Supporting Information

Photo-induced Terahertz Conductivity and Carrier Relaxation in Thermal-reduced Multilayer Graphene Oxide Films

Xiao Xing[1], Litao Zhao[2], Zeyu Zhang[1], Liang Fang[1], Zhengfu Fan[1], Xiumei Liu[1], Xian Lin[1], Jianhua Xu[2], Jinquan Chen[2], Xinluo Zhao[1], Zuanming Jin[1*] and Guohong Ma[1*]

[1]*Department of physics, Shanghai Universtiy, 99 Shangda Road, Shanghai 200444, P. R. China*

[2]*State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, 200062, China*

Photoinduced conductivity of RGO films annealed at four different temperatures with several pump fluences, F

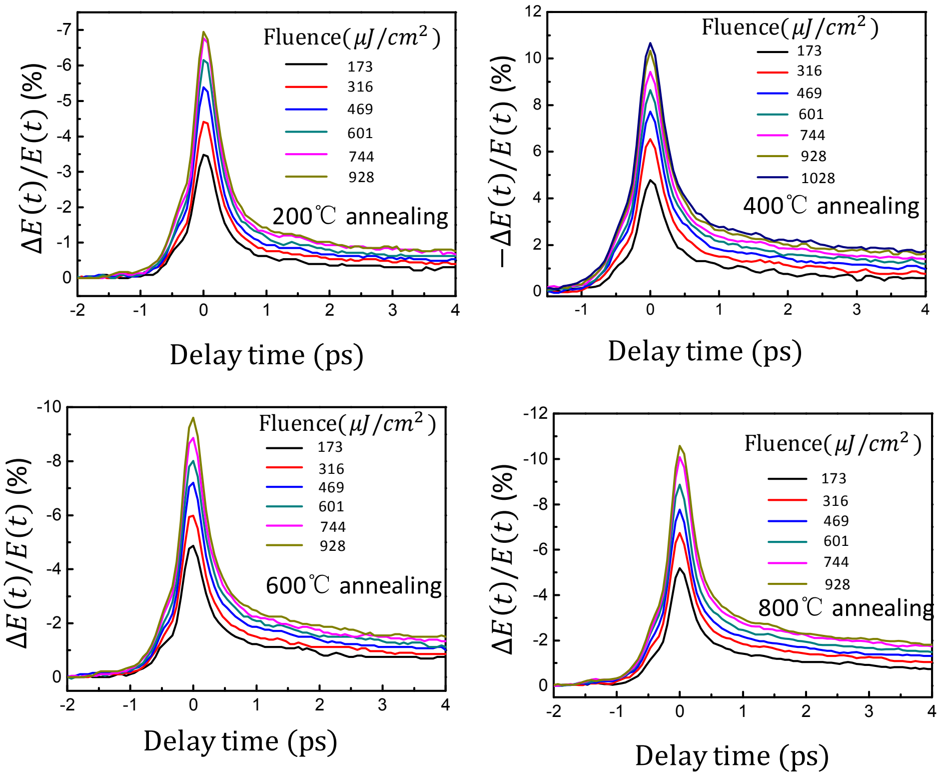

Figure S1 Photo-excited transient THz transmission response of multilayer RGO film annealed at different temperatures (200˚C 400 ˚C 600˚C 800˚C) with several fluence, F.

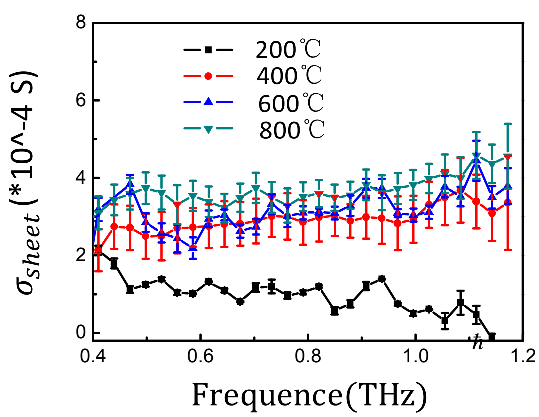

Figure S2 The real parts of the static conductivity of the multilayer RGO films at THz frequencies extracted from the amplitude and phase of relative THz-TDS transmission.

According to the intrinsic conductivity of the four RGO samples in Figure S2, we can rough estimate the $|E_F|$ by $\sigma_{DC} = \frac{\tau E_F e^2}{\pi \hbar^2}\sqrt{N}$, (N is the number of layers). Here we assume the momentum scattering time τ remains the same as that without pump excitation. Expect for the RGO sample annealed at 200 °C, all other three samples show a shorter momentum scattering time at higher annealing temperature (just as shown in table 1). This may be on account of that the RGO film annealed at 200 °C is affected more by the oxygen defects. As the intrinsic conductivity shifts from $0.9\times 10^{-4}S$ to $3.69 \times 10^{-4}S$ with the annealing temperature increases, also the scattering time τ decrease at higher annealing temperature, and $|E_F|$ turns larger as the annealing temperature increases. The magnitude of $|E_F|$ can be tuned by the annealing temperature, i.e. $|E_F|$ increases with increasing the annealing temperature.

