridiagonal matrix equations in the first and second halves of the time step, respectively.

Another approach we use to split the Hamiltonian, ADI2, follows the natural rotational symmetry of the graphene lattice. We subdivide the Hamiltonian as $\mathbf{H} = \mathbf{H}_1 + \mathbf{H}_2 + \mathbf{H}_3$. Each of the three components Hamiltonians \mathbf{H}_i are constructed by ignoring all bonds in the corresponding direction δ_i shown in Fig1, as illustrated in Fig. 4. Since each bond appears in two of the \mathbf{H}_i , their strength is halved therein, to $\frac{1}{2}t_o$ again. The onsite coupling is split equally among the three \mathbf{H}_i . The time step is now split into three sub-steps as,

$$\begin{pmatrix} \mathbf{I} + i\frac{dt}{3}\eta\mathbf{H}_1 \end{pmatrix} \boldsymbol{\psi}^{n+1/3} = \begin{pmatrix} \mathbf{I} - i\frac{dt}{3}(\mathbf{H} - \eta\mathbf{H}_1) \end{pmatrix} \boldsymbol{\psi}^n$$

$$\begin{pmatrix} \mathbf{I} + i\frac{dt}{3}\eta\mathbf{H}_2 \end{pmatrix} \boldsymbol{\psi}^{n+2/3} = \begin{pmatrix} \mathbf{I} - i\frac{dt}{3}(\mathbf{H} - \eta\mathbf{H}_2) \end{pmatrix} \boldsymbol{\psi}^{n+1/3} \cdot$$

$$\begin{pmatrix} \mathbf{I} + i\frac{dt}{3}\eta\mathbf{H}_3 \end{pmatrix} \boldsymbol{\psi}^{n+1} = \begin{pmatrix} \mathbf{I} - i\frac{dt}{3}(\mathbf{H} - \eta\mathbf{H}_3) \end{pmatrix} \boldsymbol{\psi}^{n+2/3}$$

$$\begin{pmatrix} \mathbf{I} + i\frac{dt}{3}\eta\mathbf{H}_3 \end{pmatrix} \boldsymbol{\psi}^{n+1} = \begin{pmatrix} \mathbf{I} - i\frac{dt}{3}(\mathbf{H} - \eta\mathbf{H}_3) \end{pmatrix} \boldsymbol{\psi}^{n+2/3}$$

The parameter η is set to 3/2 so that the method remains normconserving half-implicit. (I.e., such that $\eta(\mathbf{H}_1 + \mathbf{H}_2 + \mathbf{H}_3)$ from the left-hand/implicit side(s) of (6) equals $3\mathbf{H} - \eta(\mathbf{H}_1 + \mathbf{H}_2 + \mathbf{H}_3)$ from the right-hand/explicit sides(s).)

Figs. 5(a)-(c) illustrate the accuracy of these ADI methods, showing essentially identical snapshots obtained at 3 fs from an initially Gaussian wave-packet using the reference non-ADI, ADI1, and ADI2 methods, respectively. Fig. 5(d) shows the computational effort for the three different methods as a



Figure 4. Illustration of the $\mathbf{H} = \mathbf{H}_1 + \mathbf{H}_2 + \mathbf{H}_3$ split Hamiltonian of the ADI2 method.



Figure 5. Snapshot of an initially Gaussian wave-packet with a pseudospin angle of 60° at 3fs, obtained using (a) non-ADI (b) ADI1 (c) ADI2 methods, and (d) computational time per time step for the nonADI (black), ADI1 (dashed red), and ADI2 (dash-dot blue) methods as a function of simulation region size.

function of the number of atoms in the simulation region for an ~20 nm wide graphene ribbon of varying lengths. Both ADI methods exhibit only linear growth in simulation time with simulation region size, in contrast to the super-linear growth with the non-ADI method. (That the effort required does not diverge earlier is a testament to the UMFPACK matrix solver used in MATLAB). While the ADI1 method requires slightly less effort per time step, a more detailed analysis finds the ADI2 method to be slightly more accurate for a given time step. The differences are small enough that convenience may be the best determinant of which to use.

