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Phys. Rev. Lett. **117**, 257401 — Published 13 December 2016

DOI: [10.1103/PhysRevLett.117.257401](https://doi.org/10.1103/PhysRevLett.117.257401)

Tunable Ultrafast Thermal Relaxation in Graphene Measured by Continuous-Wave Photomixing

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Abstract

Hot electron effects in graphene are significant because of graphene's small electronic heat capacity and weak electron-phonon coupling, yet the dynamics and cooling mechanisms of hot electrons in graphene are not completely understood. We describe a novel photocurrent spectroscopy method that uses the mixing of continuous-wave lasers in a graphene photothermal detector to measure the frequency dependence and nonlinearity of hot-electron cooling in graphene as a function of the carrier concentration and temperature. The method offers unparalleled sensitivity to the nonlinearity, and probes the ultrafast cooling of hot carriers with an optical fluence that is orders of magnitude smaller than in conventional time-domain methods, allowing for accurate characterization of electron-phonon cooling near charge neutrality. Our measurements reveal that near the charge-neutral-point the nonlinear power dependence of the electron cooling is dominated by disorder-assisted collisions, while at higher carrier concentrations conventional momentum-conserving cooling prevails in the nonlinear dependence. The relative contribution of these competing mechanisms can be electrostatically tuned through the application of a gate voltage – an effect that is unique to graphene.

When graphene absorbs electromagnetic radiation, its electrons heat up and produce a measurable thermoelectric response, even at room temperature. Because of graphene’s gapless dispersion relation, small electron heat capacity, and anomalously weak electron-phonon coupling, this photothermal detection mechanism is broadband (from DC to visible), highly sensitive, and fast [1–5]. The speed, temperature-, and power-dependence of these detectors depend critically upon how fast and by what mechanisms the hot carriers relax [6–8]. Two primary cooling mechanisms have been identified: supercollision cooling, in which disorder-assisted scattering allows for non-momentum-conserving transitions, and conventional momentum-conserving electron-phonon cooling [6, 7, 9–14]. Evidence for conventional momentum-conserving cooling (which is linear with temperature) has been observed only at low temperatures in high-quality graphene[8]. In experimental measurements, the cooling process is inferred from how the photoresponse depends on temperature, power, or time for either pulsed or continuous-wave illumination. Time-domain methods that are used to study thermal relaxation dynamics typically employ intense optical pulses, which significantly disturb the electron temperature, and can in some cases excite higher energy optical phonons in addition to acoustic phonons [7, 15–17]. Moreover, as we show here, the factors that govern the power dependence of the photothermal response can be different from those that determine the cooling rate. It has been shown that, uniquely in graphene, the relative strength of the two competing cooling channels can be controlled by the carrier concentration [6, 8, 12].

Here we employ a new nonlinear photomixing method to simultaneously quantify the nonlinearity in the photoresponse and the carrier-density dependence of electron cooling in graphene. This method easily distinguishes between sub-linear and super-linear power dependence, which indicate supercollision cooling and conventional cooling, respectively. Our measurements show that while supercollision dominates the nonlinear response near the charge neutral point, at higher carrier densities, conventional cooling is the dominant contribution to the nonlinearity. Furthermore, we show that when two detuned near-IR lasers co-illuminate the graphene, the resulting DC photovoltage depends upon their heterodyne difference frequency. This enables direct measurement of the electron cooling rate in the frequency domain with orders of magnitude weaker optical excitation (smaller temperature rise) than traditional time-domain methods, by simply tuning the wavelength of one of the continuous-wave lasers.

