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Dirac charge dynamics in graphene by infrared spectroscopy

Z. Q. Li^{1*}, E. A. Henriksen², Z. Jiang^{2,3}, Z. Hao⁴, M. C. Martin⁴, P. Kim², H. L. Stormer^{2,5,6}, and D. N. Basov¹

¹ Department of Physics, University of California, San Diego, La Jolla, California 92093, USA

² Department of Physics, Columbia University, New York, New York 10027, USA

³ National High Magnetic Field Laboratory, Tallahassee, Florida 32310, USA

⁴ Advanced Light Source Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

⁵ Department of Applied Physics and Applied Mathematics, Columbia University, New York, New York 10027, USA

⁶ Bell Labs, Alcatel-Lucent, Murray Hill, New Jersey 07974, USA

*e-mail: zhiqiang@physics.ucsd.edu

A remarkable manifestation of the quantum character of electrons in matter is offered by graphene, a single atomic layer of graphite. Unlike conventional solids where electrons are described with the Schrödinger equation, electronic excitations in graphene are governed by the Dirac Hamiltonian¹. Some of the intriguing electronic properties of graphene, such as massless Dirac quasiparticles with linear energy-momentum dispersion, have been confirmed by recent observations²⁻⁵. Here we report an infrared (IR) spectromicroscopy study of charge dynamics in graphene integrated in gated devices. Our measurements verify the expected characteristics of graphene and, owing to the previously unattainable accuracy of IR experiments, also uncover significant departures of the quasiparticle dynamics from predictions made for Dirac fermions in idealized, free standing graphene. Several observations reported here indicate the relevance of many body interactions to the electromagnetic response of graphene.

We investigated the reflectance $R(\omega)$ and transmission $T(\omega)$ of graphene samples on a SiO_2/Si substrate (inset of Figure 1(a)) as a function of gate voltage V_g at 45K (see Methods). We start with data taken at the charge neutrality point V_{CN} : the gate voltage corresponding to the minimum DC conductivity and zero total charge density (inset of Fig. 1(c)). Figure 1(a) depicts $R(\omega)$ of a graphene gated structure (graphene/ SiO_2/Si) at $V_{\text{CN}}=3\text{V}$ normalized by reflectance of the substrate $R_{\text{sub}}(\omega)$. $R_{\text{sub}}(\omega)$ is dominated by a minimum around 5500 cm^{-1} due to interference effects in SiO_2 . A remarkable observation is that a monolayer of undoped graphene dramatically modifies the interference minimum of the substrate leading to a suppression of $R_{\text{sub}}(\omega)$ by as much as 15%. This observation is significant because it allows us to evaluate the conductivity of graphene near the interference structure, as will be discussed below.

Both reflectance and transmission spectra of graphene structures can be modified by a gate voltage. Figure 1 (b) and (c) display these modifications at various gate voltages normalized by data at V_{CN} : $R(V)/R(V_{CN})$ and $T(V)/T(V_{CN})$, where $V = V_g - V_{CN}$. These data correspond to the Fermi energy E_F on the electron side and similar behavior was observed with E_F on the hole side (not shown). At low voltages ($<17V$) we found a dip in $R(V)/R(V_{CN})$ spectra. With increasing bias this feature evolves into a peak-dip structure and systematically shifts to higher frequency. The $T(V)/T(V_{CN})$ spectra reveal a peak at all voltages, which systematically hardens with increasing bias. A voltage-induced increase in transmission ($T(V)/T(V_{CN}) > 1$) signals a decrease of the absorption with bias. Most interestingly, we observed that the frequencies of the main features in $R(V)/R(V_{CN})$ and $T(V)/T(V_{CN})$ all evolve approximately as \sqrt{V} .

In order to explore the quasiparticle dynamics under applied voltages, it is imperative to discuss first the two dimensional (2D) optical conductivity of charge neutral graphene, $\sigma_1(\omega, V_{CN}) + i\sigma_2(\omega, V_{CN})$, extracted from a multilayer analysis of the devices (see Methods). Theoretical analysis⁶⁻⁸ predicts a constant “universal” 2D conductivity $\sigma_1(\omega, V_{CN}) = \pi e^2/2h$ for ideal undoped graphene. Our $R(\omega)/R_{sub}(\omega)$ data are consistent with this prediction. Fig. 1(a) shows a comparison between experimental $R(\omega)/R_{sub}(\omega)$ spectrum and model spectra generated assuming constant $\sigma_1(\omega, V_{CN})$ values. The constant universal conductivity offers a good agreement (within $\pm 15\%$) with the experimental spectra in the range $4000-6500 \text{ cm}^{-1}$. Outside of this spectral region, our IR measurements do not allow us to unambiguously determine the absolute value of $\sigma_1(\omega, V_{CN})$; therefore the uncertainty of $\sigma_1(\omega, V_{CN})$ increases as shown by the shaded region weighted around the $\pi e^2/2h$ value. However, recent IR studies of graphene revealed a constant conductivity $\sigma_1(\omega, V_{CN}) = \pi e^2/2h$ between 2400 and 24000 cm^{-1} (Ref.[9] and Mak, K.F. & Heinz, T. 2008 APS March Meeting, Abstract: L29.00006, unpublished). The universal conductivity is only weakly modified in bulk highly ordered pyrolytic graphite¹⁰ (HOPG) and extends down to 800 cm^{-1} . Thus in the following discussion, we will assume $\sigma_1(\omega, V_{CN}) = \pi e^2/2h$ throughout the entire range of our data.

