



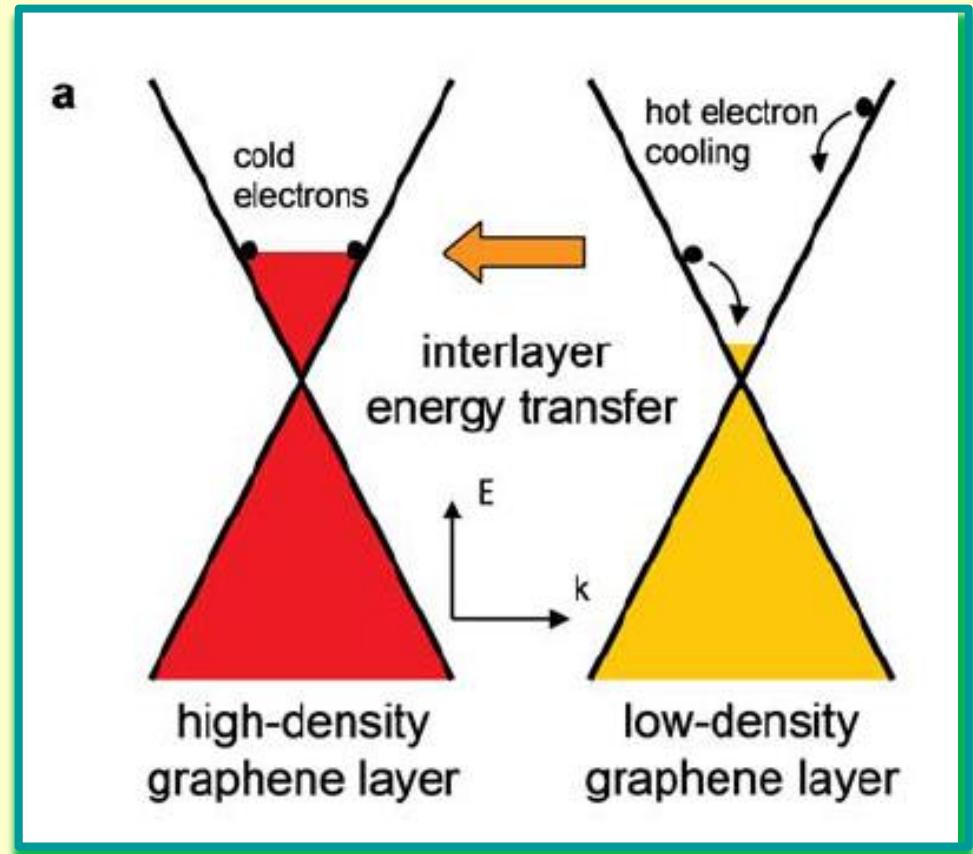
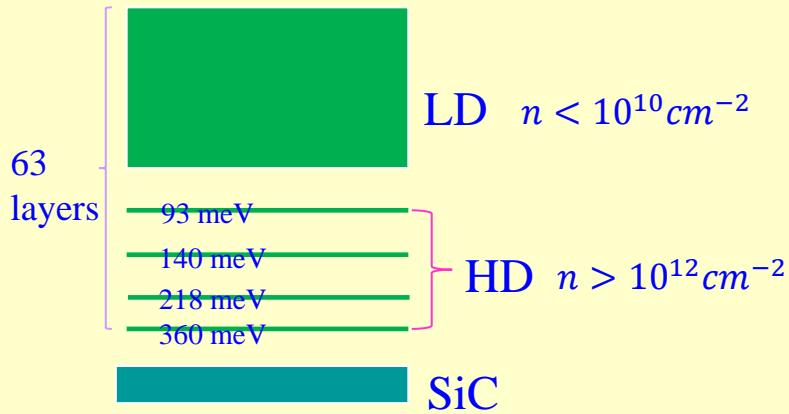
# Electronic Cooling in Multilayer Epitaxial Graphene

Reza Asgari

[asgari@ipm.ir](mailto:asgari@ipm.ir)

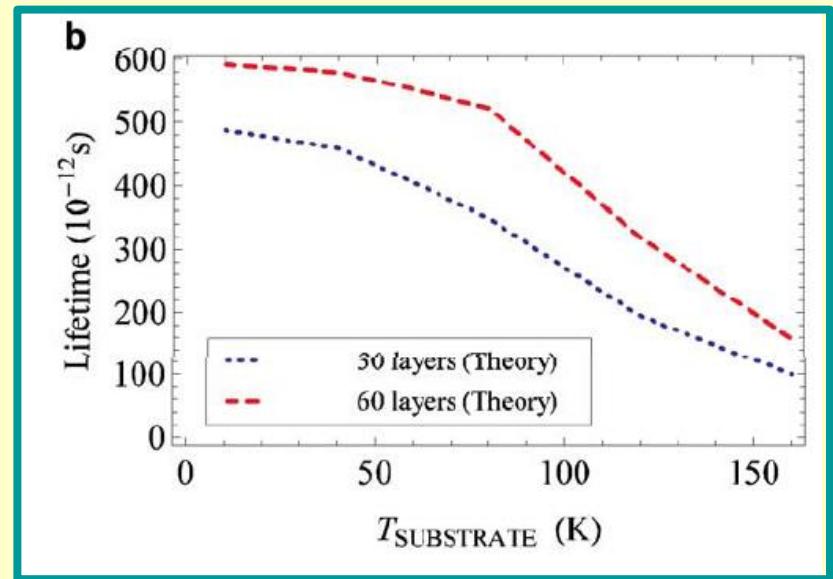
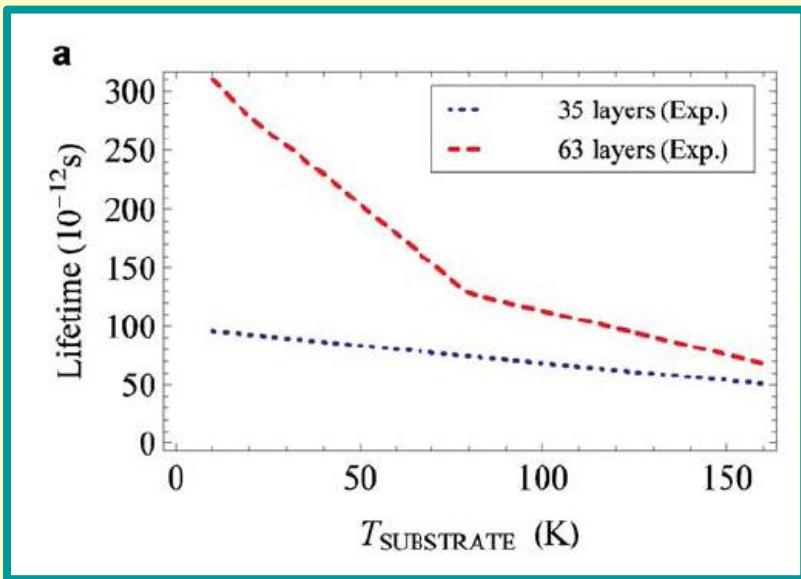


