Effects of charged impurity clusters on the conductivity of supported graphene

N. Sule,* K. J. Willis,† S. C. Hagness,* and I. Knezevic*
*University of Wisconsin Madison, Madison, WI 53706 USA
†AWR Corporation, 11520 North Port Washington Road, Mequon, WI 53092, USA
e-mail: sule@wisc.edu

Charged impurities can be introduced into graphene samples by typical fabrication and processing techniques, and can persist even after annealing [1]. Recent imaging experiments using scanning tunneling microscopy show electron-hole puddles in graphene with an average size of $20\ \mathrm{nm}$ [2], and point towards a clustered impurity distribution in graphene samples. It is evident that clustered or spatially correlated impurities play an important role in determining the conductivity of graphene [3], [4]. A reliable and quantitative determination of the effects of such impurities on carrier transport in graphene supported on SiO_2 is important for the design of graphene-based applications. We have combined particle-based transport simulations using the ensemble Monte Carlo (EMC) method, with numerical long-range and short-range field solvers using the finite-difference time-domain (FDTD) technique and molecular dynamics (MD), respectively, to simulate the formation of electron-hole puddles, and study the effect of charged impurity distributions on the conductivity of graphene supported on SiO_2. The coupled EMC-FDTD-MD algorithm has been used to calculate the high-frequency conductivity of bulk silicon [5], [6] with very good agreement to experimental data.

The simulated structure, shown in Figure 1, consists of a monolayer of graphene on top of an SiO_2 substrate. Clusters of impurity ions are present at and near the interface between graphene and the substrate. We simulate an infinite graphene monolayer by using periodic boundary conditions on the fields and the carrier momentum. Doing so eliminates any edge-effects, thus focusing on the effects of impurities on carrier transport. The charged impurity clusters are stochastically initialized using a correlation length parameter to define the average size and distribution of the clusters.

By simulating carrier dynamics without any external fields, we calculate the steady-state electron and hole density distributions for a uniform random, as well as a clustered impurity distribution (Figs. 2a and b, respectively). The average size of the electron-hole puddles, calculated using the full width at half maximum (FWHM) of the normalized spatial auto-correlation functions of the density distributions, shown in Figs. 2c and d, are about $4\ \mathrm{nm}$ and $20\ \mathrm{nm}$ for the uniform random and clustered impurity distributions, respectively. Figure 2e shows a plot of the average electron-hole puddle size as a function of the average impurity cluster size. Thus, we show that impurity clusters between the sizes of 30 and $40\ \mathrm{nm}$ are responsible for producing electron-hole puddles quantitatively similar to those seen in experiments [2].

We calculate the conductivity in graphene as a function of the carrier density, under *dc* excitation for impurity-free, as well as for uniform random and clustered impurity distributions, with an impurity density of 10^{11}–$10^{12}\ \mathrm{cm}^{-2}$, and average cluster size of $36\ \mathrm{nm}$ (Figs. 3a–d). It is seen that, for impurity densities greater than $10^{11}\ \mathrm{cm}^{-2}$, the distribution of impurities (random or clustered) significantly affects both the sublinearity and the slope of the linear region in the carrier-density dependence of conductivity. By turning off/on specific terms in the MD to elucidate the effect of short-range carrier-carrier and carrier-ion interactions, we find that the sublinearity in conductivity as a function of carrier density is due to direct and exchange carrier-carrier interactions limiting transport. Moreover, the slope of the linear region is strongly dependent on the short-range carrier-ion Coulomb interaction, and therefore on the density and distribution of the impurities.

In conclusion, we show that clustered impurities, as well as the resulting short-range carrier-ion, and carrier-carrier interactions play an important role in determining the room-temperature conductivity of supported graphene.

References

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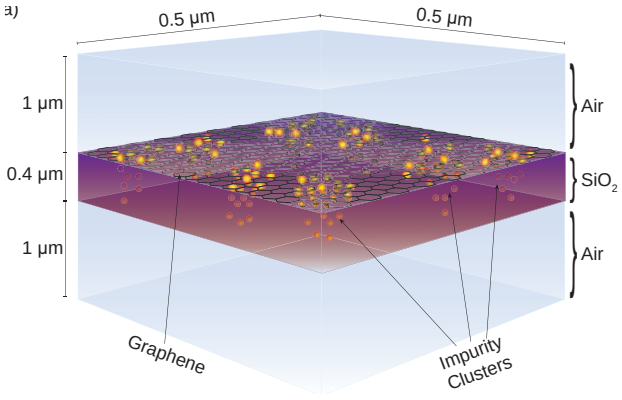

Fig. 1. A schematic of the simulated structure showing the graphene, SiO_2 substrate, and air layers, as well as impurity clusters between, and below the interface of graphene and the substrate.

