Multiphysics simulations of carrier transport and electrodynamics in two-dimensional electron systems

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Abstract

We present a multiphysics numerical technique and use it for simulating carrier dynamics under electromagnetic excitation in supported two-dimensional electronic systems. The technique combines ensemble Monte Carlo (EMC) for carrier transport with finite-difference time-domain (FDTD) for electrodynamics and molecular dynamics (MD) for short-range Coulomb interactions among particles. We explain the important criteria required for coupling the constituent methods and demonstrate the use of this coupled EMC/FDTD/MD technique by calculating the room-temperature *dc* and *ac* conductivity of graphene supported on SiO_2.

1. Introduction

Multiphysics techniques involve simulations of different physical phenomena unified with a common modeling strategy in order to examine complex coupled behavior. For example, the non-Drude-like high-frequency conductivity of doped semiconductors can be calculated by tracking the carrier dynamics coupled with electromagnetic wave propagation and particle-particle Coulomb interactions in time and space [1]. In recent years, several applications have been proposed and developed [2], [3] that rely on the coupled effects of electromagnetic waves interacting with carriers in supported two-dimensional (2D) materials, such as single-layer graphene [4] or MoS_2 [5], and quasi-2D materials, such as semiconductor membranes [6]. Realizing these applications requires an understanding of carrier transport in 2D materials in the presence of electromagnetic fields, while accounting for the strong influence of the supporting substrate and the impurities found near the interface between the substrate and the 2D electron system (2DES). A multiphysics numerical solver that combines ensemble Monte Carlo (EMC) simulation of carrier transport with the finite-difference time-domain (FDTD) technique for solving Maxwell's curl equations and molecular dynamics (MD) modeling of short-range particle interactions [1], [7] can provide insight into the carrier transport and electrodynamics of 2DESs.

We present the EMC/FDTD/MD technique as applied to simulating carrier transport in 2DESs. We consider a structure with a single graphene layer resting on top of an SiO_2 substrate, with impurity ions near the interface, as shown in Figure 1. We describe the constituent techniques and the procedure for coupling them and give examples of the *dc* and *ac* conductivity of supported graphene in the presence of charged impurities, as calculated using EMC/FDTD/MD.

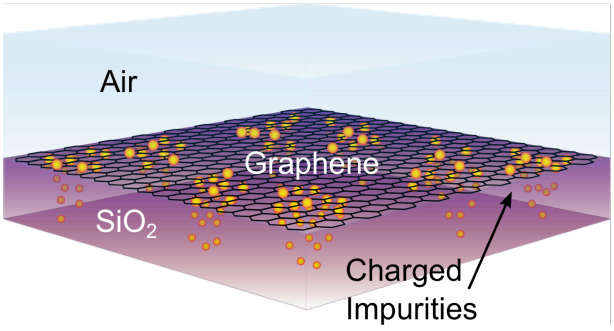

Figure 1. Schematic of the simulated structure: single-layer graphene rests on an SiO_2 substrate, with charged impurities present near the interface between the two. Carrier transport is simulated by including both electrons and holes in the graphene layer, while the positively charged ions near the interface and within the SiO_2 substrate remain stationary.

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2. The Constituent Techniques

The EMC method is used to simulate carrier dynamics in the diffusive transport regime by tracking the evolution of a large ensemble of carriers [typically $O(10^5)$] over time. This method yields a solution to the Boltzmann transport equation by using statistically appropriate stochastic sampling of the relevant carrier relaxation mechanisms, free-flight times, and angular distributions of momenta [8]. The FDTD method [9] is a grid-based technique commonly used for electrodynamics simulations. The time-dependent Maxwell's curl equations are discretized using a centered-difference scheme for the partial derivatives in both space and time on a staggered uncollocated space grid. The discretized equations are solved via leapfrog time integration with the electric and magnetic fields staggered in time, yielding a fully explicit scheme for evolving the spatial distribution of the electric and magnetic fields over time. MD is used to simulate short-range interactions in classical many-particle systems [10]. For a collection of electrons, holes, and charged ions, the direct and exchange Coulomb forces among carriers (electrons and holes), and the direct Coulomb forces between carriers and ions present within a small volume (3×3×3 grid cells) are calculated using MD. The carriers in the EMC module drift under the action of the fields and scatter according to the appropriate scattering mechanisms resulting in a current density that can be calculated from carrier velocities. The positions of the carriers also change, leading to different short-range electrostatic interactions. The current densities and the new carrier positions are used to adjust the fields acting on the carriers in the FDTD and MD modules thus coupling these with the EMC module.