# *My purpose I:*



# *My purpose II:*

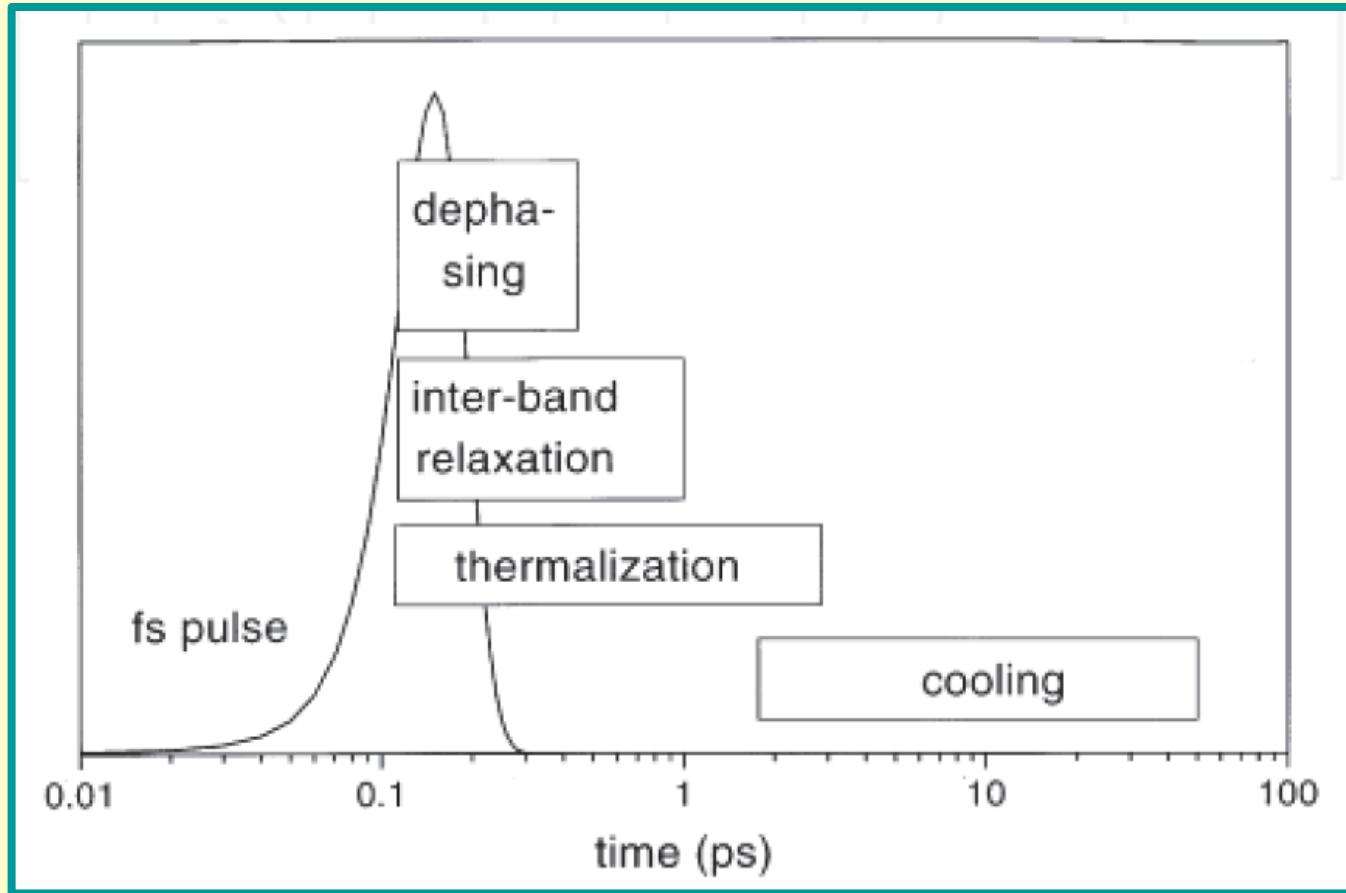
Electronic cooling life time



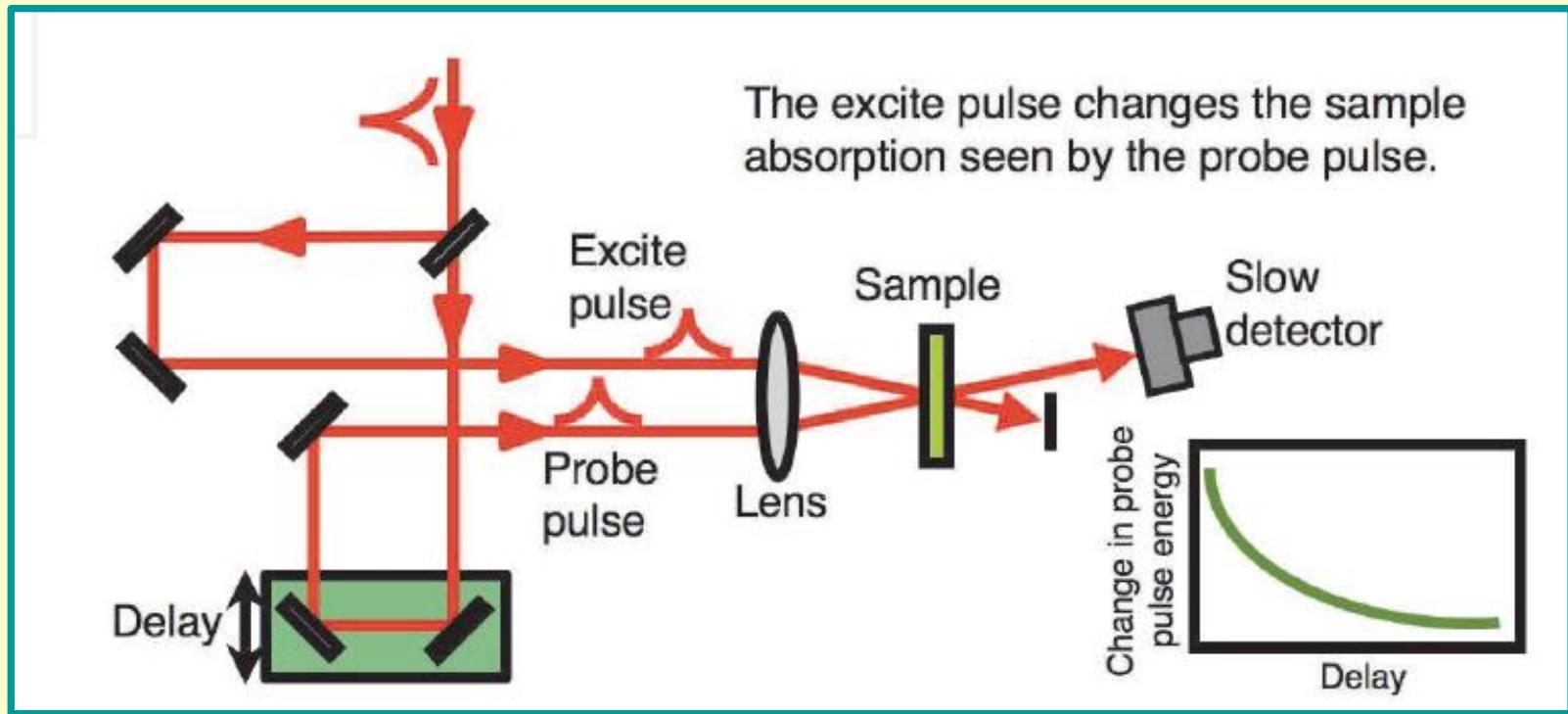
# *Outlook*

1. Introduction  
brief overview on electronic cooling and ultrafast spectroscopy
2. Electronic cooling and different mechanisms  