Fig. 1a depicts the heterodyne photomixing setup used here to characterize the pho-

tothermal response of graphene. Two fiber-coupled continuous-wave near-IR lasers, one wavelength-tunable ($\lambda_1 = 1540\text{--}1565$ nm) and one fixed-wavelength ($\lambda_2 = 1545$ nm), were amplified, spatially combined, polarized, and focused using an aspheric lens to a $3\text{ }\mu\text{m}$ spot on the graphene channel. The position of focused beam was chosen to maximize the photovoltage, which occurs when the beam is focused close to one of the contacts [18, 19]. The combined optical power illuminating the first (second) device was about 6 mW (2.1 mW), from which we estimate the total absorbed intensity to be $I = 850\text{ W/cm}^2$ ($I = 300\text{ W/cm}^2$). The graphene photodetector device was held in a liquid helium cryostat with short working distance optical access to controllably vary the lattice temperature T_L between 10 K and room temperature. The two lasers were mechanically chopped using a twin-slot (5/7) chopping wheel at frequencies $f_1 = 500$ Hz and $f_2 = 700$ Hz. The photovoltage was synchronously detected at both f_1 and the difference $f_2 - f_1$, using a dual-reference digital lock-in amplifier (Signal Recovery 7270), which simultaneously records the photovoltages V_1 and V_Δ . The phases of the two lock-in detection channels were calibrated to produce the correct sign, relative to one another. Measurements were performed as a function of gate voltage V_G , and the optical difference frequency $\Delta\nu = \Omega/2\pi$, which was swept from -0.6 THz to $+2.5$ THz by tuning laser 1.

To better elucidate the role of disorder, we considered two different graphene detectors shown in Fig. 1b: one using an edge-contacted hexagonal boron nitride encapsulated graphene channel[20, 21], and a second fabricated from an un-encapsulated exfoliated flake[5]. The Supplementary Material details the fabrication and DC electrical characterization of the devices (section S4).

The electron temperature T in the graphene evolves according to the nonlinear differential equation [6, 9, 22]

$$\alpha T \frac{dT}{dt} + \beta_1(T - T_L) + \beta_3(T^3 - T_L^3) = I(t) \quad (1)$$

where T_L is the lattice temperature, αT is the specific heat of graphene carriers, the coefficients β_1 and β_3 are the rate coefficients for conventional and supercollision cooling mechanisms, respectively, and $I(t)$ is the absorbed near-infrared optical intensity. For the two-laser illumination shown in Fig. 1a, the absorbed intensity is $I(t) = I_1 + I_2 + 2\sqrt{I_1 I_2} \cos \Omega t$, where I_1 and I_2 represent the absorbed intensities of lasers 1 and 2 respectively and $\Omega \equiv 2\pi c(\lambda_2^{-1} - \lambda_1^{-1})$ is their heterodyne difference frequency.

It is assumed that the electrons are in the degenerate regime ($E_F \gg k_B T$), and that

electron-electron collisions are fast enough to allow the temperature of the electron gas to be well defined [9, 23]. The hot-electron diffusion length is $\xi = (\kappa/\gamma\alpha T)^{1/2} = v_F(\gamma\Gamma)^{-1/2}$, where κ is the electronic thermal conductivity, Γ is the carrier scattering rate, and the Wiedemann-Franz law was used in the second equality. Even for the encapsulated device considered here, by estimating Γ from DC measurements in Fig. S3, we estimate that $500 \text{ nm} < \xi < 1.5 \text{ }\mu\text{m}$ which is smaller than the optical beam size employed. We therefore ignore spatial inhomogeneity in $I(t)$ and thermal diffusion of hot carriers out of the laser beam.

The three model parameters α , β_1 and β_3 appearing in (1) depend implicitly on the Fermi level E_F (determined by gating) and disorder mean-free path l (related to the quality of the graphene) as $\alpha = 2\pi k_B^2 E_F / (3\hbar^2 v_F^2)$, $\beta_1 = V_D^2 E_F^4 k_B / (2\pi \rho \hbar^5 v_F^6)$, $\beta_3 = \zeta(3) V_D^2 E_F k_B^3 / (\pi^2 \rho \hbar^4 v_F^3 s^2 l)$, where v_F is the Fermi velocity, ρ is the areal mass density, s is the speed of sound in graphene, $\zeta(3) \simeq 1.202$ is the Riemann zeta function, and V_D is the acoustic deformation potential. We note that the substrate surface polar phonons may also play a role in hot electron cooling in graphene[24–26], and their effect on the photoresponse can be regarded as a linear cooling term (β_1) in (1)[27]. At temperatures far below the Bloch-Grüneisen temperature (1) must be modified to include a cooling term proportional to T^4 [22]. We estimate that even at the lowest temperatures and carrier concentrations experimentally considered here ($T = 25 \text{ K}$, $E_F \sim 60 \text{ meV}$), the measurement temperature matches or exceeds the Bloch-Grüneisen temperature.

