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Systematic THz study of the substrate effect in limiting the mobility of graphene

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We explore the substrate-dependent charge carrier dynamics of large area graphene films using contact-free non-invasive terahertz spectroscopy. The graphene samples are deposited on seven distinct substrates relevant to semiconductor technologies and flexible/photodetection devices. Using a Drude model for Dirac fermions in graphene and a fitting method based on statistical signal analysis, we extract transport properties such as the charge carrier density and carrier mobility. We find that graphene films supported by substrates with minimal charged impurities exhibit an enhanced carrier mobility, while substrates with a high surface roughness generally lead to a lower transport performance. The smallest amount of doping is observed for graphene placed on the polymer Zeonor, which also has the highest carrier mobility. This work provides valuable guidance in choosing an optimal substrate for graphene to enable applications where high mobility is required.

Introduction

Graphene, an atomic layer of carbon atoms arranged in a honeycomb lattice, is not only a rich playground for uncovering new physical phenomena, but also a potential component in future electronic and optoelectronic technologies¹⁻³. For example, graphene has already been implemented into a broadband image sensor array for enhanced photodetection⁴ as well as memristors based on layered two-dimensional materials⁵. Graphene was also demonstrated to be a sensitive chemical and biological sensor⁶⁻⁸. With the development of large-scale production techniques such as epitaxial growth or chemical vapor deposition (CVD), large area graphene shows increasing promise for implementation into macroscale modern devices⁹.

Among its many properties, the high mobility of the massless Dirac fermions in graphene plays a key role in the implementation of high-speed electronic devices. Therefore, it is imperative to develop better understanding and control over the factors that affect scattering mechanisms in graphene films. Factors that limit large-area graphene's mobility can be either intrinsic or extrinsic¹⁰. Intrinsic limiting factors include scattering from lattice defects, grain boundaries¹¹⁻¹⁴, or phonons^{15,16}. Due to the formation of grain boundaries and defects during the CVD growth, such scattering mechanisms could be more pronounced in large-area graphene compared to micron-scale mechanically exfoliated samples^{17,18}. Among extrinsic limiting factors, scattering due to charged impurities is known to play a significant role¹⁹. Charged impurities can be present

in the underlying substrate²⁰, trapped between the film and substrate²¹, or on the surface of graphene in fabrication residues²².

Graphene supported by common substrates such as SiO₂, Si₃N₄, quartz or silicon is reported to have carrier mobilities varying between 500-10,000 cm²V⁻¹s⁻¹^{19,23-25}. Efforts to engineer substrates that minimize the presence of trapped charges and mechanical deformations led to three orders of magnitude improvement, with the highest mobilities (larger than 10⁶ cm²/(Vs)) reported in freely suspended devices^{26,27} or on top of atomically flat boron nitride (BN) crystals^{17,28}. It is therefore critical that the graphene-substrate interaction be well understood for successful integration of large-area graphene films with high mobility. Substrates that ensure high mobility for graphene films could be exploited for improving device performance in future graphene integrated electronics.

To date, graphene-substrate interactions have been studied in devices by measuring the unpumped/non-excited carrier dynamics with both optical techniques and electrical transport techniques. For example, far-infrared spectroscopy (3-16 THz) was used to characterize graphene on different polar dielectric and organic polymer films. Graphene on hexamethyldisilazane (HMDS) notably exhibits a mobility four times higher than on SiO₂^{29,30}. Electrical transport was also used to investigate large-scale graphene deposited on several dielectrics commonly used in the semiconductor industry. The highest mobility was achieved when graphene was placed on Si₃N₄, which outperformed SiO₂, HfO₂, Al₂O₃, and tetraethyl orthosilicate. This work indicated that carrier density fluctuation caused by the substrates is one of the main contributing factors for mobility degradation³¹. Other studies relied on time-resolved THz spectroscopy to explore the substrate effect on graphene placed on sapphire, silicon, germanium, polyethylene naphthalate (PEN), polyethylene terephthalate (PET), and quartz^{32,33}. This technique provided information about the graphene DC conductivity, but the experimental spectral bandwidth, limited to frequencies up to 3 THz, could not resolve features attributed to carrier scattering. As a result, no information was obtained on carrier density or mobility. Finally, there has been more work that measured^{23,34-36} and even spatially resolved^{24,25,37} the transport properties of wafer scale graphene using THz spectroscopy. These studies, however, are restricted to only one specific substrate, or do not investigate the substrate dependent dynamics at play.