Electrostatic doping of graphene introduces two fundamental changes in the optical conductivity $\sigma_1(\omega, V) + i\sigma_2(\omega, V)$: a strong Drude component formed in the far-IR with $\sigma_1(\omega \rightarrow 0) = 4-100 \pi e^2/2h$ accompanied by a shifting of the onset of interband transitions at $2E_F$, as schematically shown in the inset of Fig. 2(b). In order to investigate these effects, we obtained $\sigma_1(\omega, V) + i\sigma_2(\omega, V)$ (Fig. 2 (b,c)) from voltage-dependent reflectance and transmission spectra (see Methods). The key features in the conductivity spectra are independent of uncertainties in $\sigma_1(\omega, V_{CN})$ discussed above. Regardless of the choice of $\sigma_1(\omega, V_{CN})$, under applied biases we observe a suppression of the conductivity compared to $\sigma_1(\omega, V_{CN})$ and a well-defined threshold structure above which the conductivity recovers the universal value $\pi e^2/2h$. The energy of the threshold structure systematically increases with voltage, a natural expectation for a transition occurring at $2E_F$. With a scattering rate $1/\tau = 30 \text{ cm}^{-1}$ at $71V$ independently obtained from transport data, the Drude mode is rather narrow and confined below the low- ω cut-off of our measurements. We stress that the two voltage-induced transformations of the conductivity, the intraband mode and the onset of interband absorption at $2E_F$, are interdependent as suggested by

our data. Indeed, assuming the intraband component can be described with a simple Drude formula $\sigma_1(\omega) = \sigma_{DC} / (1 + \omega^2 \tau^2)$ using σ_{DC} and $1/\tau$ obtained from transport measurements, we find that the spectral weight removed from $\omega < 2E_F$ is recovered under the Drude structure, such that the total oscillator strength given by $\int_0^{\Omega_c} \sigma_1(\omega) d\omega$ is conserved at any bias with a cutoff frequency $\Omega_c = 8000 \text{ cm}^{-1}$.

Next we extracted Fermi energy values from the $2E_F$ threshold using two different methods (see Methods). We found that the $2E_F$ values (Fig. 3(a)) are symmetric for biases delivering either holes or electrons to graphene. Moreover, $2E_F$ increases with voltage approximately as \sqrt{V} (deviations from the square root law at small biases will be discussed below). Note that E_F of Dirac fermions scales with the 2D carrier density N as $E_F = \hbar v_F \sqrt{\pi N}^{2,3}$, where v_F is the Fermi velocity. In our devices, $N = C_g V / e$ where $C_g = 115 \text{ aF}/\mu\text{m}^2$ is the gate capacitance per unit area. Therefore, the observed \sqrt{V} dependence of $2E_F$ substantiates that graphene samples integrated in gated devices are governed by Dirac quasiparticles.

Interestingly, the $2E_F$ threshold in $\sigma_1(\omega, V)$ shows a width of about 1400 cm^{-1} that is independent of gate voltage and therefore of carrier density N , irrespective of a seven-fold enhancement of N between 10 V and 71 V . This effect is much stronger than the theoretical estimate for thermal smearing of the $2E_F$ feature at $45 \text{ K}^{7,8}$, which is about 500 cm^{-1} . A recent theoretical study¹¹ showed that disorder effects and electron-phonon coupling are needed to account for the width of the $2E_F$ threshold in our data. Apart from that, a spatial variation of local E_F values observed in graphene on SiO_2/Si substrates (Ref.[12] and Brar, V. et al., 2008 APS March Meeting, Abstract: U29.00003, unpublished) will inevitably lead to a broadening of the absorption onset at $2E_F$ in $\sigma_1(\omega)$, because IR measurements register the absorption averaged over a large area (a few microns in our experiments). The origin of the inhomogeneity of E_F in graphene is still an open question¹², which needs to be explored using spatially resolved probes such as near field IR conductivity studies capable of probing the response of a material with nanometer resolution over a large area¹³.

