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Two-pulse space-time photocurrent correlations at graphene *p-n* junctions reveal hot carrier cooling dynamics near the Fermi level

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Abstract. Two-pulse excitation at a graphene *p-n* junction generates a time-dependent photocurrent response that we show functions as a novel ultrafast thermometer of the hot electron temperature, $T_e(t)$. The extracted hot electron cooling rates are consistent with heat dissipation near the Fermi level of graphene occurring by an acoustic phonon supercollision mechanism.

1 Introduction

With uniform broad spectral coverage, fast response and high carrier mobility, graphene based *p-n* junctions are a promising new material for next generation optoelectronics such as photodetectors, bolometers and plasmonic devices. The response of such devices depends critically on the hot electron gas temperature and its corresponding heat dissipation rate [1,2]. Below a threshold energy of ~200 meV relative to the Fermi energy, electrons are predicted to dissipate heat predominately by the emission of acoustic phonons [2]. However, the energy dissipated by acoustic phonons emission is very small owing to the large mismatch between the slow sound velocity of the phonons and the fast Fermi velocity of the electrons. As a consequence, a cooling bottleneck has been predicted with long relaxation times, exceeding ~300 ps [2]. Thus far, empirical measurements of the hot electron cooling rate have been limited to transient absorption (TA) spectroscopy. While such measurements exhibit faster cooling kinetics, the transient signal remains convoluted by many competing contributions including electron thermalization, optical phonon emission and intraband induced absorption [2,3]. To enhance our sensitivity to the desired cooling processes near the Fermi level, we require a technique that is independent of the spectral probe window and directly measures the hot electron temperature.

Recently, it was shown that photocurrent (PC) generated at graphene *p-n* junctions can be used as a thermometer of the hot electron temperature (T_e) using the photothermal effect.[1,7] To capture the timescales of hot electron cooling, we show how the PC generated by a femtosecond two-pulse excitation serves as a novel ultrafast thermometer of the transient electron temperature, $T_e(t)$. Unlike existing measurements, the resulting transient photocurrent (TPC) kinetics are approximately independent of the excitation wavelength, and directly measure the temperature hot electronic carriers near the Fermi energy.

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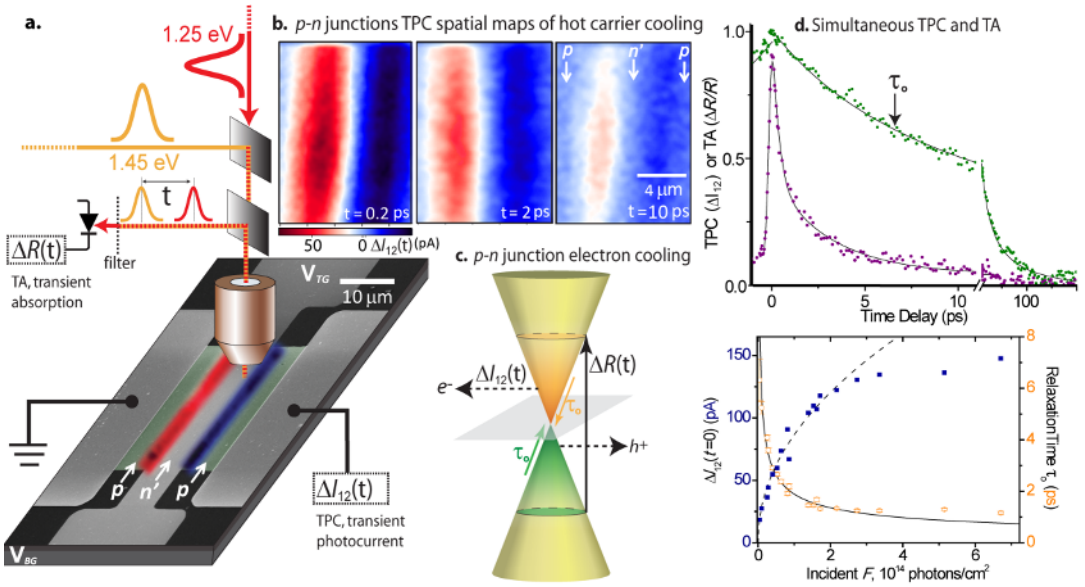

Fig. 1. (a) SEM image of a graphene (green, false color) dual *p-n* junction device. The two-color, two-pulse correlation geometry shown measures: (*i*) $\Delta R(t)$, mostly Pauli blocking of probe beam by the hot electrons. (*ii*) $\Delta I_{12}(t)$, which serves as a photothermal thermometer of the hot electron temperature. **(b)** Spatial maps of $\Delta I_{12}(t,\boldsymbol{r})$ show the decay of the TPC signal with increasing pulse delay time, *t*. **(c)** Thermalized hot electrons relax with timescale τ_o. We collect both the reflectivity of the probe (black arrow) and the subsequent photothermal current generated (dashed arrows). **(d)** Simultaneously acquired hot electron kinetics show the fast absorptive ($\Delta R(t)/R$) and long TPC ($\Delta I_{12}(t)$) responses at the *p-n* junction. (lower panel) Extracted TPC amplitudes (square root fit, dashed line) and relaxation times τ_o (inverse square root fit, solid line) as a function of photon flux.