electron-phonon interactions  
electron-impurity interactions  
electron-electron interactions  
Multilayer epitaxial graphene
3. Conclusion

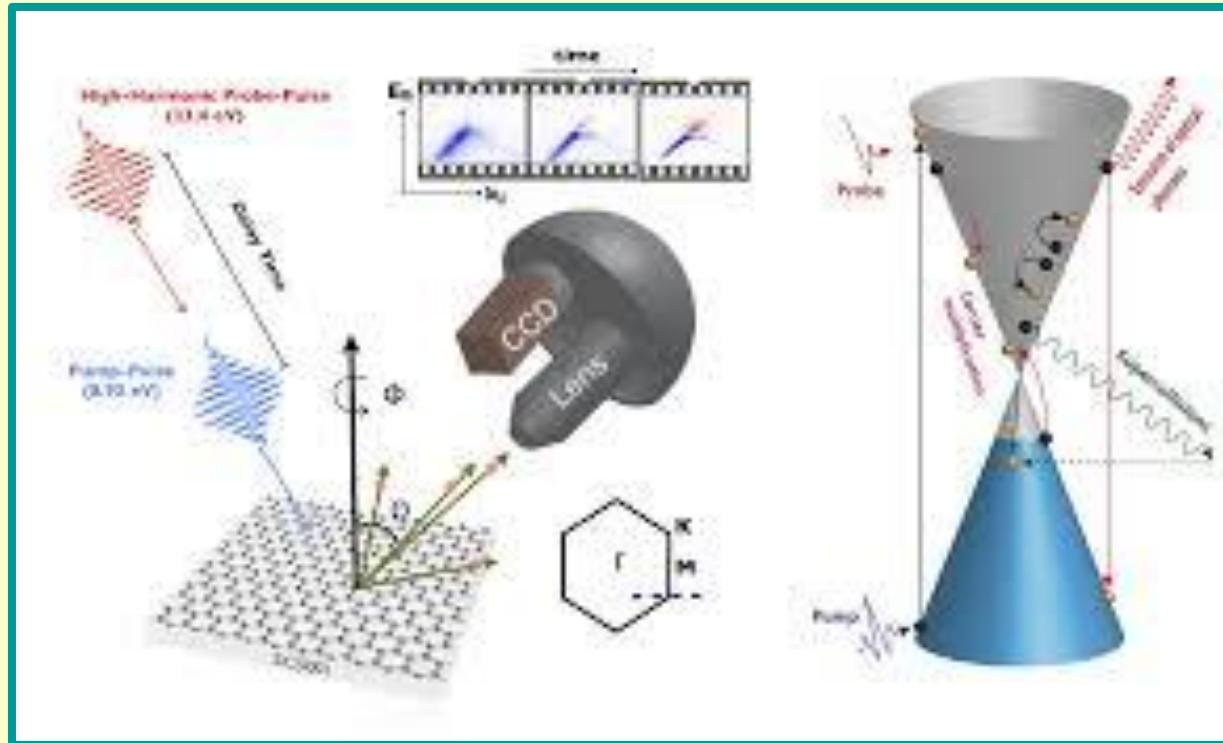
# *Ultrafast Spectroscopy*



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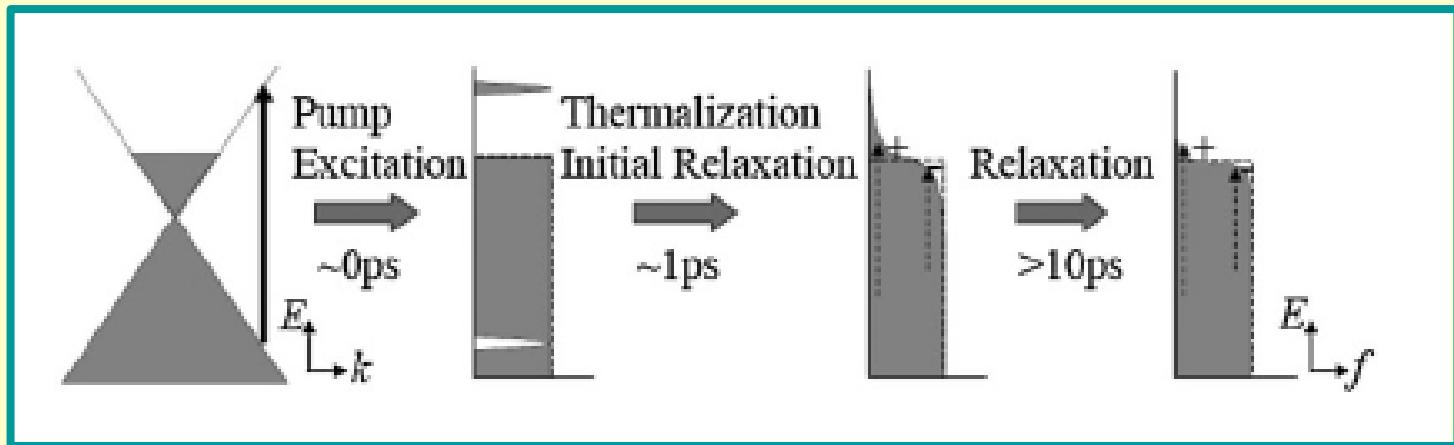