Our study has uncovered several new properties of graphene that are beyond the ideal Dirac fermion picture¹⁴. First, our study revealed unexpected features of $\sigma_1(\omega, V)$ below $2E_F$. The band structure of ideal graphene implies that the interband transition at $2E_F$ is the lowest electronic excitation in the system apart from the Drude response at $\omega=0$. Therefore, one anticipates finding $\sigma_1(\omega, V) \approx 0$ up to the $2E_F$ threshold, provided the Drude scattering rate is much smaller than $2E_F$. This latter condition is fulfilled for all data in Fig.2, and yet we registered significant conductivity below $2E_F$ (see supplementary information). This result has not been anticipated by theories developed for Dirac Fermions⁶⁻⁸. Both extrinsic and intrinsic effects may give rise to the residual conductivity in Fig. 2. Among the former, charged impurities and unitary scatterers (edge defects, cracks, vacancies, etc) were shown to induce considerable residual conductivity below $2E_F$ ¹¹. However, the theoretical residual absorption in Ref. [11] is systematically suppressed with voltage, whereas this suppression was not observed in our data. In

addition, the magnitude of the theoretical residual absorption is smaller compared to experimental values in Fig. 2. Therefore, it is likely that other mechanisms are also responsible for the residual conductivity in our data. One intriguing interpretation of the residual conductivity is in terms of many body interactions, which are known to produce a strong frequency dependent quasiparticle scattering rate $1/\tau(\omega)$. It is predicted theoretically that $1/\tau(\omega)$ in graphene increases with frequency due to electron-electron^{15, 16} and electron-phonon interactions¹⁷. The energy dependent scattering rate initiates a marked enhancement of the conductivity compared to the Lorentzian form prescribed by the Drude model. Such an enhancement in mid-IR frequencies has been observed in many systems¹⁸⁻²⁰.

A closer inspection of the voltage dependence of the $2E_F$ feature uncovers marked departures from the behavior anticipated within a single particle picture of graphene. In order to highlight these deviations, we plot $2E_F$ as a function of the Fermi vector k_F based on $k_F = \sqrt{\pi N} = \sqrt{\pi C_g V / e}$, as displayed in Fig. 3(a). The $E_F(k_F)$ plot has a clear physical meaning: it is a direct representation of the band dispersion. We then examine the ratio $E_F / \hbar k_F$ that is directly related to the Fermi velocity v_F . The $E_F / \hbar k_F$ plot as a function of $V^{1/2}$ and k_F in Fig. 3(b) reveals a departure from linear dispersion with a single value of $E_F / \hbar k_F$ expected within a single particle picture. Moreover, $E_F / \hbar k_F$ increases systematically with decreasing k_F values compared to that at high k_F values. The observed systematic enhancement of $E_F / \hbar k_F$ at low k_F is indicative of many body interactions^{14, 21, 22}. Signatures of band renormalization were also observed in a previous magneto-optical study of graphene⁴. Importantly, even the smallest $E_F / \hbar k_F$ values in Fig. 3(b) are higher than that of the bulk graphite²³ ($\sim 0.9 \times 10^6$ m/S), which also supports the hypothesis of $E_F / \hbar k_F$ renormalization in graphene. Complimentary information on the $E_F / \hbar k_F$ renormalization in graphene can be obtained from photoemission, which is another potent probe of many body effects in solids. Currently available photoemission data were all collected for epitaxial graphene grown on SiC^{24, 25}. This complicates a direct comparison with IR results for exfoliated samples on SiO₂/Si substrates reported here. We conclude by noting that the strong deviations of the experimental electromagnetic response from a simple single particle picture of graphene reported in our study challenge current theoretical conceptions of fundamental properties of this interesting form of carbon and also have implications for its potential applications in opto-electronics.

Methods:

Sample fabrication and infrared measurements

In the graphene devices studied here, monolayer graphene mechanically cleaved from Kish graphite was deposited onto an IR transparent SiO₂(300nm)/Si substrate^{2, 3}, which also serves as the gate electrode. Then standard fabrication procedures were used to define multiple Cr/Au (3/35 nm) contacts to the sample. The devices studied here exhibit

mobility as high as $8700 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ measured at carrier densities of $\sim 2 \times 10^{12} \text{ cm}^{-2}$. The characteristic half-integer quantum Hall effect is observed in these samples^{2,3}, confirming the single layer nature of our specimen. IR experiments were carried out using an IR microscope operating with synchrotron source at the Advanced Light Source (ALS) in the frequency range of $700\text{-}8000 \text{ cm}^{-1}$. The synchrotron beam is focused in a diffraction limited spot, which is smaller than the sample. We measured the reflectance $R(\omega)$ and transmission $T(\omega)$ of the graphene devices as a function of gate voltage V_g with simultaneous monitoring of the DC resistivity.