On the other hand, photoconductivity, $\Delta\sigma$ is related to electron temperature T_e by [1]

$$\Delta\sigma = \sigma_{intra}(T_e) - \sigma_{intra}(T_0)\ ,\ \sigma_{intra}(T) = \frac{2G_0\tau k_B T}{\hbar(\omega^2\tau^2+1)}\ln[2\cosh(\frac{E_F}{2k_B T})], \quad (1)$$

Here, the lattice temperature T_0=300 K, τ is obtained from fitting frequency dependence of the dynamical photoconductivity. The electron temperature (T_e) therefore can be determined by the photoinduced conductivity ($\Delta\sigma$).

For the Supercollision (SC) mechanism the rate equation governing the cooling of the electron temperature is given by [2]

$$T_{el}(t) = \frac{T_{el,0}}{1+(A/\alpha)(t-t_0)T_{el,0}} \quad (2)$$

Here $T_{el,0}$ is the initial temperature, A/α is the SC rate coefficient. If the Fermi level $|E_F|$ is smaller than $2K_BT$ (at room temperature), $\Delta\sigma \sim T_{el}$. Therefore, the dynamics of photo-induced conductivity can be fitted by Eq (2), as shown in the Figures below. The best fit is obtained with the fitting parameter of A/α = 6.3~7.8 $\times 10^8$ $K^{-1}s^{-1}$, which is very close to the reported rate coefficient for the suspended graphene ($A/\alpha = 5.5\times 10^8$ $K^{-1}s^{-1}$) [3] and chemical reduced graphene oxide ($A/\alpha = 2.6 \times 10^8$ $K^{-1}s^{-1}$) [1]. We find A/α is independent on the annealing temperature. Predicted by the SC model, $\tau = [(A/\alpha)T_0]^{-1}$ (T_0 is the initial electron temperature), the cooling time (τ) is independent on the annealing temperature in the case of similar electron temperature (T_0), as shown in Fig.S3.

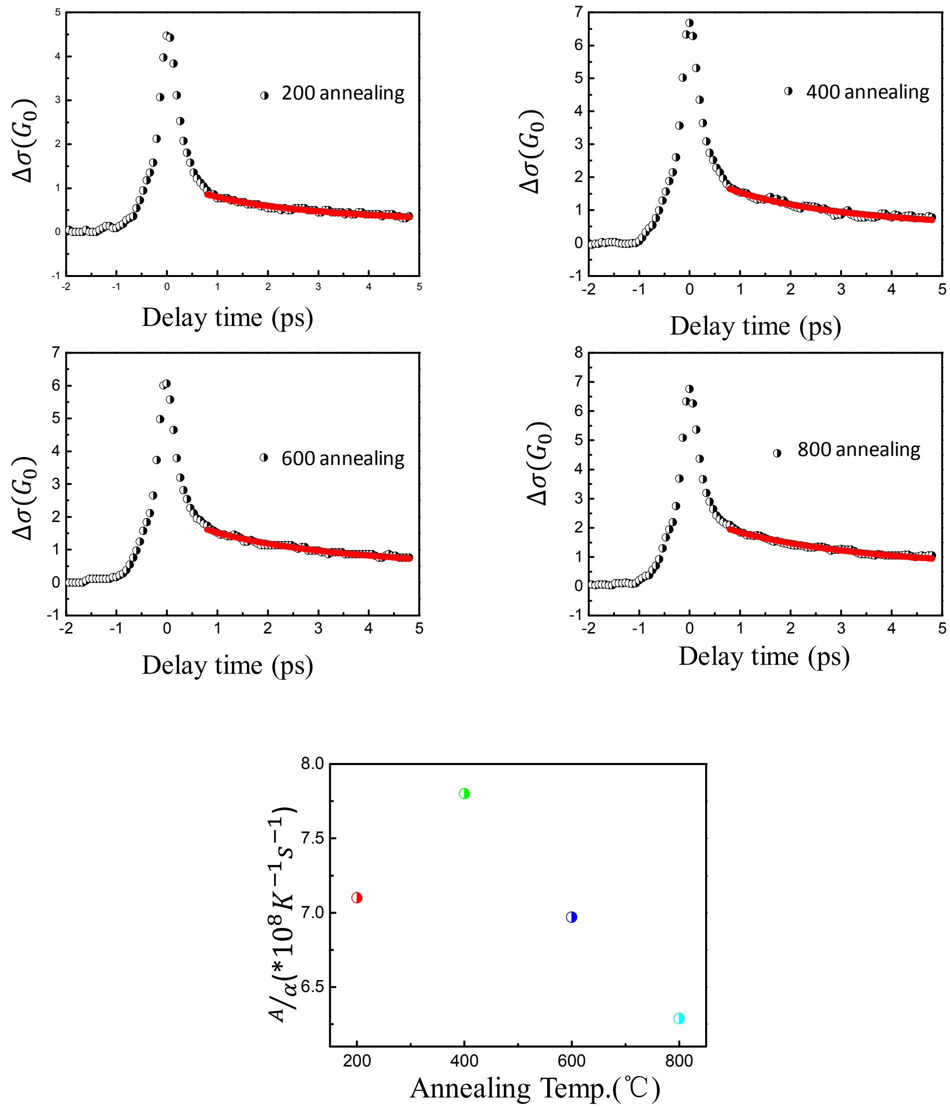

Figure S3 The SC rate coefficient with the RGO samples at four annealing temperatures

References:

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Photocurrent Measurements of Supercollision Cooling in Graphene. *Nat. Phys.* **2013**, *9*, 103-108.