Non-Drude CVD graphene terahertz conductance dynamics
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Abstract
Using ultra-broadband terahertz time-domain spectroscopy (THz-TDS), we report on the first observation of Non-Drude equilibrium charge carrier dynamics in a large-area graphene film grown by chemical vapour deposition on copper foil. Sheet conductance spectra for CVD graphene films grown under 2 different sets of growth conditions are obtained by ultra-broadband THz-TDS based on terahertz time-domain spectroscopy as well as conventional THz-TDS based on photoconductive antennas, spanning the entire frequency range from 0.1 to 15 THz associated with intraband optical conductance. In the case of CVD graphene grown under optimized conditions, a Drude intraband conductance spectrum is observed, while a conductance spectrum showing distinct non-Drude characteristics are observed in the case of less optimized CVD graphene growth. The observations are interpreted in the framework of the Drude-Smith model[1] for restricted nanoscopic carrier movement caused by preferential carrier backscattering on extended electronic barriers. The sheet conductance is extracted from an analysis of the Fresnel coefficients of reflection and transmission for the sample geometry. For the pulse transmitted directly through the sample and the 1st roundtrip echo, the sheet conductance is given as

\[
\tilde{\sigma}_{s,1}(\omega) = \frac{1}{Z_0} \frac{n_A}{\bar{T}_{film}(\omega)} - n_A
\]

\[
\tilde{\sigma}_{s,2}(\omega) = \frac{\pm n_A n_B^2 \bar{T}_{film}(\omega) - \bar{n}_A^2 - 2 n_A n_B^2 \bar{T}_{film}(\omega)}{2 n_B Z_0 \bar{T}_{film}(\omega)},
\]

respectively, where \(\bar{T}_{film}(\omega) = \bar{E}_{sub+graphene}(\omega) / \bar{E}_{sub}(\omega)\), \(n_A = n_{sub} + 1\), \(n_B = n_{sub} - 1\) and \(Z_0 = 377\Omega\) is the vacuum impedance. To improve the S/N ratio of measurements on a graphene film with a lower THz response, the 1st roundtrip echo was in one case analyzed.

We observe that the complex sheet conductance spectrum of a high quality CVD graphene film grown under optimized conditions on a copper substrate and transferred to a SiO2/high-resistivity-silicon substrate intimately follows the Drude model, as seen from the data in figure 1(a) and (b). In contrast, the complex sheet conductance spectrum of a CVD graphene film grown under less optimized conditions on a copper substrate and transferred to a SiO2/high-resistivity-silicon substrate, shows distinct features that cannot be reproduced by the Drude model, as seen from the data in figure 1(c) and (d). These features, which include a slight, but distinct suppression of the real conductance from DC to around 2 THz and an imaginary conductance that goes slightly negative in the same region, have commonly also been observed in nano-disordered, nano-defected or nano-patterned systems such as e.g. semiconductor nano-crystal systems[2]–[5]. We therefore suggest an analogous interpretation of the observed conductance spectrum in terms of restricted nanoscopic carrier movement via preferential carrier back-scattering within the Drude-Smith framework.

The conductance spectrum measured for high quality CVD graphene grown under optimized conditions agrees extremely well with the Drude model, allowing an accurate determination of the carrier scattering time \(\tau = 68 \text{ fs}\) and \(\sigma_{DC} = 2.19 \text{ mS}\), corresponding to a mean free path of approx. 61 nm and carrier mobility and carrier density of approx. \(\mu = 2500 \text{ cm}^2/\text{Vs}\) and \(N = 5.45 \times 10^{11} \text{cm}^{-2}\), respectively, under the assumption of transport limited by long-range scattering on charged impurities. In contrast, the conductance spectrum for the CVD graphene grown under less optimized conditions shows a good fit with the Drude-Smith model given by

\[
\tilde{\sigma}(\omega) = \frac{W_D}{1-i\omega\tau} \left[1 + \frac{c}{1-i\omega\tau}\right],
\]

with a Drude weight \(W_D = 1.54 \text{ mS}\), a carrier scattering time \(\tau = 43 \text{ fs}\) and a back-scattering parameter \(c = 0.60\). The Drude-Smith model describes a suppression of DC and low frequency conductance as well
as a negative low frequency imaginary conductance as a consequence of charge carrier localization caused by preferential back-scattering on extended electronic barriers on the characteristic probing length scale of the measurement, given as \( L = \sqrt{D/(2\pi f)} \), as illustrated in Figure 2. For frequencies between 0.1-15 THz \( L \) is on the order of 5-100 nm in the investigated films. Likely origins of extended electronic barriers on this length scale in the graphene film include crystal domain boundaries, sub-unity growth coverage and transfer-related damages such as e.g. rips, fractures or ripples.

References


Figures

Figure 1: Ultra-broadband terahertz time-domain spectroscopy of CVD graphene sheet conductance. Terahertz transients transmitted through HR-silicon wafers and (a) high quality CVD graphene on HR-silicon, grown under optimized conditions and (c) CVD graphene on HR-silicon, grown under less optimized conditions. Complex, frequency-dependent sheet conductance extracted from time-domain measurements for (b) high quality CVD graphene on HR-silicon, grown under optimized conditions and (d) CVD graphene on HR-silicon, grown under less optimized conditions.

Figure 2: Preferential carrier backscattering on extended defects (e.g. crystal domain boundaries, transfer-induced rips, wrinkles, incomplete growth in CVD graphene) with a characteristic length scale smaller than or similar to the probing length scale of the THz-TDS experiment gives rise to suppression of low frequency conductance and negative imaginary conductance, as described by the Drude-Smith model [1].