3. Coupled EMC/FDTD/MD

Accurate and stable coupling of the EMC, FDTD, and MD methods requires several criteria to be satisfied. The EMC method requires the time step size (Δt) to be much smaller than the relaxation time of the carriers. FDTD also constrains the size of Δt via the Courant stability condition [9]. To correctly represent fields arising from the charge densities in FDTD, the initial field distribution must satisfy Gauss's law and the continuity equation must be enforced at each time-step [1]. Therefore, the initial field distribution is calculated from the solution of Poisson's equation. The use of grid-based methods necessitate that the charges be assigned to the grid, which is achieved via the cloud-in-cell (CIC) method [11]. The continuity equation is enforced thereafter by using the same CIC scheme for calculating the current density sourcing the FDTD fields. Moreover, the fields at the location of a carrier that is found inside a given grid cell are determined by first averaging the FDTD fields on the grid faces and grid lines surrounding it in order to calculate the fields at the grid points. Then, from the fields at each grid point, the same CIC weights of each carrier are used to interpolate the fields and obtain the values at the carrier's actual location. Adding the grid-based FDTD and short-range MD fields, and then subtracting the short-range contribution of the grid-based fields yields the accurate total fields acting on the carriers without double counting the contribution from FDTD and MD. However, calculating the short-range contribution of the grid-based fields is a complicated process for layered structures with different material interfaces.

4. Numerical Constraints

Properly initializing the grid cell size in the computational domain is crucial for ensuring computational efficiency and accuracy of the EMC/FDTD/MD method. Unfortunately, the accuracy and computational efficiency constraints of FDTD are opposite to those of MD. For example, an FDTD simulation with larger Δx yields a faster but less accurate result, while with MD, it yields a more accurate calculation of the intra-grid-cell fields but is also computationally more intensive due to more number of particles per grid cell (for a fixed charge density). The computational burden in MD scales as N^2, where N is the number of particles involved.

To avoid double counting fields from FDTD and MD, and to reduce the computational burden of calculating the total forces on charged particles, we pre-calculate reference short-range fields for MD and also the grid-based short-range field contribution. The short-range grid-based field contribution is calculated by solving the Poisson's equation for a unit charge placed on a grid point near the center of the domain. Utilizing the linearity of the solution the fields at the neighboring grid points are calculated by shifting the original solution. The fields from all the grid points in a cell are finally combined using the CIC weights to obtain the total short-range fields for an ion in any location within the cell. The Poisson's equation has to be solved once for each material and the shifted fields are also weighted according to the material in order to represent the discontinuity across interfaces [7]. This procedure of precalculating the short-range component of grid-based fields needs to be done only once at the initialization stage.

A simulation with an an appropriate incident wave source condition is run until a steady state is reached, which is indicated by a saturation of the ensemble averaged electron energy and velocity. A typical EMC/FDTD/MD simulation of graphene on SiO_2 with a carrier density of 5×10^{12} cm^{-2}, impurity ion density of 5×10^{11} cm^{-2}, and a 500-

GHz sinusoidal excitation takes about 16-17 hours to run on a desktop PC with an Intel® Core i7™ 2.8 GHz CPU and 8 GB of RAM.

5. Application of EMC/FDTD/MD: Calculating the Conductivity of Graphene on SiO_2

Figure 2 shows the results of applying the EMC/FDTD/MD technique to calculate the *dc* and *ac* conductivity of graphene for an impurity sheet density of 5×10^{11} cm^{-2} with a uniform random distribution (blue squares) and a clustered distribution (red diamonds; correlation length of 40 nm). We compare these results with the conductivity of impurity-free graphene (black circles), also computed using EMC/FDTD/MD. The *dc* conductivity results (Figure 2a) reproduce important features of the conductivity vs. carrier density curve observed in experiment [12]. The curve displays a sublinear increase at high carrier densities (>4×10^{12} cm^{-2}) for "clean" graphene. Moreover, for a given sheet density of charged impurities, the conductivity for the clustered impurity distribution is lower than that for the uniform random distribution. The clustered impurity-conductivity behavior has also been observed in experiment [13] and predicted in the calculations of carrier-impurity scattering rates with a structure factor describing correlations [14]. Our EMC/FDTD/MD simulation makes no assumptions about the screening length or structure factor, and uses real-space impurity positions and the corresponding carrier-impurity interactions to calculate the conductivity. Our results also show a flattening of the conductivity curve near the Dirac point for clustered impurity distributions, similar to that observed in conductivity measurements involving intentional potassium doping [12].