Here, we perform time-domain terahertz transmission spectroscopy over a bandwidth extending from 0.5 to 5.5 THz to measure graphene's transport properties when supported by substrates with different charged impurity densities, dielectric constants, and surface roughness. Our THz spectral window, extending beyond the limit of most commercial systems, is sufficiently broad to capture the spectral-dependent change in conductivity associated to the Drude roll-off frequency and therefore allows for measurement of both carrier density and Drude mobility. In comparison to other characterization methods based on electrical measurements, THz spectroscopy is a non-invasive technique that does not alter the graphene structure or rely on the deposition of electrical contacts, which composition and configuration often affect the measured transport parameters³⁸.

As such, this all-optical technique has a great potential to map out properties of large graphene sheets to optimize its integration into electronics or opto-electronics devices^{24,25,37}.

The selection of substrates in this work is guided by the promise for graphene to be implemented into “beyond Moore” technologies¹. We compare the mobility of graphene deposited on different materials: two gate dielectrics (SiO₂ and Si₃N₄) which are traditional materials for computational-based applications, silicon with three doping concentrations, and a hydrophobic, flexible, transparent polymer (Zeonor). We find that substrates with minimal amount of charged impurities and substrates that are flat generally exhibit the highest carrier mobilities. Notably, graphene on high-purity silicon and insulating Zeonor outperform the popular gate dielectrics. Our results guide the field of graphene integrated technologies, by providing valuable insights into the transport potential of graphene supported by substrates commonly used in devices.

Results and Discussion

In this work all samples are identically prepared, using graphene synthesized by CVD on Cu foil and transferred onto a substrate using a standard PMMA-based wet etching technique, schematically represented in Fig. 1a. A micrograph of transferred graphene on SiO₂ in Fig. 1b shows continuous coverage across a large area. Atomic force microscopy (AFM) and Raman spectroscopy confirmed monolayer thickness as demonstrated by the height profile (~1 nm) and Raman spectrum ($I_{2D}/I_G = 1.94$) shown in Fig. 1c and Fig. 1d.

For the measurements, we employ a home-built time-domain terahertz spectrometer, schematically represented in Fig. 1e. The system is based on an optical source delivering ~50 fs pulses centered at 1035nm³⁹. These pulses are focused onto a nonlinear GaP crystal where THz transients are generated by difference frequency mixing. The time-domain detection relies on electro-optic sampling inside a second GaP crystal⁴⁰, and the maximum signal sensitivity is found at 3.5 THz. The graphene sample, at room temperature and under normal ambient conditions, is placed at the THz focus and the measurement is performed in a transmission geometry. Note that we purge the system with dry air (< 0.5% RH) to minimize THz absorption in water vapor. Fourier transform of the time-domain data yields the complex field amplitude spectrum $\tilde{E}_s$ transmitted through the graphene sample. As shown in Fig. 2a, a reference field amplitude spectrum $\tilde{E}_{ref}$ is collected through the bare substrate, on a region without graphene, to calculate the complex transmission $\tilde{T} = \tilde{E}_s / \tilde{E}_{ref}$. Each measurement is repeated 10 times (Fig. 2c) under the same experimental conditions to obtain (i) an averaged signal with reduced noise and (ii) a frequency-dependent standard deviation indicative of the scan-to-scan experimental error.