# *Ultrafast Spectroscopy*



# *Ultrafast Spectroscopy*

## Thermalization of the hot electron



# *Outlook*

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# *Electron-phonon interactions*

Heat transfer rate

$$Q_{e-ac} = V_d \sum (T_e^\delta - T_{ac}^\delta)$$

- $\delta = 5$  3D metals, F. Wellstood, et al, PRB 49, 5942 (1994)
- $\delta = 6$  Discorded thin film, Sergeev and Mitin , PRB 61, 6041 (2000)
- $\delta = 3$  Clamped 3D but vibrations are 1D, Hekkila, PRB 77, 033401 (2008)

$$\delta = 2+d$$

# *Electron-phonon interactions: graphene*

$$Q_{e-ac} = V_d \Sigma (T_e^\delta - T_{ac}^\delta)$$

$\delta = 4$     2D graphene, Kubakadda , PRB 79, 075417 (2009)

$$Q_{e-ac} = V_d g(\mu, T_e) (T_e - T_{ac})$$

2D graphene, Bistritzer and MacDonald PRL 102, 206410 (2009)

# *Electron-phonon interactions: Graphene*

Classical Boltzmann equation

$$\partial_t f_k^\alpha = S_{e-ph}(f_k^\alpha)$$

Scattering integral

$$S_{e-ph}(f_k^\alpha) = - \sum_{p\beta} [f_k^\alpha(1-f_p^\beta)W_{k\alpha \rightarrow p\beta} - f_p^\beta(1-f_k^\alpha)W_{p\beta \rightarrow k\alpha}],$$

Golden-rule scattering rate

$$W_{k\alpha \rightarrow p\beta} = \frac{2\pi}{\hbar} \sum_{q\gamma} w_{kp,q}^{\alpha\beta,\gamma} [(n_q^\gamma + 1)\delta_{k,p+q}\delta(\epsilon_{kp}^{\alpha\beta} - \omega_q^\gamma) + n_q^\gamma\delta_{k,p-q}\delta(\epsilon_{kp}^{\alpha\beta} + \omega_q^\gamma)].$$

$$\epsilon_{kp}^{\alpha\beta} = \epsilon_k^\alpha - \epsilon_p^\beta$$

$$Q = -\partial_t \sum_{k\alpha} \epsilon_k^\alpha f_k^\alpha = - \sum_{k\alpha} \epsilon_k^\alpha S_{e-ph}(f_k^\alpha).$$

# Electron-phonon interactions: Graphene

Electron-phonon Hamiltonian:

$$\hat{H}_{e-ac} = \sum_{k\alpha, p\beta} \sum_{\mathbf{q}} M_{pk,q}^{\beta\alpha} c_{p\beta}^\dagger c_{k\alpha} (b_{\mathbf{q}} + b_{-\mathbf{q}}^\dagger)$$

D is the coupling constant

$$M_{pk,q}^{\beta\alpha} = -\sqrt{\hbar^2/(2M\omega_q)} D q \langle p\beta | e^{i\mathbf{q}\cdot\mathbf{r}} | k\alpha \rangle$$

$$\omega_q = \hbar c q$$

Effective interaction term

$$w_{kp,q}^{\alpha\beta} = \frac{\hbar^2}{2M\omega_q} D^2 q^2 F_{\alpha\beta}(\theta)$$

$$F_{\alpha\beta}(\theta) = (1 + s_\alpha s_\beta \cos n\theta)/2$$

$$\theta = \phi_p - \phi_k$$

# Electron-phonon interactions: Graphene

$$\begin{aligned}
 Q_{\text{ind}} = & -\frac{g_e A \hbar D^2}{2(2\pi)^2 \rho} \sum_{\alpha\beta} \int_0^\infty dk k \int_{q_{\min}^{\alpha\beta}}^{q_{\max}^{\alpha\beta}} dq q^2 \\
 & \times \frac{1 + s_\alpha s_\beta y(k, q)}{\sqrt{1 - [x(k, q)]^2}} |\hbar v_k^\alpha [p(k, q)/k]^{n-2}|^{-1} \\
 & \times n_{ac}(\hbar c q) [f(\epsilon_k^\alpha - \hbar c q) - f(\epsilon_k^\alpha)]
 \end{aligned}$$

$$p(k, q) = [s_\alpha s_\beta - s_\beta \hbar c q \gamma_1^{n-1} / (\hbar v k)^n]^{1/n} k$$