The resulting photothermoelectric voltage V produced by the Seebeck effect is then related to the electron temperature by $V = rT(T - T_L)$, where rT is the Seebeck coefficient of graphene[2, 28]. This nonlinear relationship between temperature and photovoltage could be generalized to include a nonlinearity in the Seebeck coefficient[29, 30], but the temperature- and power-dependence of the observed nonlinearity indicate that this effect is small in comparison to the nonlinearity in cooling. Although other photoresponse mechanisms, such as photoelectric effect[31], might also contribute to the graphene photoresponse, a photo-thermo-electric model can adequately describe the photoresponse at the graphene-metal interface[5, 13, 32].

Equation (1) can be solved using a power series expansion (Supplementary Material),

and resulting DC photovoltage is found to be

$$V(I_1, I_2) = a_1(I_1 + I_2) + a_2(I_1^2 + I_2^2) - a_3(I_1^3 + I_2^3) \dots \\ + 2a_2I_1I_2 \left[1 + \frac{\gamma^2}{\Omega^2 + \gamma^2} \right] - 3a_3I_1I_2(I_1 + I_2) \left[1 + \frac{2\gamma^2}{\Omega^2 + \gamma^2} \right] \quad (2)$$

where $\gamma \equiv (\beta_1 + 3\beta_3T_L^2)/\alpha T_L$ is the linearized cooling rate from both mechanisms. The coefficients a_1 , a_2 and a_3 are given by

$$a_1 \equiv \frac{r}{\alpha\gamma}, \quad a_2 \equiv \frac{r\beta_1}{(\alpha\gamma T_L)^3}, \quad a_3 \equiv \frac{3r\beta_3^2}{T_L^2\alpha^5\gamma^5} \quad (3)$$

The final two terms in (2) which contain the factor I_1I_2 , represent a nonlinear interaction of the two beams, which occurs only when both beams are present. In order to sensitively detect only these mixing products, we employ a double-modulation configuration in which laser 1 is mechanically chopped at a frequency f_1 , laser 2 is chopped at f_2 , and the photovoltage V is synchronously detected using a lock-in amplifier at the chopping difference frequency $\Delta f \equiv f_1 - f_2$ (not to be confused with the heterodyne difference frequency). The resulting photovoltage $V_\Delta \equiv V(I_1, I_2) - V(I_1, 0) - V(0, I_2)$ can be positive or negative, depending on the nonlinearity in the photothermal response. We also simultaneously measure the Fourier component at f_1 , denoted $V_1 \equiv V(I_1, 0)$, which gives the photovoltage produced by laser 1 alone.

By simply comparing the magnitude of the two terms that compose the linearized cooling rate γ , one can determine a condition for which process makes the largest contribution to the cooling rate. For nearly all of the experimental cases considered here and reported elsewhere, the cooling rate is largely limited by the supercollision term. (3) reveals that despite this, the photoresponse can be either super-linear or sub-linear in intensity, depending on the carrier density and graphene quality. As explained below, neither cooling effect can be ignored when analyzing the nonlinearity of the response.

When the heterodyne frequency exceeds the cooling rate ($\Omega \gg \gamma$), (2) simplifies to $V(I) = a_1I + a_2I^2 - a_3I^3$, where $I \equiv I_1 + I_2$ is the total absorbed optical intensity. The quadratic and cubic terms have opposite sign, and therefore describe super-linear or sub-linear dependence on the optical intensity. From (3), one sees that the super-linear coefficient is proportional to β_1 , which we associate with momentum-conserving cooling, while the sub-linear coefficient is proportional to β_3 , which arises from supercollision cooling.