The complex sheet conductance $\tilde{\sigma}_s$ can be straightforwardly extracted from $\tilde{T}$ considering graphene as an infinitely thin conducting film²⁴, such that:

$$\tilde{\sigma}_s = \frac{1+\tilde{n}_s}{Z_0} \left(\frac{1}{\tilde{T}} - 1 \right), \quad (1)$$

where $\tilde{n}_s$ is the substrate refractive index and Z_0 the vacuum impedance (376.6 Ω). Fig. 2b shows a typical measurement and the corresponding transmission amplitude in the inset with error bars representing scan-to-scan uncertainties. Although Eq. 1 contains complex values, we do not consider the imaginary part of the conductivity in our analysis since this value can induce systematic errors as it is extremely sensitive to local variations of the substrate thickness. Therefore, we consider only the real part of the conductivity σ_s , which has proven to be sufficient to extract reliable graphene properties²⁵. The frequency-dependent Drude model, which has been broadly used to describe carrier intra-band scattering behavior in graphene^{24,25,34-36}, yields:

$$\sigma_s = \frac{\sigma_{DC}}{1 + \omega^2 \tau^2}, \quad (2)$$

where σ_{DC} is the DC conductivity, ω is the radial frequency, and τ is the scattering time. As a result, we are able to extract values for both σ_{DC} and τ from the THz data. In general, the analysis of time-domain THz measurements is restricted to data points within a carefully selected spectral window centered around the maximum THz signal, where experimental noise is considered negligible. Here, we use instead a scan-to-scan statistics to include a larger bandwidth while error bars objectively determine the precision of each spectral data point. More specifically, experimental data are fitted to the Drude model (Eq. 2) with a weighting factor inversely proportional to the square of the frequency-dependent uncertainty. This quantitative approach ultimately allows more data points to be considered in the analysis and can therefore improve accuracy and reliability. To determine another type of uncertainty related to sample anisotropy, we collect data on more than 5 different positions on each sample and perform again the analysis described above (Fig. 2c). We rely on these multiple measurements and their standard deviation to evaluate a relative uncertainty of σ_{DC} and τ , which is, in turn, used to calculate the relative uncertainty on other carrier transport parameters displayed in Figs. 3, 4 and 5. Finally, the full data acquisition process on each graphene sample is repeated at least twice on different days to test the reproducibility of our THz spectroscopy technique. This last step provides at least two data points, with their respective error bar, for all measured transport parameters and each substrate considered in our study.

We use the experimental procedure described above to obtain the parameters σ_{DC} and τ of graphene samples deposited on seven different substrates: three gate dielectrics on Si wafer (Si_3N_4 , wet thermal grown SiO_2 , and dry thermal grown SiO_2), Si with three doping concentrations corresponding to a resistivity of 10 $\text{k}\Omega\text{-cm}$ (intrinsic doping), 15 $\Omega\text{-cm}$ (p-doped), and 5 $\Omega\text{-cm}$ (p-doped), and finally the flexible polymer Zeonor, a cyclo-olefin copolymer. We note that all bare substrates display high transparency to our broadband THz probe. Assuming a diffusive Drude-type transport regime⁴¹⁻⁴⁴, we can then calculate graphene's carrier concentration (N_s) and the mobility (μ) from the extracted σ_{DC} and τ using the following relations:

$$N_s = \frac{\pi \hbar^2}{v_F^2 e^4} \left(\frac{\sigma_{DC}}{\tau} \right)^2, \quad (3)$$

$$\mu = \frac{\sigma_{DC}}{e N_s} = \frac{v_F^2 e^3}{\pi \hbar^2} \frac{\tau^2}{\sigma_{DC}} = \frac{v_F e}{\sqrt{\pi} \hbar} \frac{\tau}{\sqrt{N_s}}, \quad (4)$$

where the Fermi velocity for graphene $v_F \approx 10^6$ m/s. As a result, THz measurements enable an effective and non-invasive method for probing the influence of the substrate on the transport properties of graphene films.

