Intrinsic mobility in graphene

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Abstract
Recent studies have shown that a high K dielectric solvent screens the impurities for room
temperature transport in graphene and the mobility has been found to increase by orders of
magnitude. This gives what is probably the intrinsic, phonon limited mobility at room
temperature, and we have confirmed this with simulation. Mobility as high as
$44\,000$ cm^2 V^{-1} s^{-1} was achieved. At very low density, impurity scattering still is the
determining factor for mobility, but this is significantly reduced in the recent experiments due to
the dielectric screening. At high density, impurity scattering becomes negligible.

(Some figures in this article are in colour only in the electronic version)

Graphene has been the subject of a considerable level of interest in the past few years [1]. This interest seems to originate from the astonishing difference between graphene and other well-known two-dimensional semiconductor systems. This single layer, one atom thick, sheet of carbon has atoms which are arranged into a honeycomb lattice, which produces a unique bandstructure where the bands and electron transport are mainly derived by the surface normal p$_z$ orbitals. Near the K point, the bandstructure itself is described by the Dirac equation, in which charge carriers imitate relativistic particles with zero rest mass. This strictly two-dimensional material exhibits exceptionally high crystalline and electronic quality, and despite its short history, has already revealed an abundance of new physics and potential applications.

While very high mobilities have been observed at low temperature [2] such high mobilities usually are not observed at room temperature, even though they have been predicted [3]. There are several factors that can affect the transport of carriers in graphene and degrade its mobility. Among the possible factors suggested have been the presence of charged impurities [4–12], corrugation of the graphene sheet [13, 14] and short-range disorder [3, 4, 15, 16]. Chen *et al* [4] investigated the effect of charged impurities on graphene's conductivity by depositing various doses of potassium atoms on the graphene surface and measuring the conductivity at 20 K temperature in ultrahigh vacuum. Their experiments clearly demonstrated the decreases of mobility with increasing impurity density. The role of remote impurity scattering was further confirmed when Bolotin *et al* [2, 17] reported

drastically reduced carrier scattering in suspended graphene devices when they removed the residual impurities by current annealing. In addition, Jang *et al* [18] have placed solid ice on the surface of graphene (on SiO$_2$) and observed an improvement in mobility, a result disputed by the Manchester–Nijmegen collaboration [19]. More recently, we have demonstrated that depositing high dielectric constant *liquids* on a graphene device screens the impurities from the graphene and leads to very high mobilities at room temperature [20, 21]. In this case, the liquid apparently penetrates between the graphene and the oxide to enhance the screening of the impurities in the oxide. Mobilities above $40\,000$ cm^2 V^{-1} s^{-1} were found [21]. Once the dielectric constant of the dielectric liquid is sufficiently high, no further increases in mobility are found, and the mobility seems to saturate. The question then arises as to whether this intrinsic graphene mobility is due to the phonons in the single layer or not. In this paper, we address this issue using a simulation based upon Rode's method [22]. We extract the mobilities for 3 high dielectric constant situations, taken from [21], and then match the theoretical mobility with these data. While impurity scattering can still affect the mobility at very low density, the behavior of the mobility at high density is consistent with scattering due to intrinsic phonons, both acoustic and optical.

It is very common to determine the mobility from the derivative of the conductance with respect to gate voltage, but this incurs a common error. In fact, this procedure gives the

general result

$$\frac{1}{C_G}\frac{\partial \sigma}{\partial V_G} = \mu + V_G \frac{\partial \mu}{\partial V_G}, \tag{1}$$

and the last term is not insignificant at low densities where the mobility varies rapidly with density, which is the case in the data of [21]. Consequently, we use a different approach to extract the mobility from the measured conductivity. The impurity density can be determined from the shift of the 'threshold voltage' and the oxide capacitance (after accounting for the dielectric liquid capacitance and the quantum capacitance of the graphene layer). Then, we use the following formulation to extract the mobility from the conductance–gate voltage data [4]

$$\mu = \frac{\sigma - \sigma_{min}}{C_G(V_G - V_{G\_min})}. \tag{2}$$

The extracted mobilities for the three solvents are shown in figure 1. As mentioned above, we used Rode's method which is an iterative procedure on a linearized Boltzmann equation. The advantage of using Rode's method over the relaxation time approximation (RTA) is that Rode's method is capable of providing adequate accuracy for calculating mobility in the presence of inelastic scattering processes, which the RTA usually fails to do. Rode introduced an iterative procedure to calculate the actual distribution function for the low field situation. The distribution function is written as

$$f(\mathbf{k}) = f_0(E) + g(\mathbf{k})\cos(\theta). \tag{3}$$

Here θ is the angle between electric field $\mathbf{F}$ and the momentum $\mathbf{k}$ vector. For an isotropic system, $g(\mathbf{k})$ can be calculated as

$$g(\mathbf{k}) = \frac{S_{in} - evF\frac{df_0}{dE}}{S_{out} + \frac{1}{\tau_m}}, \tag{4}$$