Fig. 2a plots V_1 (black) and V_Δ (green) as a function of the gate voltage for the HBN-encapsulated device. These measurements were performed with $\Omega/2\pi = 2.5$ THz, which is much faster than the expected cooling rate at room-temperature. The sign of the photothermal voltage V_1 depends on the gate voltage, as expected from the photothermoelectric effect [2, 3, 5]. For carrier densities near the charge-neutral point (CNP), V_1 and V_Δ have opposite sign (as indicated by the blue shading), revealing a sub-linear power dependence, characteristic of supercollision cooling [7, 13]. In this regime the Fermi surface is small, and the allowed phonon energy space for momentum-conserving collision is strongly constrained, thereby suppressing conventional electron-phonon cooling [6, 12]. At higher carrier densities, the behavior changes to super-linear (red shading), indicating that conventional cooling becomes stronger and dominates the photothermal nonlinearity. Fig. 2b plots the single-beam photovoltage as a function of the incident optical power, confirming the sub- and super-linear behavior, respectively. Fig. 2c illustrates the two cooling mechanisms schematically in k -space, along with the predicted sublinear and superlinear power dependence. The transitions outside of the Dirac cone represent supercollision cooling, in which the spatial disorder in the graphene compensates for the electron-phonon momentum mismatch.

The threshold between these two nonlinear regimes can be approximated by equating the opposing terms in V_Δ , which gives,

$$2a_2 \gtrless 3a_3I \quad (4)$$

where the upper and lower inequalities describe the conditions under which conventional cooling or supercollision cooling prevails in the nonlinear response, respectively. The relative importance of the two competing cooling channels depends on temperature, intensity, the carrier concentration (Fermi level), and indirectly on the material quality, which is related to the disorder mean-free-path l . Even though the linearized cooling rate γ is limited by supercollision cooling, both effects are evident in the nonlinear response reported here.

In the Supplementary Material (section S2), we present results of similar measurement performed on lower-mobility exfoliated graphene on SiO_2 . Similar to the HBN-encapsulated device, we observe an expected transition from supercollision cooling to conventional cooling. The transition happens around $E_F = 80$ meV, and we use (4) to determine the ratio of the two rate coefficients, $\beta_1/\beta_3 = 5300 \text{ K}^2$. At room temperature, the supercollision contribution to the cooling rate γ is nearly $50\times$ larger than the contribution from conventional cooling. Despite this, both effects have non-negligible role in the nonlinearity of the photoresponse,

and their relative significance depends on the carrier density.

When the heterodyne difference frequency Ω is comparable to or smaller than the cooling rate γ , the electron temperature can follow the interferometric beating of the two lasers, which produces a larger photothermal voltage than when the lasers are widely detuned. The final two terms in (2) reveal that the nonlinear mixing signal V_Δ exhibits a Lorentzian dependence on the heterodyne difference frequency Ω , with a spectral width that is proportional to the cooling rate γ . As before, the double-chopping configuration allows for sensitive detection of this heterodyne photomixing signal.

Fig. 3a plots the measured photovoltage V_Δ as a function of the gate voltage and heterodyne difference frequency, for the non-encapsulated graphene detector. In addition to the expected gate-voltage dependence discussed previously, the photoresponse exhibits distinct spectral peak around $\Omega = 0$. Fig. 3b shows the photomixing spectrum at a fixed gate voltage, along with the best-fit Lorentzian curve. From the linewidth, we estimate a cooling time of $\gamma^{-1} = 1.42$ ps, which is consistent with the time-domain pulse coincidence measurements [5, 33] reported for similar devices.

In order to confirm the thermal model for the photomixing, we repeated the heterodyne spectral measurements at temperatures from room temperature down to 25 K for the exfoliated sample on SiO_2 near the charge neutral point. As shown in Fig. 4a, in all cases the photomixing signal exhibits a Lorentzian spectral dependence, but with a spectral width that decreases with temperature, as expected. The solid blue curve in Fig. 4b shows a fit to the linearized cooling rate, based on the model presented here. For the data points above $T_L = 80\text{K}$, and for conditions at the charge neutral point, the assumption of $E_F \gg k_B T$ (degenerate regime) is no longer valid, which requires a modification of the cooling rate (Supplementary Material S3). We therefore excluded these points when fitting the blue curve. However, when the parameters from the low-temperature fit were incorporated into the modified thermal model, it correctly predicts the observed high-temperature asymptotic behavior, indicated by the red curve, with no additional free parameters.