The frequency-dependent *ac* conductivity, shown in Figure 2b for a carrier density of 3×10^{12} cm^{-2}, is computed by varying the frequency of the source excitation from 500 GHz to 13 THz. In this range, carrier transport is dominated by intraband processes and is captured very well in our simulation. These results are in line with experimental measurements of frequency-dependent conductivity [15]. For frequencies greater than 4 THz, our results show that the total impurity density and distribution do not affect the conductivity of graphene. However, for lower frequencies (<4 THz), there is a significant dependence of conductivity on the impurity density and distribution. (As expected, the low-frequency conductivity limit obtained from *ac* calculations is very close to the values calculated in the *dc* simulations.)

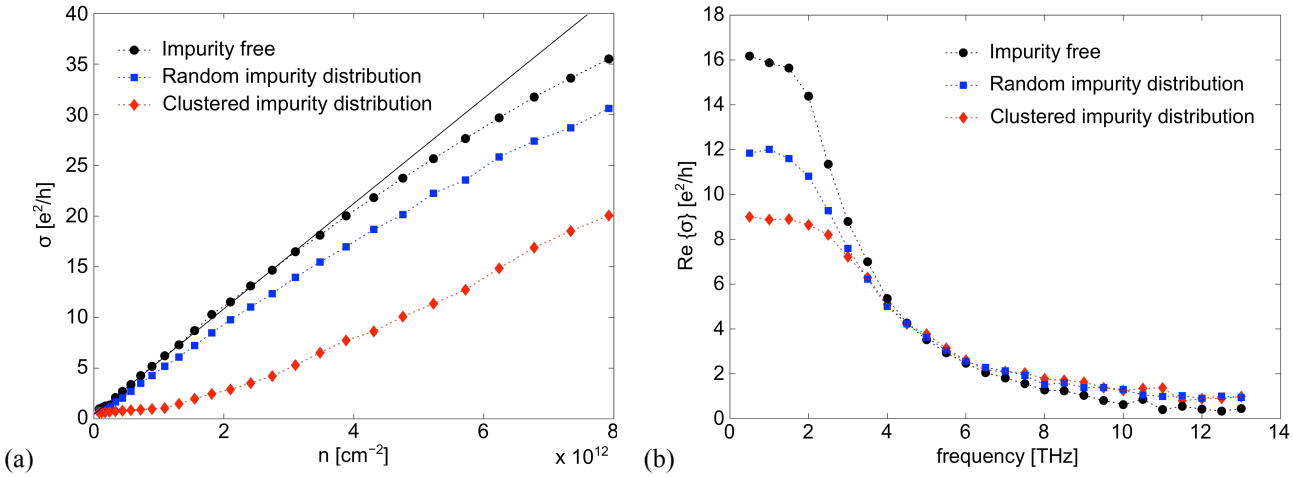

Figure 2: (a) *dc* conductivity of graphene supported on SiO_2 as a function of the carrier density. Black circles denote the results for the impurity-free case, blue diamonds for a uniform random distribution, and red diamonds for a clustered distribution (40 nm average cluster size) of charged impurities with a sheet density of 5×10^{11} cm^{-2}. The black line is a linear fit to the low-density (<3×10^{12} cm^{-2}) part of the impurity-free curve to indicate the sublinearity in the data. (b) Frequency-dependent *ac* conductivity of graphene on SiO_2 for the same charged impurity distribution as in panel (a) with a carrier density of 3×10^{12} cm^{-2}.

6. Conclusion

We have presented the implementation and application of the coupled EMC/FDTD/MD simulation technique to carrier transport in supported graphene. We have described the constituent techniques, as well as the important steps for their self-consistent coupling. The general implementation can also be applied to transport simulations of other 2D or quasi-2D materials. We have demonstrated the use of the EMC/FDTD/MD method by calculating the room temperature

dc and *ac* conductivity of graphene supported on SiO_2.The EMC/FDTD/MD multiphysics simulation method is a versatile tool that can be used for characterizing high frequency electronic properties of materials and also simulating THz devices.

7. Acknowledgments

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