Temperature of the graphene sample

Data reported here were obtained in a micro-cryostat with sample mounted on a coldfinger in vacuum. The temperature of our graphene sample is warmer than that of the coldfinger, due to thermal radiation from room temperature KBr optical windows and electrical isolation of the devices from the coldfinger that compromises thermal contact. A sensor mounted in the immediate proximity to the Si substrate of the devices read $T=45 \text{ K}$ at the lowest temperatures attainable at the coldfinger. Because both the temperature sensor and the device are in nearly identical environment, we assumed this reading to be accurate for graphene as well.

Extracting the optical constants of graphene

The graphene device contains four layers: (1) graphene with 2D optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$, (2) SiO_2 gate insulator, (3) Si accumulation layer that forms at the interface of SiO_2/Si under the applied bias and (4) Si substrate. Properties of layers 2 and 4 are independent of the gate voltage whereas layers 1 and 3 are systematically modified by V_g . In our analysis of these multilayer structures we followed the protocol detailed in Reference [26]. Specifically, we carried out reflection, transmission, and ellipsometric measurements on the Si substrates and SiO_2/Si wafers used in our devices and thus obtained the optical constants of layers (2) and (4). We then investigated IR properties of test devices $\text{Ti}/\text{SiO}_2/\text{Si}$ as a function of gate voltage and thus extracted the optical constants of the Si accumulation layer in wafers used for graphene devices. We find that the response of the Si accumulation layer is confined to far-IR frequencies²⁷ and gives negligible contribution to mid-IR data in Fig. 1. Finally, we employed a multi-oscillator fitting procedure²⁶ to account for the contribution of $\sigma(\omega)$ of graphene to the reflectance and transmission spectra shown in Fig 1 using standard methods for multilayered structures.

Extracting Fermi energy E_F from conductivity spectra

Because of the broadening of the $2E_F$ threshold in $\sigma_1(\omega, V)$, the E_F values can be determined most accurately from the imaginary part of the optical conductivity spectra $\sigma_2(\omega, V)$ depicted in Fig. 2(c). Indeed, these spectra reveal a sharp minimum at $\omega = 2E_F$ in agreement with previous theoretical prediction²⁸. The minimum in $\sigma_2(\omega, V)$ spectrum is found from the frequency where the derivative of $\sigma_2(\omega, V)$ with respect to frequency is zero. The uncertainties of $2E_F$ obtained from this method are related to the accuracy in

defining the minimum in $\sigma_2(\omega, V)$ spectrum. Alternatively, $2E_F$ values can be extracted from the center frequency of the $2E_F$ threshold in $\sigma_1(\omega, V)$. The second method has larger uncertainties as shown in Fig. 3, due to the ambiguity of defining the center of the $2E_F$ threshold in $\sigma_1(\omega, V)$.

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Competing Financial Interests

The authors declare that they have no competing financial interests.

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Figures and Figure Legends:

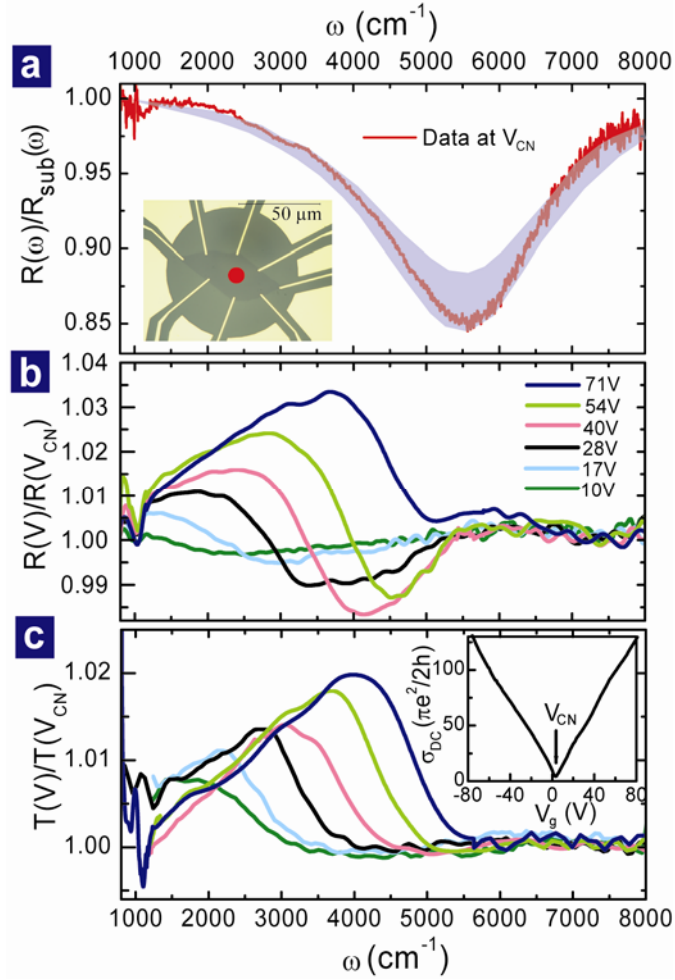


Figure 1: The reflectance $R(\omega)$ and transmission $T(\omega)$ of a graphene device under applied gate voltages. (a): the reflectance of the graphene device (graphene/SiO₂/Si) $R(\omega)$ normalized by that of the SiO₂/Si substrate $R_{\text{sub}}(\omega)$ at the charge neutrality voltage V_{CN} . A set of $R(\omega)/R_{\text{sub}}(\omega)$ spectra generated from the multilayer model using a constant $\sigma_1(\omega, V_{\text{CN}})$ in the range of $(1 \pm 0.15)\pi e^2/2h$ are shown as shaded area. The upper and lower boundary of the shaded area are defined by $\sigma_1(\omega, V_{\text{CN}})$ with values of $0.85 \cdot \pi e^2/2h$ and $1.15 \cdot \pi e^2/2h$, respectively. Inset of (a): a photograph of a graphene device together with a schematic of the focused synchrotron beam (red dot). (b) and (c): $R(V)/R(V_{\text{CN}})$ and $T(V)/T(V_{\text{CN}})$ spectra of the graphene device at several voltages corresponding to E_F on the electron side, where $V = V_g - V_{\text{CN}}$. Inset of (c): the smoothed DC conductivity data of the sample as a function of gate voltage V_g .

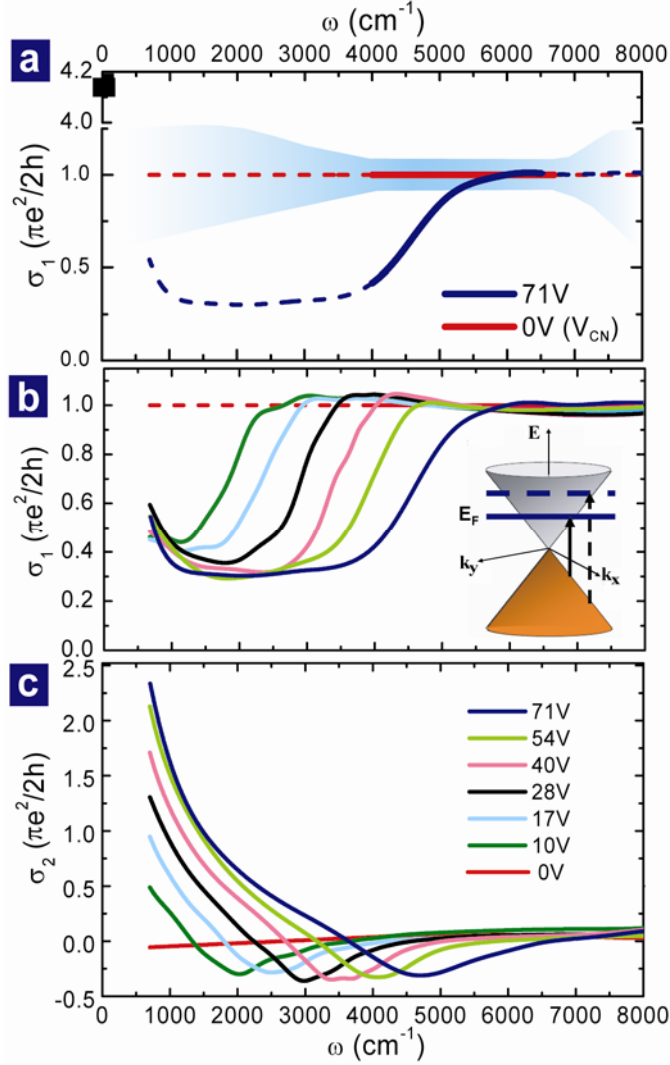


Figure 2: The optical conductivity of graphene at different voltages. (a), the real part of the 2D optical conductivity $\sigma_1(\omega)$ at V_{CN} and 71V. The solid red line displays the region where our data support the universal result. The uncertainty of $\sigma_1(\omega, V_{\text{CN}})$ is shown by the shaded area with the theoretical $\sigma_1(\omega) = \pi e^2/2h$ plotted as dashed line. The blue dashed line is $\sigma_1(\omega)$ at 71V evaluated for the theoretical spectra: $\sigma_1(\omega, V_{\text{CN}}) = \pi e^2/2h$ (red dashed line). The key spectral features of $\sigma_1(\omega, V)$ are independent of uncertainties in $\sigma_1(\omega, V_{\text{CN}})$ indicated by the shaded area, as discussed in the text. Black square on the left axis: DC conductivity at V_{CN} . (b) and (c), $\sigma_1(\omega)$ and $\sigma_2(\omega)$ of graphene at several voltages with respect to V_{CN} corresponding to E_F on the electron side based on $\sigma_1(\omega, V_{\text{CN}}) = \pi e^2/2h$. The absolute values of the $\sigma_2(\omega)$ spectra in (c) have uncertainties due to the uncertainties of $\sigma_1(\omega, V_{\text{CN}})$ as discussed in the text. Inset of (b), the band structure of graphene near the Dirac point and the interband transition at $2E_F$.