This nonlinear heterodyne photovoltage spectroscopy method has two important advantages over the traditional time-domain measurement using pulse coincidence [5, 7, 33]: (i) The frequency range and resolution is limited only by the tuning range and resolution of the laser, while in time-domain measurements the response is limited by the optical pulsewidth and repetition period. (ii) Continuous-wave illumination produces a far smaller thermal

stimulus to the graphene electrons than intense ultrafast pulses, thereby allowing measurement of the temporal dynamics and nonlinearity of photodetection under low photothermal excitation for which the electron temperature is near the lattice temperature.

The model and measurements described here show that there are two competing cooling channels for hot electrons in graphene, and (4) describes the relative importance of each in the nonlinear response. In time-domain experiments reported elsewhere, the instantaneous absorbed intensity is orders of magnitude higher than the continuous-wave illumination considered here, in which case (4) predicts that supercollision cooling is the dominant contribution to the nonlinearity at all practically attainable doping concentrations. Moreover non-encapsulated graphene samples have a much smaller disorder mean-free-path l , which further contributes to the relative importance of supercollision cooling over conventional momentum-conserving cooling. In these cases, the photothermal response is often adequately described by supercollision cooling alone, for a wide range of carrier densities and temperatures [13, 34, 35]. For continuous-wave measurements on encapsulated devices, (4) also predicts that at sufficiently low temperatures, conventional cooling will prevail, consistent with temperature-dependent measurements reported recently[8].

We show that nonlinearity in the photothermoelectric effect causes photomixing when graphene is illuminated by near-infrared beams, and we describe a new heterodyne spectroscopy method that accurately measures this nonlinearity in the frequency domain. Exceedingly small nonlinearities in the photoresponse can be probed using continuous-wave illumination, which accurately elucidates the physical mechanisms behind the nonlinearity and cooling. In particular, the measurements reveal the role that disorder plays in the cooling of hot electrons, and the interplay of different cooling channels at different carrier concentrations. The method permits direct measurement of the cooling rate in graphene using swept laser spectroscopy, which offers several advantages over pump-probe or pulse-coincidence measurements. The work also implies that nonlinear photomixing in graphene is very promising for the development of new optical/THz photomixing devices.

This work was sponsored by the US ONR (N000141310865) and the US NSF (ECCS 1309750). C.T. was supported by a DOD-AFOSR, NDSEG fellowship under contract FA9550-11-C-0028, 32 CFR 168a. C.T. and J.H. acknowledge financial support from the Nanoelectronics Research Initiative (NRI) through the Institute for Nanoelectronics Discovery and Exploration (INDEX). K.W. and T.T. acknowledge support from the Elemental Strat-

egy Initiative conducted by the MEXT, Japan and a Grant-in-Aid for Scientific Research on Innovative Areas “Science of Atomic Layers” from JSPS. M.S.F. was supported in part by an ARC Laureate Fellowship.