where

$$S_{in} = \sum_{\mathbf{k}'} g(\mathbf{k}')\{P^{in}(\mathbf{k}',\mathbf{k})[(1 - f_0) + P^{in}(\mathbf{k},\mathbf{k}')f_0]\}\cos(\theta_0)$$

$$S_{out} = \sum_{\mathbf{k}'}\{P^{in}(\mathbf{k},\mathbf{k}')[(1 - f_0) + P^{in}(\mathbf{k}',\mathbf{k})f_0]\} \tag{5}$$

are the net in- and out-scattering functions. Here, $P^{in}(\mathbf{k},\mathbf{k}')$ is the inelastic scattering rate and θ_0 is the angle between $\mathbf{k}$ and $\mathbf{k}'$ vectors. Initially it is assumed that $g(\mathbf{k}) = 0$ and therefore, $S_{in} = 0$. If S_{out} is known then $g(\mathbf{k})$ for the next iteration step can be calculated and an updated S_{in} will be determined. In the case for which the inelastic scattering is isotropic, as here in graphene, this iteration requires only the first update. Once $g(\mathbf{k})$ is known, mobility in graphene can be calculated as

$$\mu(n_s) = \frac{2v_F \int_0^{k_F} g(k)k\,dk}{\pi n_s F}. \tag{6}$$

It is apparent from figure 1 that the two lower dielectric constant materials give essentially the same high density behavior, which is likely due to the intrinsic phonon scattering in the graphene. The fact that the highest dielectric constant
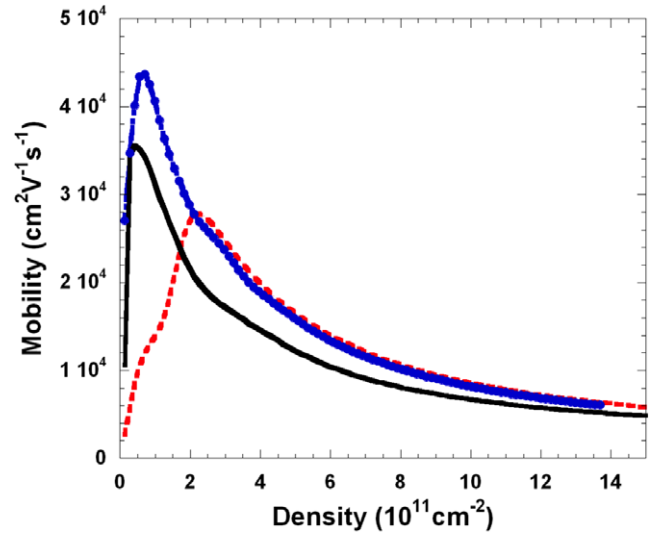
Figure 1. Extracted mobility using equation (2) from the measured data of conductance for the three high K dielectric solvents, taken from [21]. The solid (black), dot–dashed (blue), and dashed (red) curves are for dielectric constants of 189, 66, and 47, respectively.

material leads to a lower mobility suggests that some other scattering process is present, a point to which we return later. As a result of the match in mobilities, we use the high density parts of the curves to determine the coupling constants particularly for acoustic phonon scattering.

In studying the 'intrinsic' mobility in graphene, Chen *et al* [3] considered acoustic modes of the graphene phonons, but neglected the optical modes. Instead, they assumed that remote interface modes [23], derived from the polar interaction in SiO$_2$, would be important. But, these modes are known not to be very important, even in the Si/SiO$_2$ case [24, 25]. Moreover, the interaction of these modes is predominantly Coulombic in nature, and this will be screened by the high dielectric constant materials. Instead, we consider scattering by the non-polar optical phonons, arising from the K point phonons, which couple the K and K' valleys of the conduction band. From studies of the phonon structure, this is the LA + LO mode with an energy of 150 meV [26]. Both the long wavelength intravalley optical mode and the zone edge TA mode are forbidden by symmetry [27], and there is an out-of-plane mode near the latter that can give rise to ripple modes in second order [28]. In the rigid ion approximation, and with a deformation field D_0 and optical phonon frequency ω_0, the scattering rate becomes

$$\frac{1}{\tau} = \frac{D_0^2}{\rho_m \omega_0 (\hbar v_F)^2}[(E - \hbar\omega_0)(N_q + 1)u_0(E - \hbar\omega_0) + (E + \hbar\omega_0)N_q]. \tag{7}$$

The function u_0 assures that the energy is sufficient to emit a phonon. For acoustic phonons, the scattering rate derived by Hwang and Das Sarma [29] for the equipartition limit (high temperature, and corrected for an apparent typographical error) is given as

$$\frac{1}{\tau(E)} = \frac{4D^2 k_B T}{\hbar^3 \rho_m (v_{ph} v_F)^2} E(k). \tag{8}$$