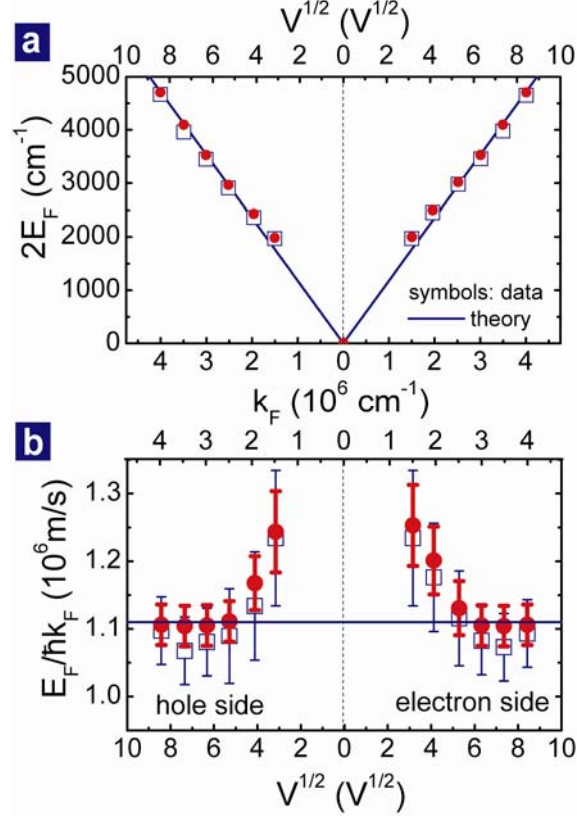


Figure 3: The Fermi energy E_F and the ratio of E_F to the Fermi wave vector $E_F / \hbar k_F$. The magnitude of $E_F / \hbar k_F$ is closely related to the Fermi velocity v_F as discussed in the text. (a), The magnitude of $2E_F$ plotted as a function of $V^{1/2}$ and k_F for the electron and hole sides with respect to the charge neutrality voltage. Red solid symbols: $2E_F$ extracted from the minimum in the $\sigma_2(\omega, V)$ spectra. Blue open symbols: $2E_F$ extracted from the center of the $2E_F$ threshold in $\sigma_1(\omega, V)$. The uncertainties of the $2E_F$ values discussed in the Methods section do not exceed the size of the symbols. Solid lines are theoretical $2E_F$ values using $v_F = 1.11 \cdot 10^6 \text{ m/s}$. (b), Symbols: $E_F / \hbar k_F$ as a function of $V^{1/2}$ and k_F . Blue line corresponds to a v_F value of $1.11 \cdot 10^6 \text{ m/s}$. The error bars reflect uncertainties in the determination of the Fermi energy $2E_F$ from the conductivity data $\delta(E_F)$ discussed in the Methods section and are calculated as $\delta(E_F / \hbar k_F) = \delta(E_F) / (\hbar \sqrt{\pi C_g V} / e)$.

Supplementary Information for

Dirac charge dynamics in graphene by infrared spectroscopy

Z. Q. Li^{1*}, E. A. Henriksen², Z. Jiang^{2,3}, Z. Hao⁴, M. C. Martin⁴, P. Kim², H. L. Stormer^{2,5,6}, and D. N. Basov¹

¹ Department of Physics, University of California, San Diego, La Jolla, California 92093, USA

² Department of Physics, Columbia University, New York, New York 10027, USA

³ National High Magnetic Field Laboratory, Tallahassee, Florida 32310, USA

⁴ Advanced Light Source Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

⁵ Department of Applied Physics and Applied Mathematics, Columbia University, New York, New York 10027, USA

⁶ Bell Labs, Alcatel-Lucent, Murray Hill, New Jersey 07974, USA

*e-mail: zhiqiang@physics.ucsd.edu

Spectral features in the raw reflectance/ transmission data and their connection to the broadening of the $2E_F$ threshold and residual absorption of graphene.