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FIGURES

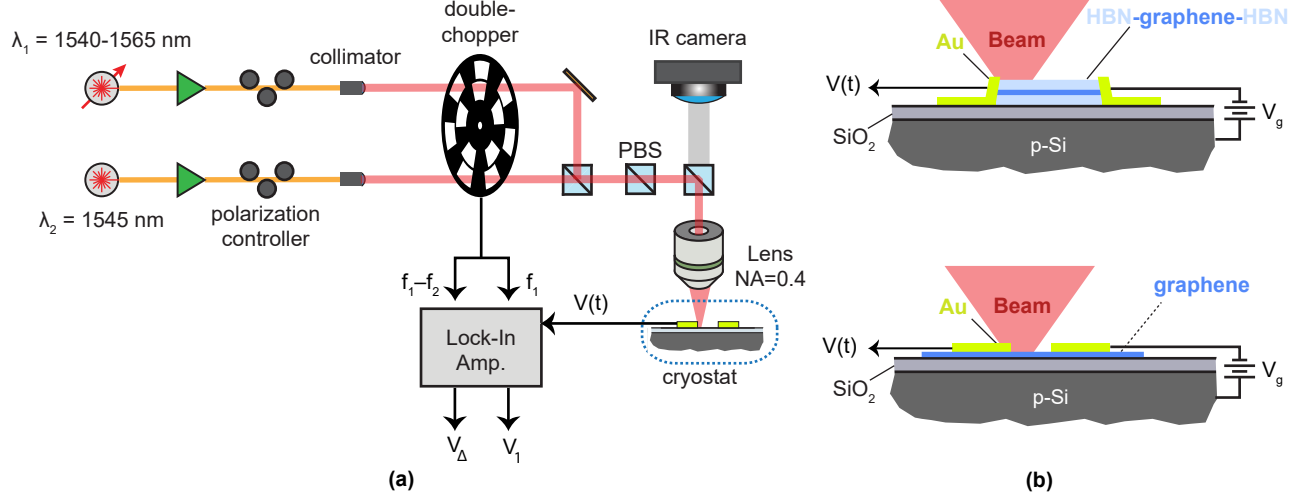


FIG. 1. Diagram of heterodyne photomixing experiment. (a) Two near-IR continuous-wave beams, one with tunable wavelength, are modulated at two different frequencies f_1 and f_2 , then overlapped and focused down to the graphene photodetector. The photovoltages produced by laser 1 (V_1) and the mixing of two beams (V_Δ) are detected at the modulation frequencies f_1 and $f_1 - f_2$ respectively. (b) Diagram of the HBN-encapsulated graphene (top) and exfoliated graphene (bottom) photodetector devices. The optical beams are illuminated close to one of the metal contacts, and carrier density of graphene is altered by applying an electrostatic voltage (V_g) between doped silicon substrate and graphene.