$$x(k, q) = -\{[p(k, q)]^2 - k^2 - q^2\} / (2kq)$$

$$y(k, q) = n[z(k, q)]^n - n + 1$$

$$z(k, q) = [k - qx(k, q)] / p(k, q)$$

# *Electron-phonon interactions: Graphene*

Limit of the low temperature

$$k_B T_e, k_B T_{ac} \ll 2(c/v)|\mu| = k_B T_{BG,MLG}$$

$$\mathcal{Q}_{e-ac} = V_d \Sigma (T_e^\delta - T_{ac}^\delta)$$

$$\Sigma = \frac{\pi^2 D^2 |\mu| k_B^4}{15 \rho \hbar^5 v^3 c^3}$$

$$\delta=4$$

# *Electron-phonon interactions: Graphene*

For high temperature regiem

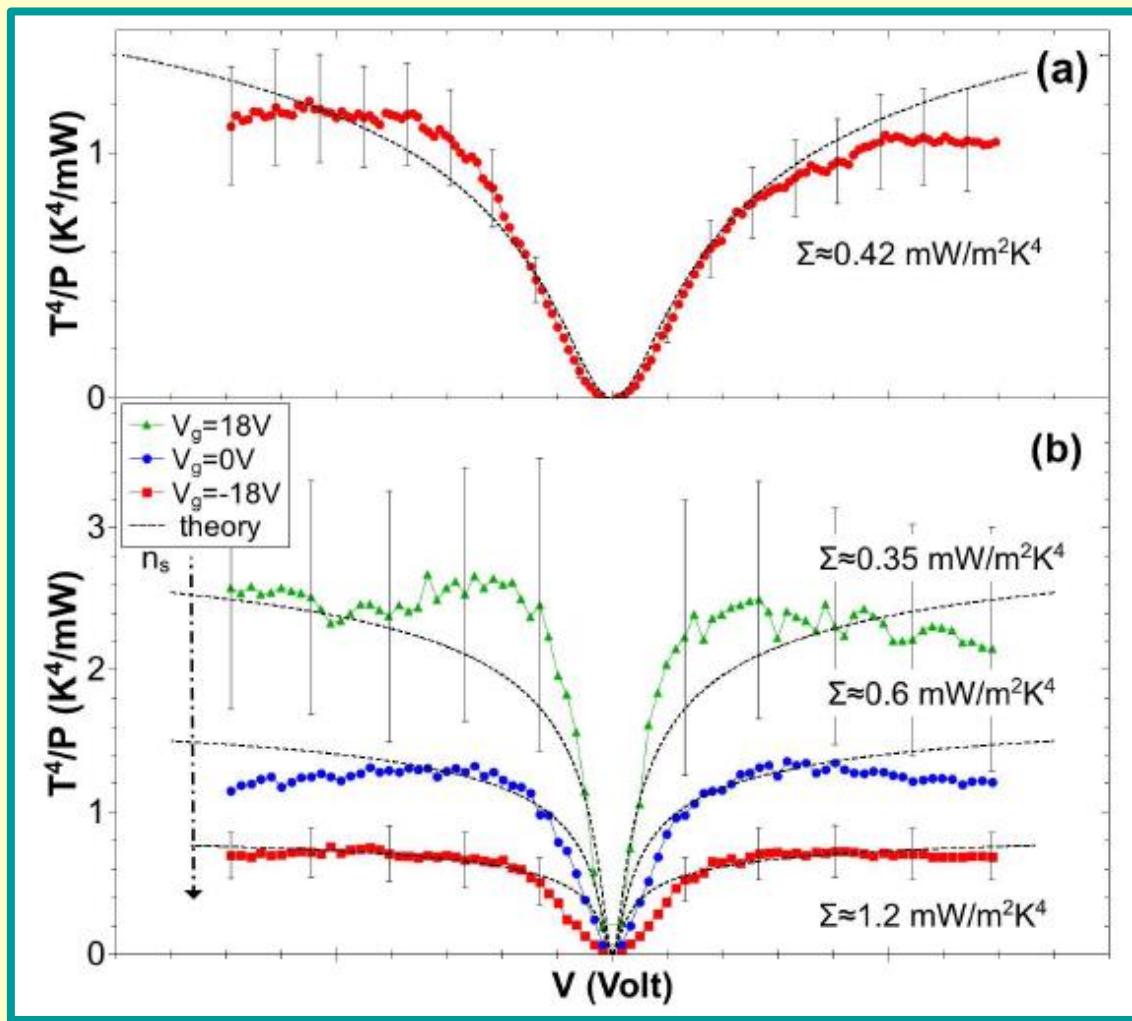
$$2(c/v)\max(|\mu|, k_B T_e) \ll k_B T_e, k_B T_{ac}$$

$$Q_{e-ac} = V_d g(\mu, T_e) (T_e - T_{ac})$$

$$g(\mu, T_e) = \frac{D^2 k_B}{30 \pi o \hbar^5 v^6} [15\mu^4 + 30\pi^2 \mu^2 (k_B T_e)^2 + 7\pi^4 (k_B T_e)^4]$$

$$g(\mu, T_e) \propto T_e^4$$

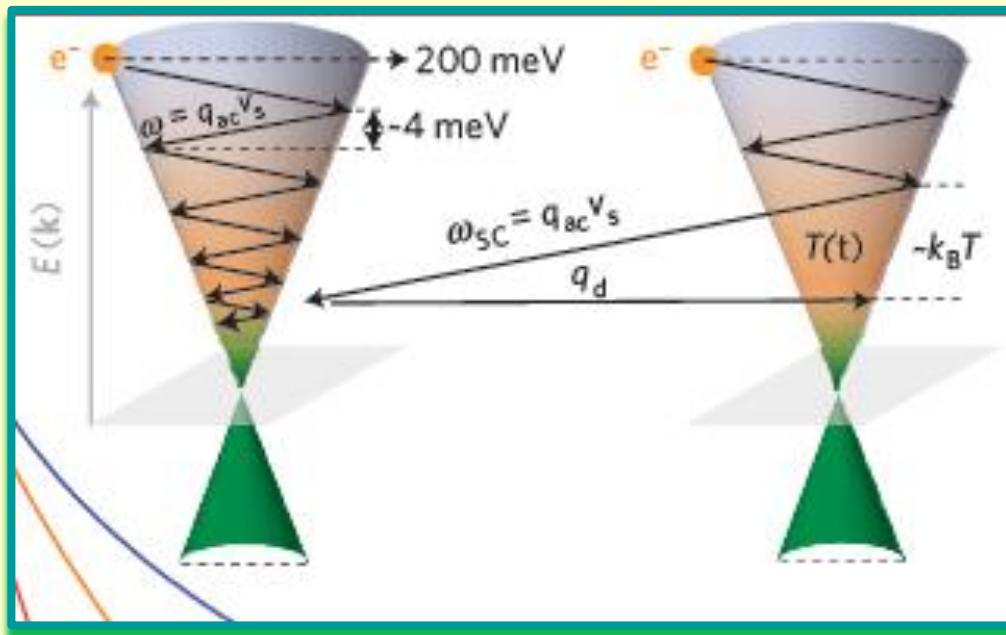
# *Electron-phonon interactions: Exp results*



