

“Observation of an electrically tunable band gap in trilayer graphene”

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1. Optical conductivity of gated ABC-stacked graphene trilayers with hole doping

Figure S1 shows the optical sheet conductivity $\sigma(\hbar\omega)$ of ABC-stacked graphene trilayers at gate biases corresponding to induced hole doping. The results are qualitatively similar to those for the case of electron doping described in the main text. For hole doping, we observe an enhancement and splitting of the main peak at $\hbar\omega = 0.35$ eV. Just as for electron doping, this behavior is the result of the induction of a band gap. The high-energy component of the split peaks is broadened and reduced in strength at the highest gate bias voltages. We attribute this behavior to the lateral inhomogeneity in the electric field of the polymer electrolyte top gate rather than to the inherent material response of the trilayer graphene.

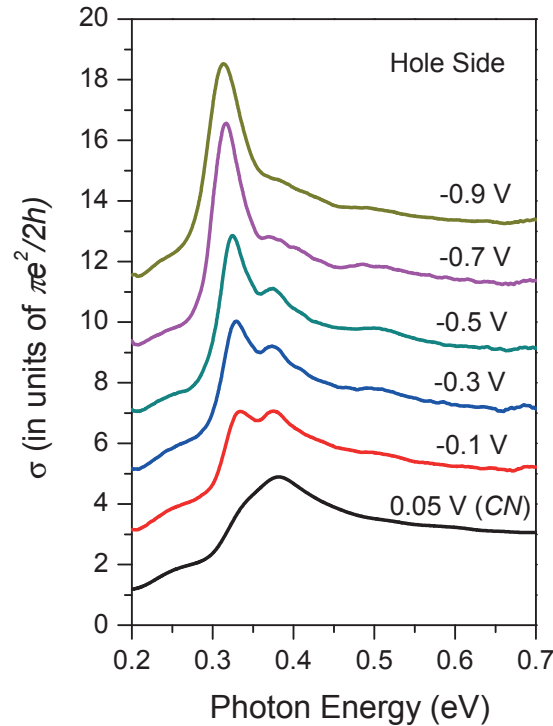


Figure S1 Measured optical conductivity spectra $\sigma(\hbar\omega)$ of an ABC-stacked graphene trilayer at gate biases corresponding to hole doping. The different spectra are displaced for 2 units for clarity. The gate voltage V_g and charge neutrality (CN) point are denoted on the spectra. The corresponding spectra for electron doping are shown in figure 2b of the main text.

2. Electron-hole asymmetry in ABA-stacked graphene trilayers

In contrast to the result for the ABC-stacked trilayer structure, the ABA trilayer displays a clear difference in its optical conductivity $\sigma(\hbar\omega)$ for applied fields corresponding to electron and hole doping (figure S2a). At the charge neutrality point of $V_{CN} = -0.65$ V, $\sigma(\hbar\omega)$ exhibits two peaks, one at $\hbar\omega = 0.520$ eV and the other at $\hbar\omega = 0.585$ eV. For increasing gate bias V_g (on the electron side), the amplitude of the higher-energy transition (0.585 eV) grows and the peak position red shifts, while the lower-energy transition (0.520 eV) subsides. As we decrease V_g (on the hole side), the low-energy peak grows and the high-energy peak subsides. The evolution of the energy of the absorption peak with gate bias is summarized in figure S2d.

The observed behavior in the ABA-stacked trilayer can be understood within the framework of a tight-binding (TB) model that includes not only the dominant intralayer (γ_0) and

interlayer (γ_I) couplings, but also the on-site energy difference δ and the parameter $v_4 = \gamma_4/\gamma_0$ describing the next-nearest neighbor interlayer coupling. The observed electron-hole asymmetry is attributed to the band structure asymmetry between the valence and conduction bands, as previously discussed for graphene bilayers [S1-4]. According to the TB analysis of ABA trilayer (Figure 1c in the main paper), sites A1, B2, A3 and B1, A2, B3 possess different energies (δ) because of the different crystal field environment. This leads to different transition energies for electron and hole doping. The next-nearest-neighbor interlayer coupling parameter v_4 produces different dispersion properties for the conduction and valence bands. This parameter is thus responsible for the different evolution of the electron and hole transition peaks with the gate voltage. The calculated band structure (figure S2e) clearly shows the role of coupling parameters δ and v_4 .

For a quantitative understanding of the ABA trilayer data, we have simulated the ABA trilayer IR conductivity by means of the Kubo formula, including a 20-meV phenomenological broadening. The conductivity at different induced charge densities n is calculated within a self-consistent scheme that takes into account the different potentials at the individual graphene layers that result from an uneven charge distribution in the sample (see Methods in the main text). We find that the main features of the experimental conductivity spectra and the dependence of absorption peak on the gate voltage are reproduced well for $\gamma_I = 371$ meV, $\delta = 37$ meV and $v_4 = 0.05$ (figure S2b and blue solid line in figure S2d). We have used a top-gate capacitance of $C = 0.8$ μFcm^{-2} to obtain the best description of the data. For comparison, we show the predicted $\sigma(\hbar\omega)$ spectra for a TB model with the same parameters, but under the neglect of any induced modification of the band structure (figure S2c). The calculated spectra are quite similar to those found when the change of the band structure is considered (figure S2b). The predicted absorption peaks are also in reasonable agreement with the experimental data (green dashed lines in figure S2d). We conclude therefore that the gate-induced modification of band structure is not needed to explain our experimental data.

