Wafer-scale characterization of carrier dynamics in graphene

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***Abstract*—The electronic properties of single-layer graphene, such as surface conductance, carrier concentration, scattering time and mobility, can be characterized in a noncontact manner by THz time-domain spectroscopy. Standard spectroscopic imaging reveals the AC conductance over large areas with a few hundred µm resolution, and spectroscopic imaging on back-gated graphene allows for extraction of both the carrier concentration and the mobility. We find that spatial variations of the conductance of single-layer CVD-grown graphene are predominantly due to variations in mobility rather than in carrier concentration.**

I. INTRODUCTION

GRAPHENE is being targeted for an increasing number of commercially oriented applications and industrial development, due to its combination of advantageous electronic, optical, and mechanical properties as well as the increasing availability of synthesized large-area graphene films. In particular, there is a strong interest in the commercial adaptation of large-area graphene of high electrical quality for electronics applications, including terahertz (THz) electronics and transparent, flexible, and durable electrodes for graphene-based displays, touch-screens and solar cells. There have already been several demonstrations of THz time-domain spectroscopy (THz-TDS) for rapid and contact-free conductance measurements of large-area graphene, making practical implementations of large-scale, in-line spatial mapping of graphene sheet conductance possible. For most electronic applications, however, the conductance of a graphene film in itself gives an incomplete picture of the performance. For many scientific and commercial applications, the carrier mobility and the background carrier density originating from chemical doping are the crucial parameters. The most straightforward way to obtain this information is through the electric field effect, which requires a variable gate potential. Such characterization in contacted graphene devices ultimately results in the dissection and processing of the sample, if any kind of spatial or statistical information is required. This is a destructive process, and thus the question as to whether the final devices truly represent the initial state of the unprocessed film of graphene, is difficult to answer.

Here we give an overview of recent efforts on contact-free characterization of large areas of single-layer graphene, with emphasis on a detailed understanding of the relation between THz-frequency and DC conductivity, effects of domain boundaries and growth conditions [1-4]. In particular we demonstrate quantitative mapping of the field-effect carrier mobility in a large-area monolayer CVD graphene film based on in-situ electrically gated THz-TDS imaging. We employ a low-mobility, high carrier concentration gate-electrode material, in this case p^+ boron-doped, nano-crystalline silicon, to ensure a negligible THz response from free carriers injected into the gate-electrode. This allows for isolation of the graphene sheet conductance from non-contact THz-TDS transmission measurements and thus a quantitative extraction of the graphene field-effect mobility. The experimental configuration is shown in Figure 1.

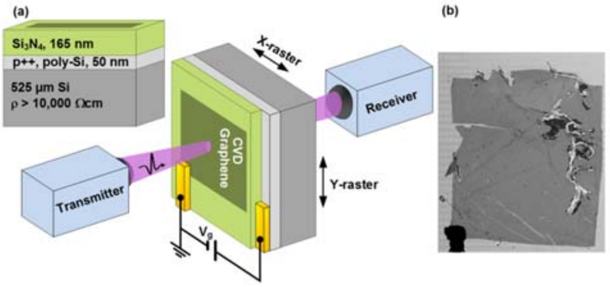

Figure 1: (a) Sample composition and experimental setup for back-gated THz conductance mapping of single layer graphene, (b) high-resolution image of the sample investigated here (reproduced from [4]).

II. RESULTS

THz transmission spectral maps are recorded, and converted into conductance maps [1]. These maps are recorded at various gate voltages, exemplary spectra in the 0.3-1.2 THz range are shown in Figure 2(a). A Drude-like behavior of the conductance is observed, with a slight indication of carrier confinement on the length scale of the mean free path of the carriers. The carrier concentration is related to the gate voltage relative to the charge neutrality point (CNP),

$$\left|\Delta n_S\right| = C_g / (e\Delta V_g) = \varepsilon_0 \varepsilon_{SiN} / (t \cdot e\Delta V_g)\,, \qquad (1)$$

where C_g is the gate capacitance and ε_{SiN} and t are the permittivity and thickness of the Si_3N_4 gate dielectric. Within the regime of long-range, charged impurity scattering, there is a linear relation between the sheet conductance σ_s and the carrier concentration n_s, leading to a relation for the field-effect mobility μ_{FE},

$$\left|\Delta\sigma_S\right| = e\mu_{FE}\left|\Delta n_S\right| = \frac{\mu_{FE}\varepsilon_0\varepsilon_{SiN}}{t}\Delta V_g\,, \qquad (2)$$

$$\mu_{FE} = \frac{t}{\varepsilon_0\varepsilon_{SiN}}\left|\frac{\Delta\sigma_S}{\Delta V_g}\right|. \qquad (3)$$