# *Outlook*

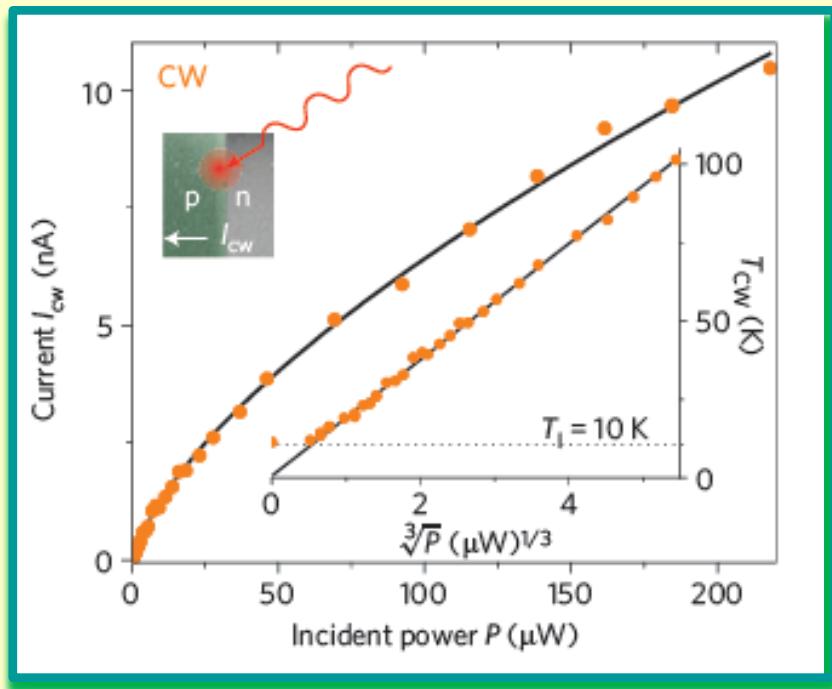
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# Electron-impurity interactions: supercollision

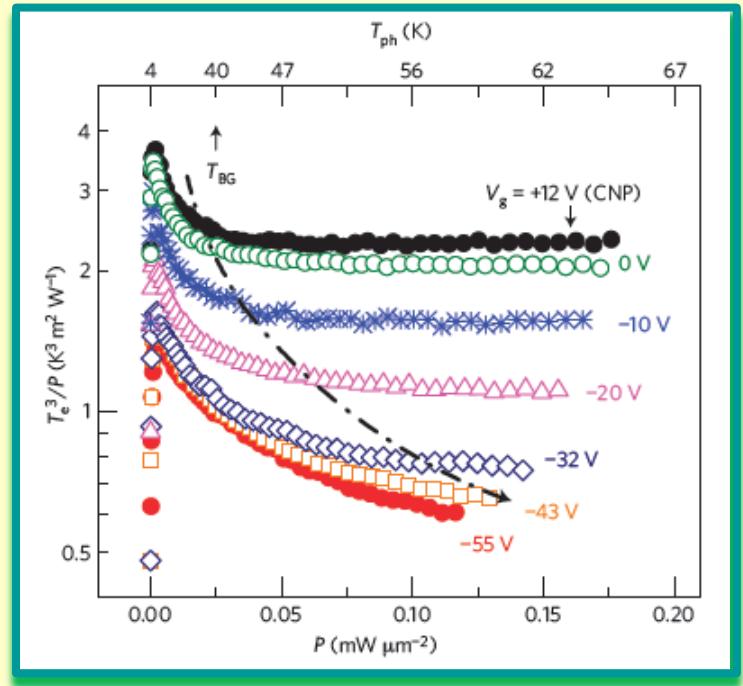


$$J_{\text{SC}} = A (T_e^3 - T_{\text{ph}}^3), \quad A = 9.62 \frac{g^2 v^2(E_F) k_B^3}{\hbar k_F l}$$

# Electron-impurity interactions: Evidences!

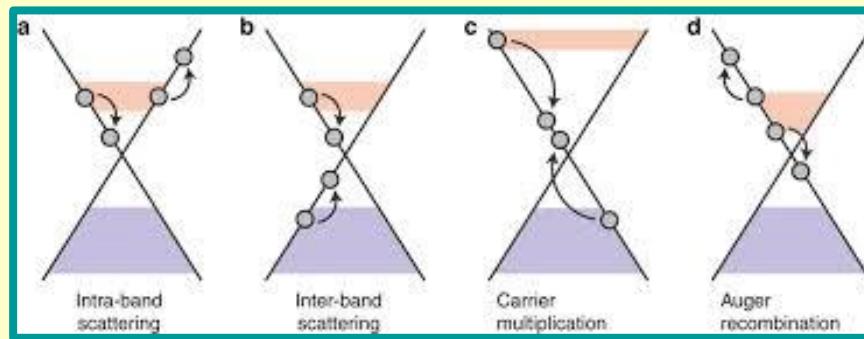
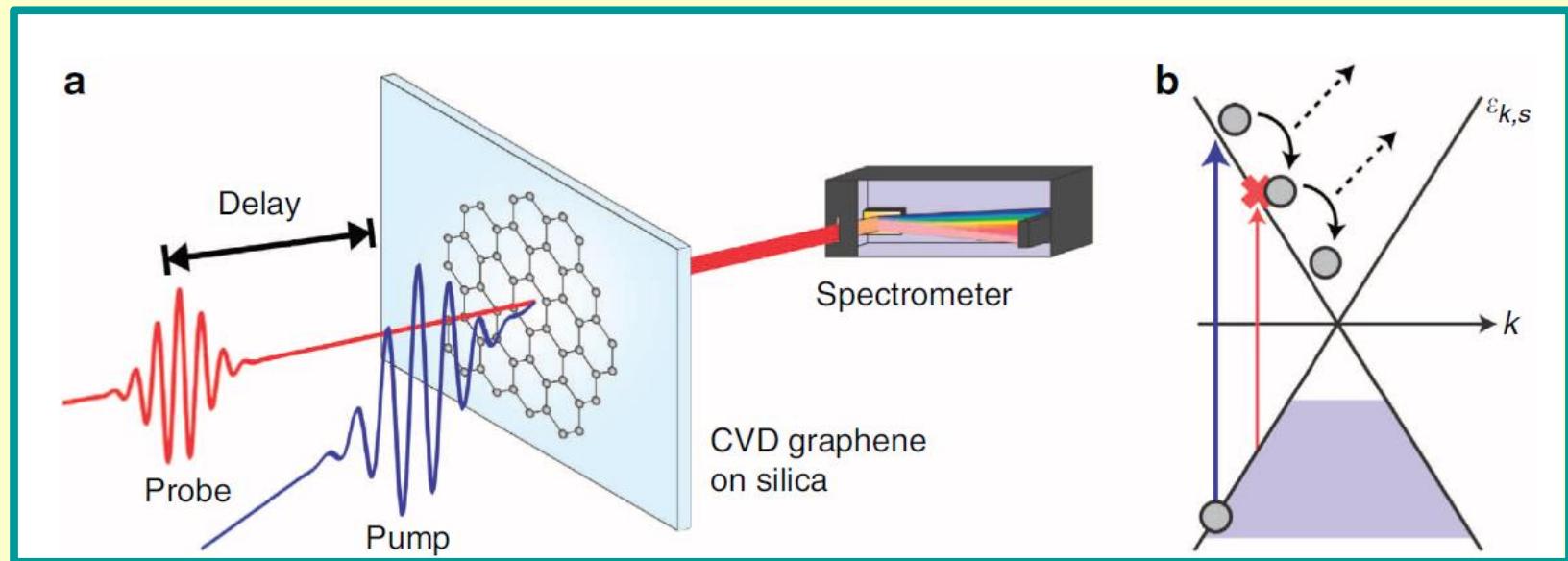