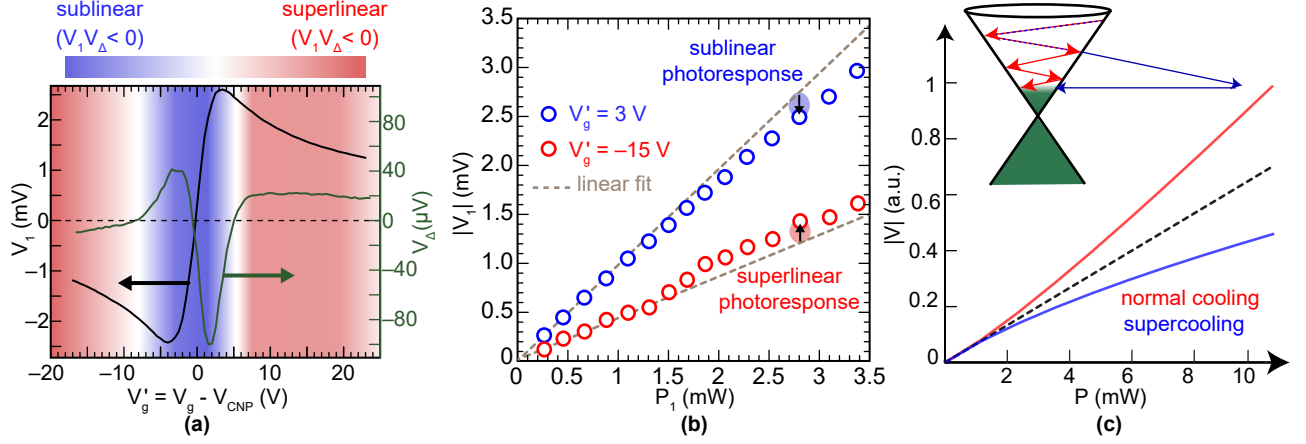


FIG. 2. Photovoltage vs. carrier concentration and incident power, measured at room temperature (a) Single-laser photovoltage (V_1), and nonlinear photomixing signal (V_Δ) measured vs. the applied gate voltage V_g . V_{CNP} denotes the charge neutral point. Red ($V_1 V_\Delta > 0$) and blue ($V_1 V_\Delta < 0$) regions indicate the conditions where super- and sub-linear power dependence is observed. (b) Measured photovoltage V_1 of a single laser vs. incident optical power at two different gate voltages, showing sublinear and superlinear behavior. (c) Calculated photothermoelectric voltage (in arbitrary units) vs. input optical power for the case of $\beta_1 = 0$ (blue) and $\beta_3 = 0$ (red), illustrating the sublinear and superlinear behavior, respectively. Inset: energy diagram illustrating the two different cooling mechanisms.