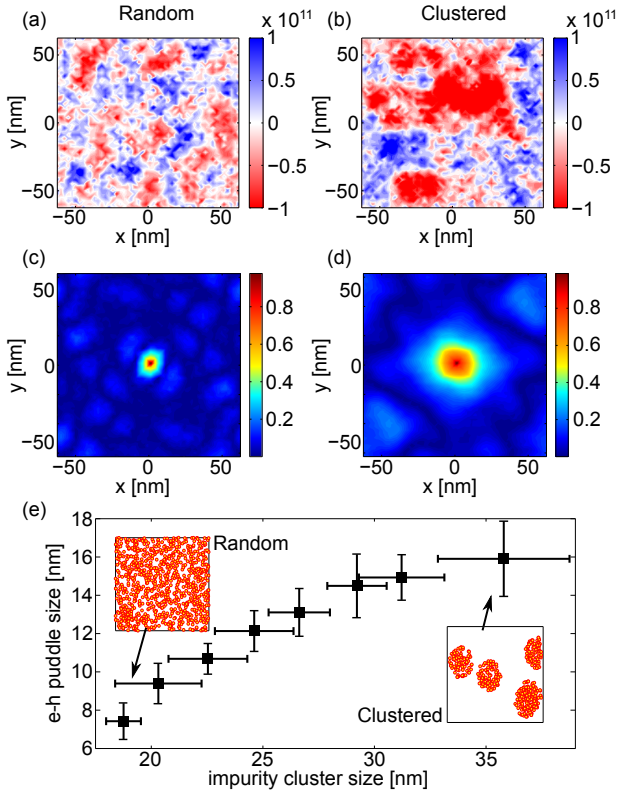

Fig. 2. Carrier-density distribution depicting the electron-holes puddles formed in graphene for (a) uniform random and (b) clustered impurity distributions with impurity density equal to 8×10^{11} cm^{-2}. The average size of the electron-hole puddles is estimated from the FWHM (yellow ring) of the normalized spatial auto-correlation function (SACF), is shown in (c) and (d), corresponding to the (a) random and (b) clustered impurity distributions, respectively. The estimated puddle size from (c) is 4 nm and that from (d) is 20 nm. (e) Plot of the mean electron-hole puddle size as a function of the mean impurity cluster size. Each data point in this plot is an average of 14 simulation runs and the error bars on data points denote the standard deviations.

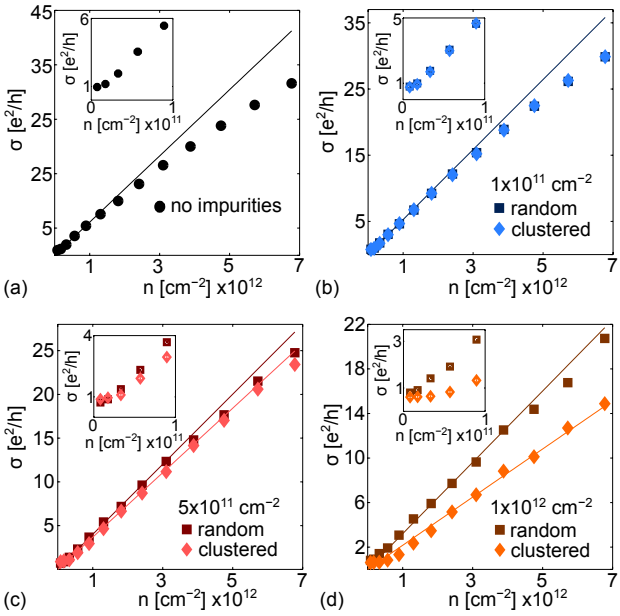

Fig. 3. Conductivity of graphene supported on an SiO_2 substrate as a function of the carrier-density for (a) impurity-free graphene, and for an impurity density of (b) 10^{11} cm^{-2}, (c) 5×10^{11} cm^{-2}, and (d) 10^{12} cm^{-2}. The square and diamond markers denote results for a uniform random and a clustered distribution of impurities, respectively. The insets in all of the above panels show a deviation from the linear decrease in conductivity at very low carrier densities ($< 10^{11}$ cm^{-2}), with the conductivity saturating near the value of e^2/h.