II. ABSORBING AND INJECTING BOUNDARY CONDITIONS

For the sake of the current discussion, consider a central (C) simulation region *through* which we wish to consider dynamic probability/charge current flow, coupled to "open" left (L) and right (R) "leads," where the wave-function in each region is ψ_C , ψ_L , and ψ_R , respectively.

Consider injection first, from the left lead for specificity. The total wave-function in the left lead can then be written as $\Psi_L = \Psi_L^{in} + \Psi_L^r$ where Ψ_L^{in} is the assumed-given incident wave-function whose time evolution in the lead is known, and Ψ_L^r is any reflected wave. The time-dependent Schrödinger's equation is transformed into the inhomogeneous/NEGF form,

$$i\hbar \frac{d}{dt} \begin{bmatrix} \boldsymbol{\Psi}_{L}^{r} \\ \boldsymbol{\Psi}_{C} \\ \boldsymbol{\Psi}_{R} \end{bmatrix} = \begin{bmatrix} \mathbf{H}_{L} & \mathbf{H}_{LC} & \mathbf{0} \\ \mathbf{H}_{CL} & \mathbf{H}_{C} & \mathbf{H}_{CR} \\ \mathbf{0} & \mathbf{H}_{RC} & \mathbf{H}_{R} \end{bmatrix} \begin{bmatrix} \boldsymbol{\Psi}_{L}^{r} \\ \boldsymbol{\Psi}_{C} \\ \boldsymbol{\Psi}_{R} \end{bmatrix} + \boldsymbol{\Psi}_{S} .$$
(7)

Here, \mathbf{H}_C , \mathbf{H}_L , and \mathbf{H}_R are the Hamiltonians of the isolated central, left and right regions, respectively. The off-diagonal elements represent the coupling between the adjacent regions. The probability source term $\boldsymbol{\psi}_s$ on right-hand-side is given by

$$\boldsymbol{\Psi}_{S} = \begin{pmatrix} \left(\mathbf{H}_{L} - i\hbar \frac{d}{dt} \right) \boldsymbol{\Psi}_{L}^{in} \\ \mathbf{H}_{LC} \boldsymbol{\Psi}_{L}^{in} \\ \mathbf{0} \end{pmatrix}.$$
(8)

Consider next absorption by the leads. In principle, one could derive boundary self-energies based on quantum transmitting boundary conditions (OTBCs) as for timeindependent NEGF. However, non-stationary OTBCs are required for the time-dependent system. In principle, creation of these requires keeping track of, and integrating over at each time step, the past history of the wave-function at the boundary, or at least a significant period thereof for a reasonable approximation. It may be possible to provide nonstationary but at least local-in-time approximate QTBCs via extrapolation of the time-dependent wave-function within the central region to (just) across the boundary, based on assumptions about its approximate form (such as in [7] perhaps aided by a transverse mode expansion). However, multi-band transport and quasi-non-dispersive transport, such that abrupt variations in the wave-function induced far from the lead boundary may remain at the boundary, make this latter approach to QTBCs challenging.

For these reasons, for simplicity, and for flexibility, we employ stationary but spatially non-local position-dependent absorbing complex potentials—self-energies—within leads of finite length, such as used in electromagnetics [8]. To be effective, however, there are some basic requirements that the combination of complex potential and lead length must meet. To avoid reflection from the end of the finite leads, the average complex potential within the lead must be sufficiently large to completely absorb any injected—and very fast in graphene—wave-function before it can reach the end of the lead and reflect all the way back to the simulation region. However, the rate of change of the complex potential with position, particularly near the boundary between the central region and the lead, cannot be so fast as to cause back reflection in and of itself.

