Electronic Supplementary Material (ESI) for Nanoscale Advances.

SUPPORTING INFORMATION

Ultrafast Hot-Carrier cooling in Quasi Freestanding Bilayer Graphene with Hydrogen Intercalated Atoms

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Cooling via disorder assisted acoustic-phonon scattering

The cooling rate of electron through disorder assisted acoustic-phonon scattering (supercollisions)[1,2,3] is given by

$$\frac{\partial T_e}{\partial t} = \frac{1}{C}\frac{\partial \varepsilon}{\partial t} \qquad (1)$$

Where $C = \alpha T$ is the electron heat capacity and α is the heat capacity coefficient. In the degenerate limit when $k_B T \ll E_F$, the electronic cooling rate assisted by supercollisions is given by

$$\frac{\partial T_e}{\partial t} = -\frac{A}{\alpha T}\left(T_e^3 - T_L^3\right) \qquad (2)$$

Which is reduced to

$$\frac{\partial T}{\partial t} = -\frac{A}{\alpha}\left(T_e^2\right) \quad \text{For } T_e \gg T_L \qquad (3)$$

The solution of the eq. 3 is:

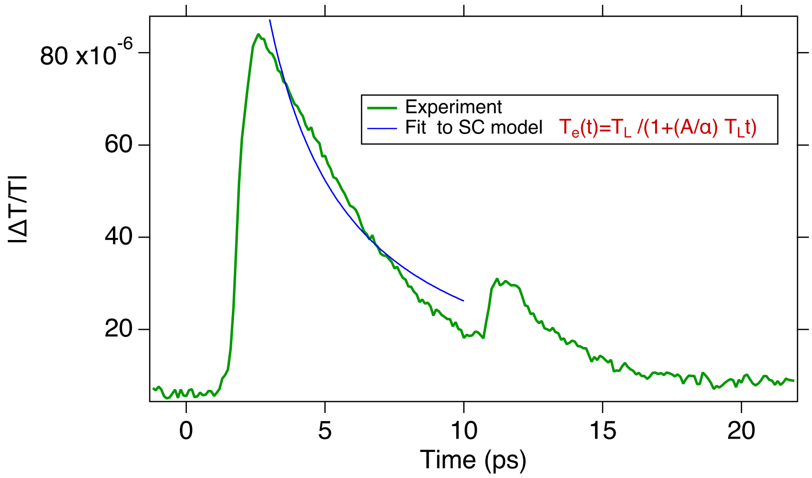
$$T_e(t) = \frac{T_L}{1 + \frac{A}{\alpha} T_L t} \quad \text{for } T_e \gg T_L \qquad (4)$$

That is the case based on the conditions of our experiment. The calculation of the cooling coefficient $\frac{A}{\alpha}$ is based on the equation

$$\frac{A}{\alpha} = \frac{2\,\lambda\,k_B}{3 k_F\, l\, \hbar} \qquad (5)$$

$where\ \lambda = \frac{2D^2 E_F}{\rho\, v_s^2 \pi (\hbar v_f)^2}$, $k_F = \frac{E_F}{\hbar v_f}$ with Fermi energy $E_F = 0.378\ eV$ of our sample, D is the deformation potential, and we chose the value of 15 eV adopted from literature [Pogna et. al., form the main text], ρ is the mass density of graphene $7.6 \times 10^{-7} kg/m^2$, $v_F = 1.1 \times 10^6\, m/s$, and $v_s = 2.1 \times 10^4 m/s$ is the Fermi velocity and the sound velocity, respectively.

The l is the mean free path given by $l = v_f \tau_{ms}$. The calculation of the momentum scattering time τ_{ms} is given by $\tau_{ms} = \frac{\mu E_F}{e v_F^2}$, where using Hall measurements we found the mobility, $\mu = 4290 cm^2 V^{-1} s^{-1}$. Based on the above equations and quantities we calculated the ratio $\frac{A}{a}$ and found it as of $\frac{A}{a} = 5.5 \times 10^7 K^{-1} s^{-1}$. This value is compatible with the values in literature [22 in the main text]. The fit of the experimental data with the SC model fails dramatically as it can be seen in Figure S1. Finally, the cooling time due to supercollisions τ_{sc} can be calculated from $\tau_{sc} = \left(3\frac{A}{a}T_L\right)^{-1}$ and we found $\tau_{sc} = 20\ ps$ which is more than 6 times larger than the cooling time we measured experimentally. Inverting the last equation along with the eq.5 and by inserting the average cooling time we measured which is 4.2 ps of all fluences we used we find an unrealistically large deformation potential exciting the value of 100 eV.



S1. The green line is the experimental measure of THz waveform transmitted through the excited sample, that reflects the dynamics of the electron temperature. The blue line is an attempt to fit with the supercollision model (eq. 6) in the main text.

Hall measurements: The two sets of experimental parameters used to determine the mobility of the carriers are listed below:

B = 2060 G, Current: 5E-5 A, Resistivity: 1.59 Ohm-cm, Mobility: 4358 cm2/Vs, Sheet Carrier density: 9.03E12 cm-2, F-factor: 0.98,

B = 2060 G, Current: 10E-5 A, Resistivity: 1.60 Ohm-cm, Mobility: 4225 cm2/Vs, Sheet Carrier density: 9.25E12 cm-2, F-factor: 0.981

The average value of the mobility has been used in the calculations in the main text.

References:

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[2] Graham, M. W., Shi, S.-F., Ralph, D. C., Park, J. & McEuen, P. L. Photocurrent measurements of supercollision cooling in graphene. *Nature Phys* **9**, 103–108 (2013).

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