In Fig. 3 we plot the calculated parameters N_S , μ , and τ for the seven different substrates described above. Fig. 3a shows that the carrier concentration N_S in graphene is strongly dependent on the substrate. This variation, by more than an order of magnitude, can be attributed to the densities of free and trapped charges in the substrate, which have previously been identified as two dominant parameters affecting N_S ^{19,42}. For the insulating substrates, trapped charges are particularly relevant, as those present at the surface may directly transfer charges to graphene and those beneath the surface may still capacitively induce charges in graphene. For the conductive substrates, N_S is largely due to direct charge transfer. In our experiment, the largest values of N_S are obtained for graphene placed on doped Si and approach 10^{13} cm⁻², corresponding to graphene's Fermi level being 350 meV away from the Dirac point. This value is comparable to epitaxial graphene grown on SiC⁴⁴. Insulating substrates, including typical gate dielectrics (SiO₂ and Si₃N₄), high-resistivity Si ($\rho = 10^4$ Ω-cm) and the polymer Zeonor ($\rho = 10^{16}$ Ω-cm), lead to a lower N_S . The lowest doping level we measured corresponds to graphene on Zeonor ($N_S = 8 \times 10^{11}$ cm⁻²), indicating that this substrate has a minimal amount of free and trapped charges contributing to the doping of the 2D material on its surface.

Fig. 3b shows the calculated mobilities for graphene films placed on the different substrates, presented in increasing order from left to right based on the calculated N_S . We obtain higher mobilities for less doped samples, consistent with the notion that substrate charged impurities create an electrostatic potential that limits the mobility^{19,20}. In particular, we find that graphene samples supported by high-resistivity Si and the insulating polymer Zeonor, show the highest mobilities $\mu \sim 5,000$ cm²V⁻¹s⁻¹, more than an order of magnitude higher than graphene on Si 5 Ω-cm and Si 15 Ω-cm substrates. This indicates that boron impurities doping the Si crystals can act as scattering centers, reducing carrier mobility in the graphene film. Interestingly, graphene on high-resistivity Si shows a higher mobility than the one on gate dielectric substrates although they all possess trapped charges and have a similar carrier concentration N_S . To understand this result, we note that a flat electrostatic potential profile, even if it causes charge doping, doesn't necessarily cause scattering. In the case of substrates with large amounts of trapped charges, their distribution can be non-uniform leading to more significant scattering⁴⁶. This concept is supported by the measured scattering times τ shown in Fig. 1c. Most samples feature a τ around 40 fs, except for graphene on high-resistivity Si, which exhibits a longer scattering time of 95 fs.

The dielectric constant κ of the substrate or environment can play an important role in the transport properties of graphene; larger values of κ imply the ability of an environment to readily screen charged potentials created by trapped scattering centers, and therefore enhance the carrier mobility^{48,49}. In Fig. 4, we plot the DC conductivity σ_{DC} , as well as τ_{scat} , N_S , and μ , as a function of the substrate's dielectric constant κ . We observe that three of these parameters: σ_{DC} , τ and N_S ,

generally increase with increasing κ , while the graphene's mobility μ does not appear to depend strongly on the substrate's dielectric constant. For the calculated τ and μ in Fig. 4b and Fig. 4d, the value obtained for graphene placed on the doped Si substrate slightly departs from the general trend. We attribute this discrepancy to a strong inhomogeneous electrostatic potential created by the boron dopants in these substrates. Surprisingly, we observe that graphene on Zeonor exhibits the highest mobility, even though this substrate has the lowest dielectric constant among the samples measured. This indicates that the screening of charged impurities in the substrate is not necessarily the main contributor to the high mobility, but other factors come into play, such as the density of trapped and free charges in the substrate, as discussed previously.

Although much less systematically studied, substrate roughness can also affect the carrier mobility in graphene since a smooth interface contributes to reduced strain of the film or eliminates wrinkles and edges that could induce scattering. Using atomic force microscopy, we measure the roughness (R_q) of the seven substrates used in this experiment. In general, we observe high carrier mobility for smooth substrates (small values of R_q). For example, Si-10k Ω -cm ($R_q = 220$ pm) out-performs wet SiO₂ ($R_q = 380$ pm). However, we note two exceptions in our experiment: (i) graphene on doped Si features a low mobility although the substrate is relatively smooth. This suggests that Coulomb scattering is a dominant mechanism in this case. (ii) At the other extreme, graphene on Zeonor has one of the highest mobilities in our study while the substrate roughness is higher, $R_q = 650$ pm. To assess the adhesion of graphene to Zeonor, we measured the roughness of sample when graphene was placed on Zeonor, which shows a smoother morphology ($R_q = 520$ pm), suggesting relatively poor adhesion of the graphene to the substrate. As a result, graphene may locally be suspended and experience a reduced influence from the substrate. This characteristic combined with the indication of minimal trapped charges at the graphene-Zeonor interface (see Fig. 3a and related discussion), explains the measured high mobility. The outperformance of graphene on a hydrophobic polymer compared to a gate dielectric is consistent with previous reports where graphene field effect transistors on parylene⁵⁰ and HMDS^{29,30} demonstrated reduced ambient doping and hysteresis compared to those on SiO₂.