2 Experimental methods

We fabricate *p-n* junctions from large-grain graphene grown by the chemical vapor deposition (CVD) method with a device carrier mobility of ~8000 $cm^2V^{-1}s^{-1}$. A tunable back gate (*BG*) and top gate (*TG*) couple to graphene electrostatically, defining two *p-n* junctions where the PC production is maximal. The collected PC amplitude is plotted as the laser is raster scanned over the *p-n* junction (see superimposed map in Fig. 1a).

We optically excite the graphene *p-n* junction region with 180 fs pulses produced by two independently-tunable oscillators plus NIR optical parametric oscillator (OPO) system that are synchronously locked. We simultaneously collect the change in reflectivity ($\Delta R(t)/R$, TA) and electrical current generated ($\Delta I_{12}(t)$, TPC) as functions of pulse delay time (t) at a lattice temperature, T_1 = 10 K (Fig. 1a). $\Delta R(t)/R$ and $\Delta I_{12}(t)$ are also measured spatially by raster scanning the collinear laser pulses. Resulting TPC spatial maps are shown in Fig. 1b, and show the decay of the measured signal in time and space.

3 Results and Discussion

Similar to previous TA on bulk graphene, we find the transient bleach signal at the graphene *p-n* junction is fast and roughly biexponential (τ_1= 0.33 ps and τ_2=3.2 ps, see Fig. 1c). In stark contrast, the simultaneously acquired TPC signal decays roughly inversely with time with long tails extending out to 100 ps. Similar strong TPC responses have been observed for exfoliated graphene source-

drain[4], and graphene *p-n* junction [5] devices. Also unlike TA, we find the kinetic decay of $\Delta I_{12}(t)$ is approximately independent of both excitation energies investigated (from 0.82 to 1.55 eV) and pulse ordering (1.25 eV pump, 1.45 eV probe or vice versa). This insensitivity to the spectral excitation window, suggests that TPC originates from the hot electron temperature near the Fermi level. To test this observation, we apply the photothermal PC generation model to quantitatively extract the transient hot electron temperature. For a single-pulse excitation, we collect the time-dependent photothermal charge given by $Q_1 = \int_0^\infty \beta T_e(t)(T_e(t) - T_l)dt = \int_0^\infty i(t, T_o)dt$, where T_o is the initial hot temperature of the thermalized distribution, T_l is the lattice temperature and β is a proportionality constant. Using a two-pulse excitation scheme, we obtain the TPC response simply by integrating piecewise about our delay time *t*, giving:

$$Q_{12}(t) = \int_0^t i(t', T_o)dt' + \int_t^\infty i\left(t' - t, \sqrt{T_o^2 + T_e^2(t)}\right)dt'. \quad (1)$$

The above TPC response function, shows how $T_e(t)$ can be extracted from the measured charge, $Q_{12}(t)$ providing that the underlying hot electron cooling rate law is nonlinear.

We find the PC generated at the *p-n* junction increases nonlinearly with the square root of photon flux, and decays on a much faster timescale than acoustic phonon emission model predcit(see Fig. 1d) [2,7]. Alternatively, Song *et al.* predicts that impurities and lattice disorder can relax the momentum conservation constraint, resulting in a more rapid energy relaxation (~1-10 ps); they call this process supercollisions (SC), and its signature is given by the cooling rate law [6]:

$$\frac{dT_e}{dt} = -A\frac{(T_e^3 - T_l^3)}{T_e}. \quad (2)$$

To date, however, this theory has not been tested. For low lattice temperatures ($T_e >> T_l \approx 10$K), the above rate law gives $T_e(t)=T_o/(1+t/\tau_o)$ and predicts that cooling rate, $\tau_o^{-1}=AT_o$ varies with the square root of incident power. Substituting $T_e(t)$ into equation (1), we fit our TPC data (solid line Fig. 1d) and extract A=4.9x10^8 K^{-1}s^{-1} [7]. Excellent fits to the kinetic decay across a wide range of electronic temperatures supports a dominant mechanism of SC-assisted hot electron cooling. In Fig. 1d, we further show the extracted hot electron cooling time (τ_o) vary from 1 to 6 ps, and scale with the inverse square root of incident laser flux, as predicted. We find we can accurately predict the PC response in graphene by using the SC cooling model [6] and our TPC response function [7]. This suggests a dominant disorder-assisted SC mechanism for hot electron cooling near the Fermi level.

4 Conclusions

We measure the dynamics of hot carrier cooling at a graphene *p-n* using a novel two-color, two-pulse excitation technique that provides time-resolved microscopy maps of both the absorptive (by TA, $\Delta R(\boldsymbol{r},t)/R$), and photocurrent (by TPC, $\Delta I_{12}(\boldsymbol{r},t)$) device response. We show the TPC kinetics are consistent with a recently predicted supercollision mechanism[6] of disorder-assisted acoustic phonons, resulting in greatly accelerated rates for hot electron cooling near the Femi energy.

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