At voltages below the CNP, conductance is dominated by holes due to environmental p-doping of graphene by residual absorbates. Above the CNP, it can be seen that the electron mobility is an order of magnitude smaller than the hole mobility. Figure 2(b) shows the extracted hole mobility, recorded at three different positions indicated in Figure 3(a). Full mobility and carrier concentration maps of the sample can now be established by repeating the above procedure for each pixel in the gate-bias dependent conductance maps, as shown in Figure 3(a) and (b).

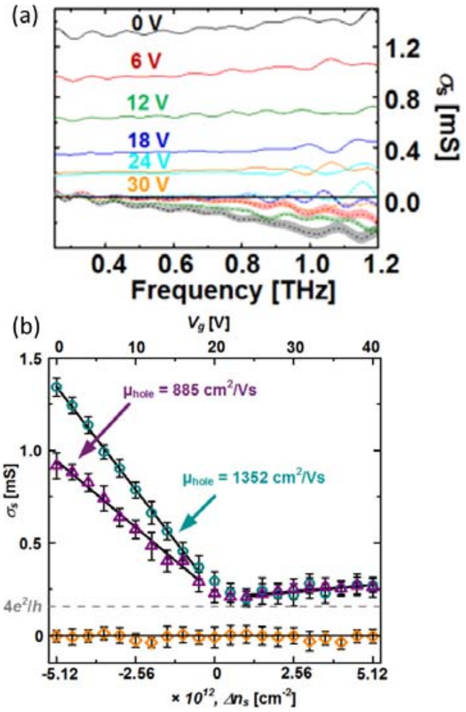

Figure 2: (a) THz conductance spectrum of graphene as function of back-gate bias voltage. (b) Extracted conductance vs. carrier concentration at selected positions on the sample (see Fig. 3) (reproduced from [4]).

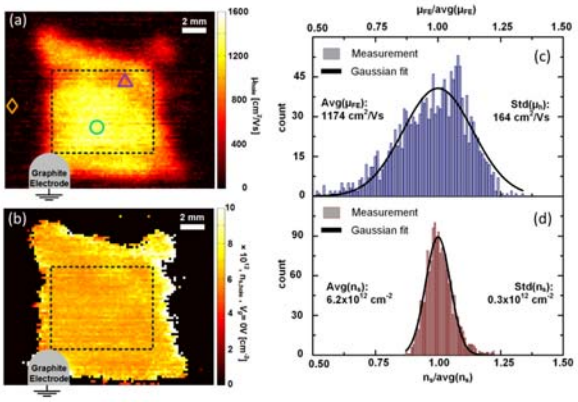

Figure 3: (a) Hole mobility map, (b) carrier concentration map, distribution of (c) mobility and (d) carrier concentration within the area marked by a dashed rectangle in (a) and (b) (reproduced from [4]).

Figure 3(c) and (d) shows a histogram of the mobilities and carrier concentration distribution within the rectangular area indicated in Figure 3(a) and (b). The relative standard deviation of the mobility distribution is significantly larger than that of the carrier concentration, indicating that variations in the conductance across the sample is due to variations in local mobility rather than local variations of the carrier concentration.

III. SUMMARY

Electrically gated THz time-domain spectroscopic imaging offers the capability of non-contact, quantitative mapping of graphene field effect mobility, here demonstrated in a 10x10 mm^2 large-area CVD-grown single-layer graphene film. While THz-TDS is a non-contact optical characterization, the frequency range is well below the inverse scattering time of the graphene charge carriers, making the obtained conductance directly comparable the DC value. The technique thus opens up the possibility for assessment of fundamental electrical transport properties such as carrier mobility and chemical doping level on basis of large statistical ensembles, or large-area spatial mapping. Far from the charge-neutrality-point, our measurements show a linear dependence of the THz sheet conductance on carrier density, which is interpreted as a signature of electrical transport limited by long-range, charged impurity scattering, also observed in most DC transport measurements on graphene. Unexpectedly, we find that the mobility varies by up to a factor of 2 on a scale of just few mm, and that significant conductance variations in the investigated CVD graphene are due to mobility rather doping variations, which highlights the importance of techniques employing statistical and spatially resolved approaches for assessing transport properties in large-area graphene.

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