To first illustrate the absorption boundary conditions only, the time evolution of an initially predominately right-directed Gaussian wave-packet out of an approximately (~) 21 nm wide by ~19 nm long section of graphene is simulated. We consider three scenarios for the right lead, as illustrated in Fig 6(a): an ~85 nm long lead region with no complex absorbing potential (NOCAP); the same lead region with an added complex absorbing potential (CAP) linearly ramped from zero at the boundary to the central region up to a purely imaginary 15 meV at its right-side hard-wall boundary; and an ~170 nm long lead region with no complex absorbing potential (NOCAP-L). (With the predominately right-directed wave, the left lead is essentially of no consequence). Figures 6(b)-(d) show the probability density within the simulation region as a function of time for the non-ADI, ADI1 and ADI2, methods. The reflection from the lead end back into the simulation region in the NOCAP case is clear. In the NOCAP-L case, the simulation region is effectively a perfect infinite lead within



Figure 6. (a) Geometries used for CAP, NOCAP and (effectively infinite lead) NOCAP-L simulations as described in text. Total probability function density in the central region as a function of time for simulation with (b) Non ADI, (c) ADI 1 and, (d) ADI 2 methods, for NOCAP (solid black) and CAP (solid red) and NOCAP-L (solid blue) simulations.

the considered 250 fs simulation period as there is no time for the wave-function of maximum velocity 1 nm/fs to traverse back and forth across the lead region, which makes the associated result the reference ideal result. The agreement between the CAP and NOCAP results, therefore, demonstrates the accuracy of the absorbing potential approach. However, although accurate here, the combination of absorbing potential and channel length has not yet been fully optimized to minimize the latter and, thus, the computational burden. Finally, we note that the required length of the leads is independent of the size of the central region, so the relative computational burden will decreases with larger central regions that will be required for device simulation.

To illustrate injection along with the absorption, we consider transient through steady-state transport through an \sim 21 nm wide metallic armchair graphene ribbon, with an \sim 23 nm long central region. ψ_{I}^{in} nominally corresponds to a monoenergetic 95 meV electron relative to the Dirac point within the metallic subband. However, the amplitude of the incident wave-function is ramped exponentially toward saturation with a 20 fs time constant, so it is not truly mono-energetic, at least prior to saturation. Also, ψ_L^{in} was defined as nonzero only within the "slice"-corresponding to the thickness of the gray rectangle along the x direction in Fig. 1—of the left lead immediately adjacent to the central region, which is all that is necessary. As shown in Fig. 7, near-steady-state conditions are achieved within the central region well within the 140 fs simulation period. Moreover, there is no apparent reflection back from the right lead at any time, as would be readily observable via a standing wave pattern. (To show more clearly the evanescing of the probability with position in the lead, the length of the right lead was extended beyond what was necessary; again, the probability need only be absorbed before traversing both ways across the length of the lead.) The results of Fig. 7(b) also evidence the continued effectiveness of the proposed ADI methods.



Figure 7. (a) Snapshots of probability density as function of position within the central region and a long right absorbing lead over 140 fs with the non-ADI implementation, with a left-injected nominally propagating eigen-mode of the ribbon, although ramped up exponentially toward steady-state with a 20 fs time constant, serving as a source term in the left lead. (b) A subset of the results of (a) but using non-ADI, ADI1 and ADI2 methods with essentially indistinguishable results.

Finally, we wish to point out that, although discussed and illustrated above in terms of a central simulation region and left and right leads, the Hamiltonian is not actually subdivided by region and source terms and, *separately*, absorbing regions could be placed anywhere within a plane of graphene as needed to simulate a particular system of interest.

III. CONCLUSION

We have presented a framework for an efficient timedependent NEGF method for simulation of dynamic through steady-state quasi-ballistic intra- and inter-band quantum transport in graphene using an atomistic-tight-binding Hamiltonian, novel alternating direction semi-implicit numerical time-evolution schemes, and injecting and absorbing boundary conditions. The essential elements of this framework were demonstrated via illustrative simulations. Extension to multi-graphene-layer systems should also be possible.

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