As discussed in the text, our study has uncovered an anomalous width of the $2E_F$ threshold and a strong residual absorption below $2E_F$ in the conductivity spectra of monolayer graphene. It is straightforward to relate both effects to features in the raw data. In Fig. S1 we compare the raw $R(V)/R(V_{CN})$ and $T(V)/T(V_{CN})$ spectra (blue curves) with similar spectra generated from a model $\sigma_1(\omega)$ spectrum for ideal graphene (black curves). The top panel details the input for these model calculations. In this panel we plot with the black line the conductivity of ideal graphene $\sigma_1(\omega)$ obtained using an analytical expression for the optical constants of graphene derived by Gusynin et al.¹:

$$\sigma_1(\Omega) = \frac{e^2 N_f}{2\pi^2 \hbar} \int_{-\infty}^{\infty} d\omega \frac{[n_F(\omega) - n_F(\omega')]}{\Omega} \frac{\pi}{4\omega\omega'} \left[\frac{2\Gamma(\omega)}{\Omega^2 + 4\Gamma^2(\omega)} - \frac{2\Gamma(\omega)}{(\omega + \omega')^2 + 4\Gamma^2(\omega)} \right] (|\omega| + |\omega'|)(\omega^2 + \omega'^2)$$

where $\omega' = \omega + \Omega$, $n_F(\omega) = \frac{1}{e^{(\omega - E_F)/T} + 1}$ is the Fermi distribution, $N_f = 2$ is the spin degeneracy, and $\Gamma(\omega)$ is an impurity scattering rate. A constant scattering rate Γ is used in the theoretical formula. In order to facilitate comparison with our mid-IR data, we have set the Fermi energy $2E_F = 5600 \text{ cm}^{-1}$. By setting the scattering rate to $\Gamma = 1 \text{ cm}^{-1}$ and temperature to $T = 45 \text{ K}$, we are able to model the threshold structure at $2E_F$ influenced by thermal broadening representing experimental conditions. We utilized the above equation in the interband region and in order to account for the free carrier response we augmented this result with the Drude Lorentzian $\sigma_1(\omega) = \sigma_{DC} / (1 + \omega^2 \tau^2)$, where $\sigma_{DC} = 100 * \pi e^2 / 2\hbar$ and a scattering rate $1/\tau = 30 \text{ cm}^{-1}$ is obtained from the transport data for our device. The

model $R(V)/R(V_{CN})$ and $T(V)/T(V_{CN})$ spectra were calculated based on the input $\sigma_1(\omega, V_{CN})$ spectrum in Fig. S1(a) using the procedure described in the Methods section. The dip-peak feature around 1000 cm^{-1} in all the experimental and model spectra in Fig. S1(b, c) is due to a phonon of SiO_2 . Vertical dashed lines in the plot show that the width $\delta 2E_F$ of the interband threshold in $\sigma_1(\omega)$ is determined by broadening of the high frequency edge in $T(V)/T(V_{CN})$. Furthermore, this connection was validated through calculations using different values of the phenomenological damping constant Γ . Thus with the guidance provided by modeling results in Fig.S-1 one can read the broadening of the $2E_F$ feature directly from the $T(V)/T(V_{CN})$ data and conclude that $\delta 2E_F \approx 1400 \text{ cm}^{-1}$ at all biases.

Model spectra are equally helpful for substantiating significant residual conductivity of graphene below the $2E_F$. For this purpose it is instructive to analyze the upper limit of the $T(V)/T(V_{CN})$ values at $\omega=2E_F$ corresponding to the maximum depletion of the conductivity under the applied bias. Our modeling shows that this upper limit is determined by the transmission of the graphene gated structure at the charge neutrality point $T(\omega, V_{CN})$ and the transmission of the Si substrate $T_{\text{sub}}(\omega)$ as $T(\omega, V_{CN})/T_{\text{sub}}(\omega)$, where $T_{\text{sub}}(\omega)$ is obtained from IR measurements and $T(\omega, V_{CN})$ is calculated from the multi-layer model using the theoretical universal conductivity $\sigma_1(\omega, V_{CN}) = \pi e^2/2h$ for graphene. Provided the residual conductivity is vanishingly small, the peak in $T(V)/T(V_{CN})$ spectra at $\omega=2E_F$ reaches the upper boundary. Under these latter conditions the amplitude of peaks in a series of spectra generated for different biases will trace the boundary of the shaded region in Fig.S-1(b). However, if the depletion of the conductivity at $\omega < 2E_F$ is incomplete, the residual absorption will reduce the amplitude of $T(V)/T(V_{CN})$ below the upper limit.. This is indeed the case for the experimental spectrum in Fig.S1-(b) taken at $V=71 \text{ V}$ and for the entire data set in Fig.1. Similarly, the amplitude of changes of reflectance is also reduced by the residual conductivity (Fig.S1-c). We note that deviations between experimental and model spectra is significant compared to the signal-to-noise of our measurements.