Graham et al Nature Physics (2012)



Betz et al Nature Physics (2012)

# Electron-electron interactions



Brida, et al, Nature Commun, DOI 10.1038 (2013)

Butscher, et al App. Phys. Lett **91**, 203103 (2007)

Sun et al, Phys. Rev. B **85**, 125413 (2012)

# Electron-electron interactions

Semiclassical Boltzmann equation

$$\frac{df_\mu(\varepsilon_{\mathbf{k},\lambda})}{dt} = \left. \frac{df_\mu(\varepsilon_{\mathbf{k},\lambda})}{dt} \right|_{e-e} + \left. \frac{df_\mu(\varepsilon_{\mathbf{k},\lambda})}{dt} \right|_{e-ph}$$

$$\begin{aligned} \left. \frac{df_\mu(\varepsilon_1)}{dt} \right|_{e-e} &= \int_{-\infty}^{\infty} d\varepsilon_2 \int_{-\infty}^{\infty} d\varepsilon_3 C_\mu(\varepsilon_1, \varepsilon_2, \varepsilon_3) \\ &\times \{ [1 - f_\mu(\varepsilon_1)][1 - f_\mu(\varepsilon_2)]f_\mu(\varepsilon_3)f_\mu(\varepsilon_4) \\ &- f_\mu(\varepsilon_1)f_\mu(\varepsilon_2)[1 - f_\mu(\varepsilon_3)][1 - f_\mu(\varepsilon_4)] \} \end{aligned}$$

Collision integral

$$\begin{aligned} C_\mu(\varepsilon_1, \varepsilon_2, \varepsilon_3) &= \frac{2\pi}{\hbar} \frac{1}{S^2} \sum_{\mathbf{Q}, \mathbf{k}_3} |V_{1234}^{(\mu)}|^2 \times \delta(|E - \varepsilon_1| - \hbar\nu |\mathbf{Q} - \mathbf{k}_1|) \\ &\times \delta(|\varepsilon_3| - \hbar\nu k_3) \times \delta(|E - \varepsilon_3| - \hbar\nu |\mathbf{Q} - \mathbf{k}_3| + \eta) \end{aligned}$$

Brida, et al, Nature Commuin, DOI 10.1038 (2013)

Butscher, et al App. Phys. Lett **91**, 203103 (2007)

Sun et al, Phys. rEv. B **85**, 125413 (2012)

$$E = \varepsilon_1 + \varepsilon_2$$

$$Q = k_1 + k_2$$

# Electron-electron interactions

$$\frac{df_\mu(\varepsilon_{\mathbf{k},\lambda})}{dt} = \left. \frac{df_\mu(\varepsilon_{\mathbf{k},\lambda})}{dt} \right|_{e-e} + \left. \frac{df_\mu(\varepsilon_{\mathbf{k},\lambda})}{dt} \right|_{e-ph}$$

$$| V_{1234}^{(\mu)} |^2 = | \mathcal{U}_{1234}^{(\mu)} - \mathcal{U}_{1243}^{(\mu)} |^2 / 2 + | \mathcal{U}_{1234}^{(\mu)} |^2$$

$$\mathcal{U}_{1234}^{(\mu)} = W(| \mathbf{k}_1 - \mathbf{k}_3 |, \omega; t) F_{s_1, s_3}^{(\mu)}(\theta_{\mathbf{k}_3} - \theta_{\mathbf{k}_1}) F_{s_2, s_4}^{(\mu)}(\theta_{\mathbf{k}_4} - \theta_{\mathbf{k}_2})$$

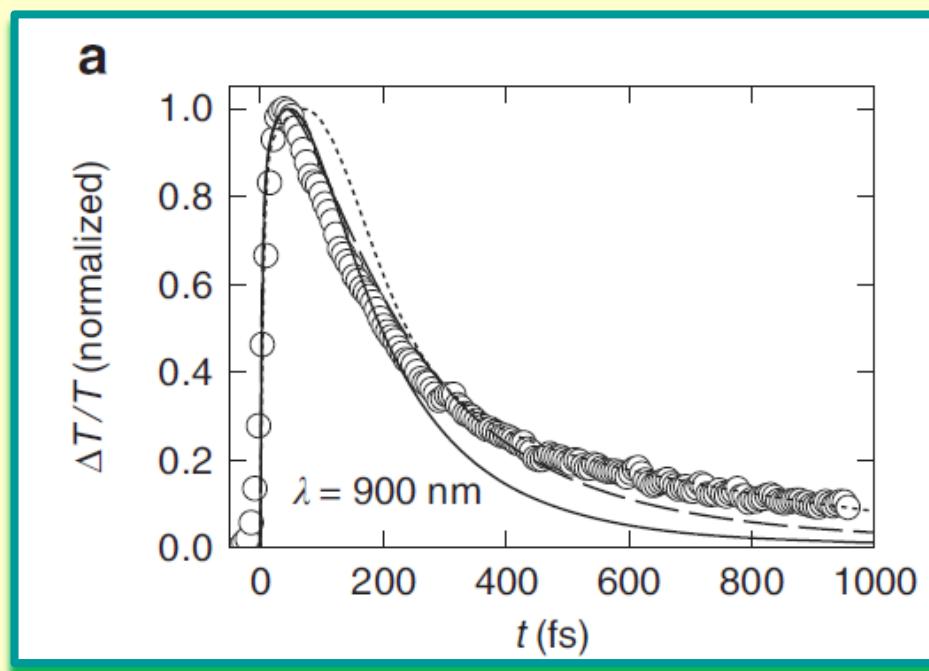
$$F_{s,s'}^{(\mu)}(\theta) = [1 + ss' \exp(i\mu\theta)]/2$$