Conclusion

In summary, we have demonstrated the implementation of terahertz transmission spectroscopy for probing the equilibrium transport properties of CVD graphene supported by seven substrates, distinct in their density of charged impurities, dielectric constant and surface roughness. We find that the resulting conductance spectra for graphene films agrees well to a Drude model for intraband behavior, consistent with other reports. We have demonstrated a method for weighted Drude fitting based on statistical averaging and extracted transport properties (N_s , μ , τ , and σ_{DC}), for graphene supported by distinct substrates. When comparing the results on different substrates, we find the highest mobility for graphene placed on pure silicon and the polymer Zeonor, which outperformed commonly used gate dielectrics. This highlights that the densities of trapped and free charged impurities in the substrate are likely key limiting factors of the graphene film's mobility. We also find the mobility of devices to be generally improved for substrates that are

smoother, except for graphene on Zeonor. In this case, we interpret the high mobility to be due to both the poor adhesion between Zeonor and graphene, minimizing the substrate effect, and to the low density of trapped charged impurities at the interface. A flexible polymer, therefore, can be considered as a promising alternative to some gate dielectrics. These results provide valuable guidance for the effective integration of graphene into technological platforms where high mobility is necessary.

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AUTHOR CONTRIBUTIONS

SS fabricated the samples, took the data, analyzed the data, and wrote the manuscript. WC took data and participated in the manuscript presentation. AL-M and J-MM coordinated the project and wrote the manuscript. All authors discussed the data and its interpretation.

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Figure 1: Sample fabrication and experimental set-up. (a) Schematic of sample fabrication: CVD graphene on Cu foil is transferred onto a desired substrate. (b) 5X optical microscope image of graphene (right section) on SiO₂ (left section). (c) Raman spectrum of a transferred graphene film featuring distinctive G and 2D peaks. (d) Atomic force micrograph of graphene transferred on SiO₂ together with a height profile across the graphene edge, indicated by the white line. (e) Schematic of the time-domain terahertz spectrometer with its optical components: beam splitter (BS), translation stage (TS), gallium phosphide crystals (GaP), germanium and silicon wafer (Ge and Si), quarter wave plate (QWP), Wollaston prism (WP), and balanced photodetectors (PD).

Figure 2: Description of the THz spectroscopy experimental procedure. (a) Geometry of the THz reference and sample measurement. (b) The real sheet conductance is extracted by comparing the transmission through the reference and sample (inset). Solid line represents experimental data and the error; dashed line represents the error weighted Drude fit to the data. (c) Description of the data collected for signal averaging and estimation of uncertainty: 10 scans are taken at a single spot on the graphene sample, indicated by a red dot. This is repeated across the sample at different spots at least 5 times, indicated by the several red dots.

Figure 3: Results obtained for seven distinct substrates by applying the Drude fit to our experimental conductance spectra. The calculated transport parameters are: (a) carrier density N_S , (b) Drude carrier mobility μ , and (c) scattering time τ . Solid squares represent the average value measured across several spots on a sample. Error bars represent one standard deviation. Measurements are repeated at least twice for each sample on a different day under similar experimental conditions.

Figure 4: Experimentally measured transport parameters plotted as a function of the substrate dielectric constant κ : (a) DC conductivity σ_{DC} , (b) scattering time τ , (c) carrier density N_s , and (d) mobility μ .

Figure 5: Mobility of graphene devices supported by substrates with different surface roughness.

Figures

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