Here we stress that the magnitude of $\sigma_1(\omega, V)$ below the $2E_F$ threshold is sensitive to ambiguities with the choice of $\sigma_1(\omega, V_{CN})$. An assumption of the universal value for $\sigma_1(\omega, V_{CN})$ implies that the residual conductivity is as strong as $0.3 \cdot \pi e^2/2h$. Within limitations of our measurements we cannot unambiguously rule out $\sigma_1(\omega, V_{CN}) < \pi e^2/2h$ at energies below 4000 cm^{-1} and dependent on the input for $\sigma_1(\omega, V_{CN})$ the residual values at $\omega < 2E_F$ may significantly vary. Within these constrains, our data indicate either a breakdown of the universal conductivity $\sigma_1(\omega, V_{CN}) = \pi e^2/2h$ or significant residual conductivity below $2E_F$ at finite doping. Note that other experimental studies²⁻⁴ attest to the validity of $\sigma_1(\omega, V_{CN}) = \pi e^2/2h$ assumption in the entire mid-IR, which implies strong residual absorption below the $2E_F$ cut-off that is nearly independent of the applied voltage.

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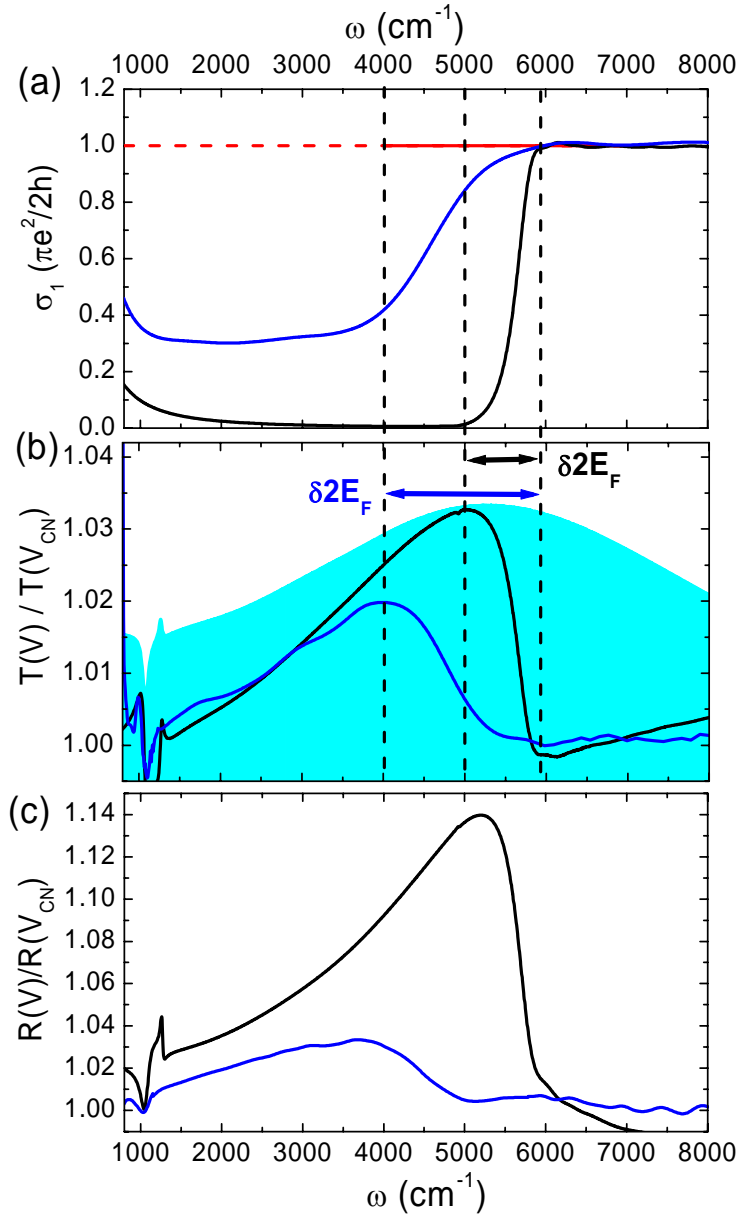


Figure S1: (a), the theoretical 2D optical conductivity $\sigma_1(\omega, V_{\text{CN}})$ at the charge neutrality point V_{CN} (red curve) and experimental $\sigma_i(\omega)$ spectrum at 71V (blue curve), together with a model $\sigma_i(\omega, 71 \text{ V})$ (black curve). The model spectrum reveals narrow width of the $2E_F$ threshold and negligible residual conductivity below $2E_F$. (b) and (c): experimental $R(V)/R(V_{\text{CN}})$ and $T(V)/T(V_{\text{CN}})$ spectra at 71V (blue spectra) and model data corresponding to the conductivity in (a) (black spectra). The upper boundary of the shaded region in (b) is the upper limit of $T(V)/T(V_{\text{CN}})$ values for different biases of our devices as described in the text. .