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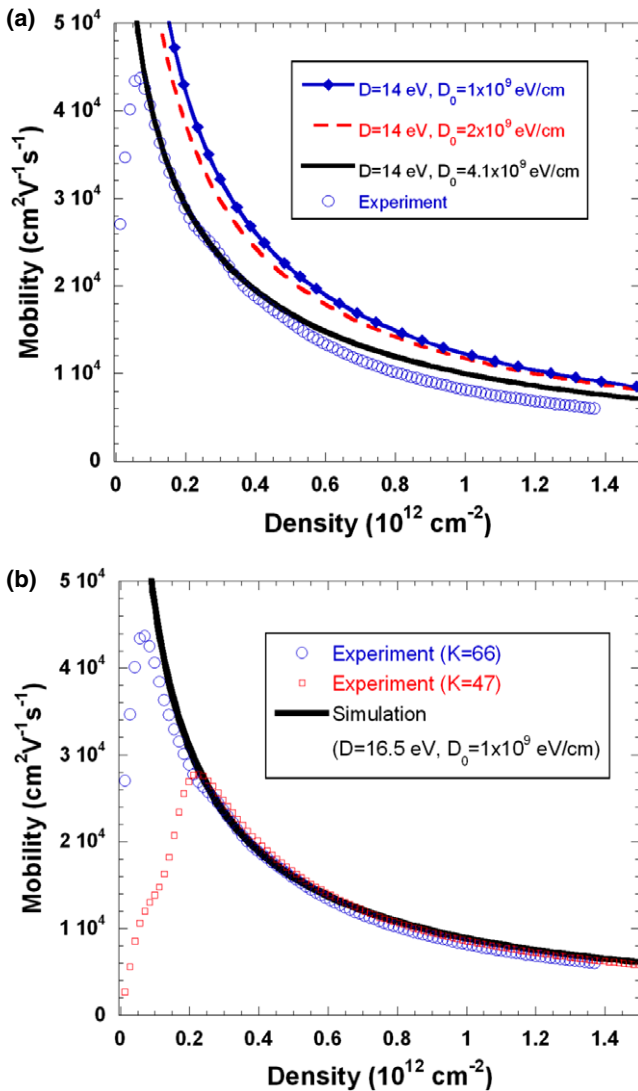
Figure 2. Fits to the mobility curve for a dielectric constant of 66. (a) Fit using an acoustic coupling constant of 14 eV, and a variety of optical mode coupling constants. Using the value of 4.1×10^9 eV cm^{-1} gives a reasonable fit. (b) Fit using an acoustic coupling constant of 16.5 eV, and an optical mode coupling constants of 1.0×10^9 eV cm^{-1} gives the best fit.

In figure 2, we use the mobility curve for the liquid with a dielectric constant of 66, as this gives the highest mobility over the entire range of densities. Clearly, there remains a Coulombic contribution from the impurities at very low densities, but this is reduced not only by the screening, but by the fact that these impurities are now further from the graphene as well. In figure 2(a), we use a value of 14 eV for the acoustic deformation potential and a range of values for the optical phonon coupling constant. The largest value (4.1×10^9 eV cm^{-1}) gives a reasonable fit to the mobility curve, however the slopes at the highest density do not match particularly well. In figure 2(b), we increase the acoustic coupling constant to 16.5 eV, closer to the value suggested (18 eV) by Chen *et al* [3]. With this larger value, the acoustic scattering is increased and the optical scattering must be decreased accordingly. Here, a value of 1.0×10^9 eV cm^{-1}

is found to fit the mobility over the entire range of densities in the figure. Moreover, we find that the fit is sufficiently good that surface roughness scattering probably does not contribute over this density range. It is clear that the screening by the dielectric fluid means that this liquid is penetrating between the graphene and the oxide. But, whether or not this smooths the graphene surface is unknown.

Because of the decreasing impurity scattering and increasing phonon scattering, mobility reaches its peak around 1×10^{11} cm^{-2} density and begins to fall afterward. We do not take account for any variation in the charge density in the puddles, which may change as the gate voltage is varied. We also assume that the total conductance is a simple addition of puddle charge conductance and gate induced charge conductance as indicated in (2). The validity of this assumption may be questioned, but we do not have any more reasonable model to use, and Morozov *et al* [28] suggest this background conductance is independent of carrier density. Nevertheless, apart from these mismatches, the simulation suggests the upper bound of the mobilities that can be achieved.

As mentioned above, the experimental mobility for the highest dielectric constant material lies well below the other two, and well below the theoretical curve. At present, we do not understand this. However, it is well known that high dielectric constants are usually accompanied by low energy phonon modes (so-called soft modes in solid dielectrics), and it may be that such a mode is affecting the mobility in this latter case as a remote (and polar) optical phonon [30]. But, this is no more than a supposition, and further work needs to be done. Although not shown here, we have also considered the possibility of scattering from a first-order interaction of the normally forbidden TA mode, but it is found that this interaction is far too weak to affect the results discussed here.

In conclusion, we find a good agreement with experimentally measured mobilities and scattering due to acoustic phonons. These show peak mobilities above $40\,000$ cm^2 V^{-1} s^{-1}, which are among the highest mobilities that have been reported for graphene at room temperature. These simulations suggest that acoustic phonon and intervalley optical phonon scattering dominate the mobility in this material. A good understanding of transport mechanisms for the puddle charges and their contribution to the total conductance is necessary for further improvements of the simulation.

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