The extracted on-site energy difference ($\delta = 37$ meV) in ABA-stacked trilayer is much larger than the corresponding value in the AB-stacked bilayer ($\delta = 18$ -25 meV) [S1-4]. The value is also much larger than the limit of $\delta < 22$ meV that we have estimated for the ABC stacked trilayer by considering the 44-meV width of the optical transition peak at $V_g = V_{CN}$ (figure 2b in the main text). The distinct behavior in the two cases is related to the difference in the crystal structure. Since δ arises from the change of local crystal field by the interlayer coupling, a trimer with two interlayer bonds is expected to have a larger value of δ than a dimer with only one interlayer bond. ABA trilayers, which feature trimers, should therefore exhibit a larger electron-hole asymmetry than that in bilayers or ABC trilayers, which only have dimers.

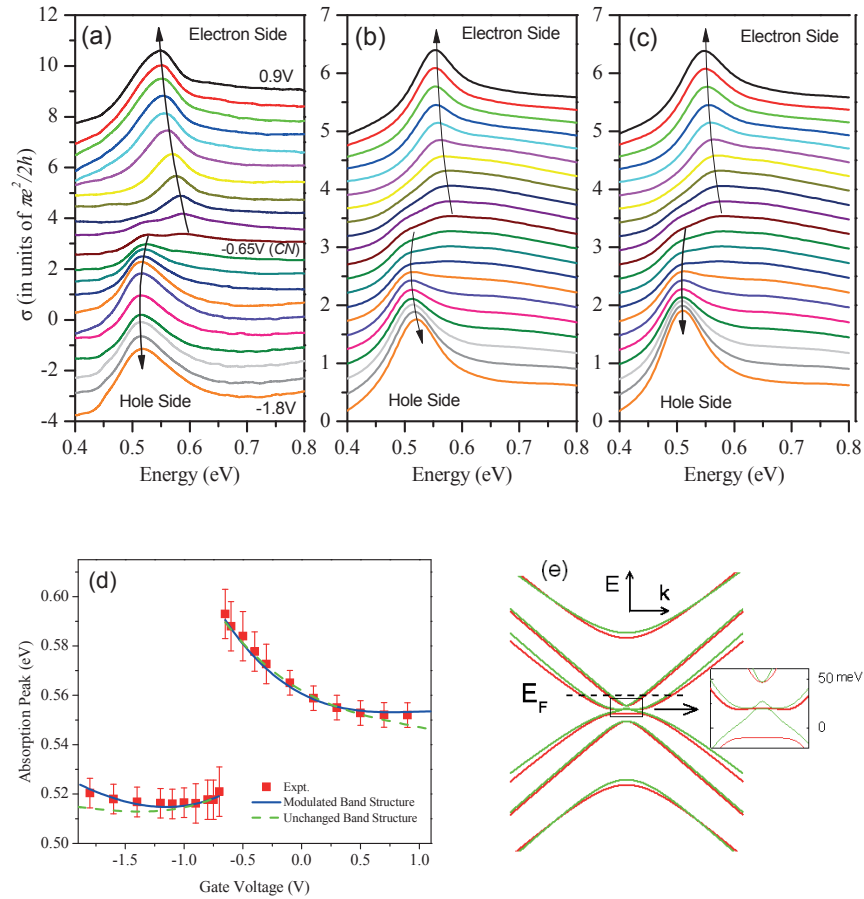


Figure S2 Comparison of the optical conductivity $\sigma(\hbar\omega)$ with theory for gated ABA-stacked graphene trilayers under both electron and hole doping. **a**, The experimental conductivity spectra $\sigma(\hbar\omega)$ as a function of gate voltage V_g . The spectra are shifted for 0.6 units for clarity. The arrows are guides for peak positions. For charge neutrality, $V_g = V_{CN} = -0.65$ V. From the top to bottom, V_g varies as 0.9, 0.7, 0.5, 0.3, 0.1, -0.1, -0.3, -0.4, -0.5, -0.6, -0.65(CN), -0.7, -0.75, -0.8, -0.9, -1.0, -1.1, -1.2, -1.4, -1.6 and -1.8 V. **b-c**, Simulated spectra for $\sigma(\hbar\omega)$ from the Kubo formula for the same conditions. The different spectra are shifted by 0.25 units for clarity. The results in (b) include the predicted modification of the band structure, while (c) is a reference calculation in which the band structure is assumed to remain unchanged. **d**, Positions of absorption peaks extracted from (a) as a function of V_g . The solid blue and dashed green lines are from the spectra calculated in (b) and (c), respectively. **e**, The calculated band structure for ABA trilayer graphene with finite values (green) and vanishing values (red) for the TB parameters δ and γ_4 .

References

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