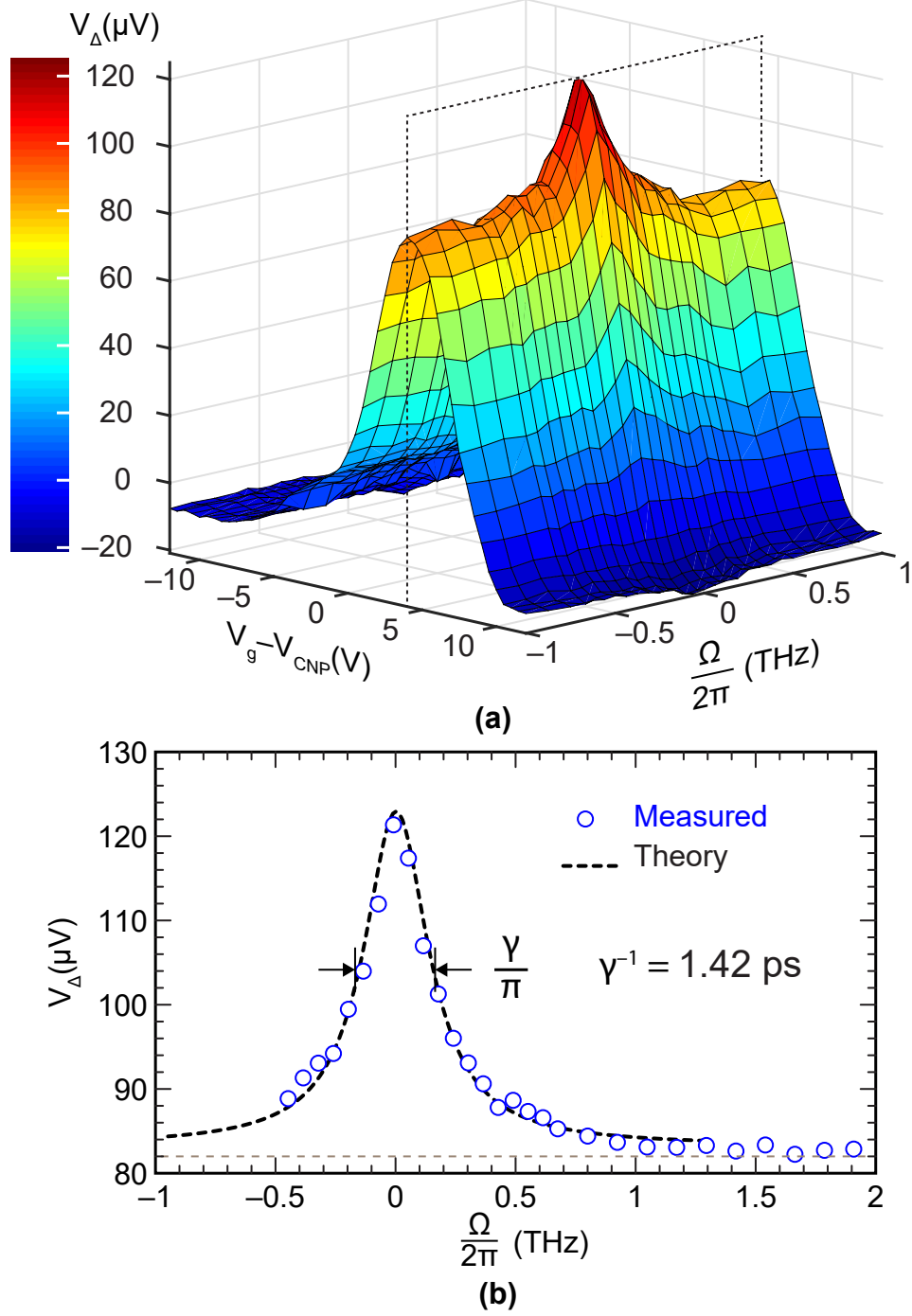


FIG. 3. Heterodyne photomixing response for the non-encapsulated device, measured at room temperature. (a) The nonlinear photomixing signal (V_{Δ}) as a function of their difference frequency (Ω) and gate voltage (V_g). At each gate voltage, V_{Δ} exhibits a Lorentzian-shaped dependence on the heterodyne difference frequency Ω . (b) V_{Δ} vs Ω for $V_g = 4$ V. The dashed curve is the theoretical calculation of the photomixing voltage based on a photothermoelectric effect. From the Lorentzian fit, the hot electron cooling time is estimated to be $\gamma^{-1} = 1.42$ ps.

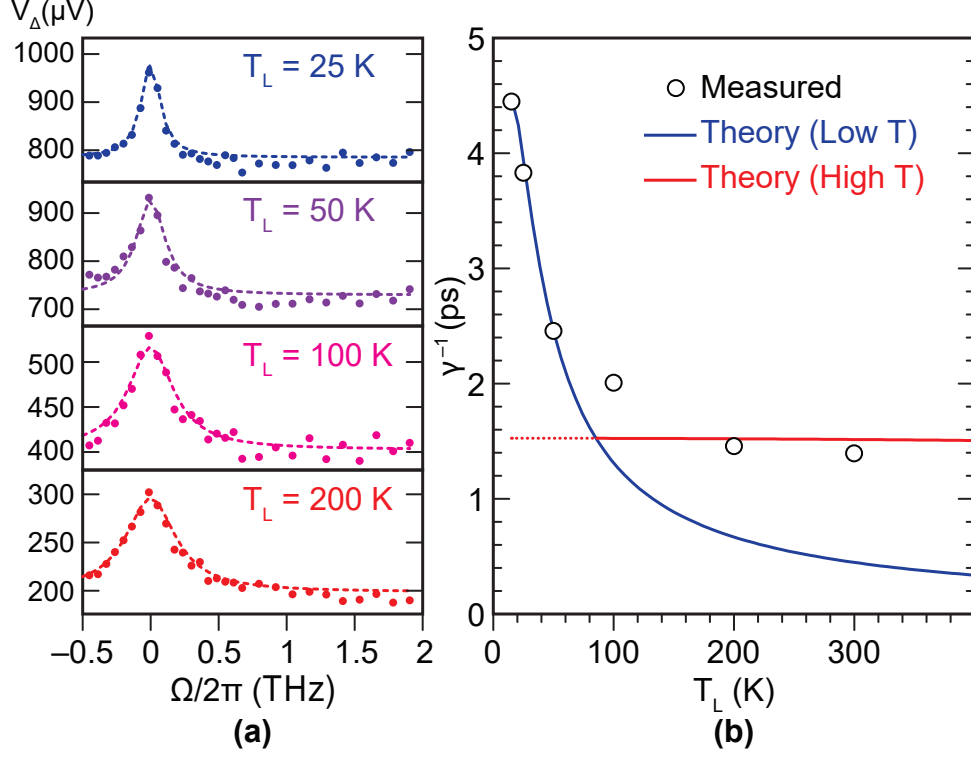


FIG. 4. Temperature dependence of cooling rate. (a) The two-beam mixing signal as a function of difference frequency measured close to the charge neutral point (sub-linear power dependence regime) at different temperatures for the exfoliated sample on SiO_2 . The dashed curves are Lorentzian fits. (b) Black circles are the extracted hot electron cooling time constant $\tau \equiv \gamma^{-1}$ from Lorentzian fits in part (a). The blue/red curve is the theory fit for low/high temperatures.