$$C_\mu(\varepsilon_1, \varepsilon_2, \varepsilon_3)|_{\text{Auger}} = \frac{1}{8\pi^2 \hbar^5 \nu^4} \sqrt{\left| \frac{\varepsilon_2 \varepsilon_3 \varepsilon_4}{\varepsilon_1} \right|} | V_{1234}^{(\mu)} |^2$$

$$\text{CM}(t) \equiv \frac{n_c(t) - n_c(-\infty)}{n_c(0) - n_c(-\infty)}$$

# *Electron-electron interactions: Delay time*

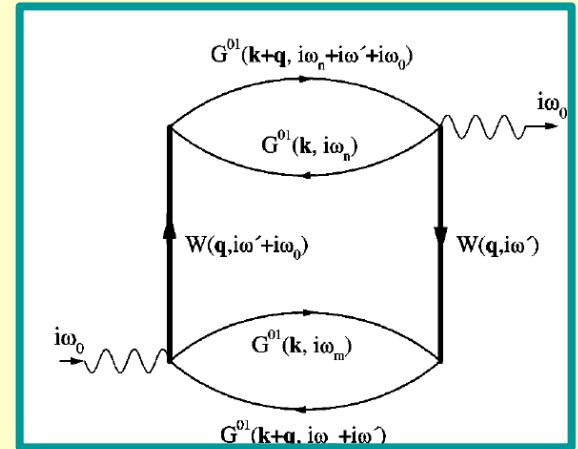
$$\frac{\Delta T}{T}(\lambda, t) = \pi\alpha[f_\mu(\hbar\omega/2) - n_F(\hbar\omega/2) - f_\mu(-\hbar\omega/2) + n_F(-\hbar\omega/2)].$$



# Electron-electron interactions: interlayer

$$\dot{\mathbf{P}}_2 = -i[\mathbf{P}_2, H] = -i\nabla_{\mathbf{R}_2}H = \sum_{\mathbf{q}} i\mathbf{q}U_d(q)\rho_{\mathbf{q}}^1\rho_{-\mathbf{q}}^2 e^{-i\mathbf{q}\cdot(\mathbf{R}_1-\mathbf{R}_2)} \\ - en_2\mathbf{E}_2 + \dot{\mathbf{P}}_{2,ep} + \dot{\mathbf{P}}_{2,ei}.$$

$$en_2\mathbf{E}_2 = i\sum_{\mathbf{q}} \mathbf{q} \int_{-\infty}^{\infty} dt_1 (-i) \theta(t-t_1) \sum_{\mathbf{k}, \mathbf{p}} \sum_{\mathbf{Q}} \\ \times e^{-i\mathbf{q}\cdot[\mathbf{R}_1(t)-\mathbf{R}_1(t_1)]+i\mathbf{Q}\cdot\mathbf{R}_1(t_1)} e^{i(1-\gamma)(\varepsilon_{k+q}-\varepsilon_k)(t-t_1)} \\ \times \text{Tr}\{ \tilde{\varrho}[ U_d(q)\tilde{c}_{\mathbf{k}+\mathbf{q}}^\dagger(t)\tilde{c}_{\mathbf{k}}(t)\tilde{d}_{\mathbf{p}-\mathbf{q}}^\dagger(t)\tilde{d}_{\mathbf{p}}(t), \\ \times U_d(\mathbf{Q}-\mathbf{q})\tilde{\rho}_{\mathbf{Q}-\mathbf{q}}^1(t_1)\tilde{\rho}_{\mathbf{q}-\mathbf{Q}}^2(t_1)] \}.$$



# *Electron-electron interactions: Interlayer*

Cooling power rate transferred between layers

$$\frac{\underline{Q}^{el}}{L^2} = \frac{\hbar}{4\pi^3} \int_{-\infty}^{\infty} \omega d\omega \int d\vec{q} |v_{LD,HD}^{sc}|^2 [n_B(T_{LD}) - n_B(T_{HD})]$$
$$\times \text{Im}\chi_{LD}(q, \omega, T_{LD}) \text{Im}\chi_{HD}(q, \omega, T_{HD})$$

# Electron-electron interactions

$$\chi^{(0)}(q, \omega, T) = g \lim_{\eta \rightarrow 0^+} \sum_{s, s' = \pm} \int \frac{d^2 k}{(2\pi)^2} \frac{1 + ss' \cos(\theta_{k, k+q})}{2} \frac{n_F(\varepsilon_{k,s}) - n_F(\varepsilon_{k+q,s'})}{\omega + \varepsilon_{k,s} - \varepsilon_{k+q,s'} + i\eta}.$$

$$\begin{aligned} \text{Im } \chi^{(0)}(q, \omega, T) = & \frac{g}{4\pi} \sum_{\alpha=\pm} \left\{ \Theta(v_F q - \omega) q^2 f(v_F q, \omega) [G_+^{(\alpha)}(q, \omega, T) - G_-^{(\alpha)}(q, \omega, T)] \right. \\ & \left. + \Theta(\omega - v_F q) q^2 f(\omega, v_F q) \left[ -\frac{\pi}{2} \delta_{\alpha, -} + H_+^{(\alpha)}(q, \omega, T) \right] \right\} \end{aligned}$$

$$\begin{aligned} \text{Re } \chi^{(0)}(q, \omega, T) = & \frac{g}{4\pi} \sum_{\alpha=\pm} \left\{ \frac{-2k_B T \ln[1 + e^{\alpha \mu_0 / (k_B T)}]}{v_F^2} + \Theta(\omega - v_F q) \right. \\ & \times q^2 f(\omega, v_F q) [G_-^{(\alpha)}(q, \omega, T) - G_+^{(\alpha)}(q, \omega, T)] \\ & \left. + \Theta(v_F q - \omega) q^2 f(v_F q, \omega) \left[ -\frac{\pi}{2} \delta_{\alpha, -} + H_-^{(\alpha)}(q, \omega, T) \right] \right\}. \end{aligned}$$

$$f(x, y) = \frac{1}{2\sqrt{x^2 - y^2}}$$

$$G_{\pm}^{(\alpha)}(q, \omega, T) = \int_1^{\infty} du \frac{\sqrt{u^2 - 1}}{\exp\left(\frac{|v_F q u \pm \omega| - 2\alpha \mu_0}{2k_B T}\right) + 1}$$

# *Electron specific heat and spin susceptibility at low temperature*

$$C_V = -T \partial^2[n\delta f(T)]/\partial T^2$$

$$\delta f_{\text{int}}(T \rightarrow 0) = \varepsilon_F \frac{\pi^2}{3} \left(\frac{T}{T_F}\right)^2 \frac{\alpha_{\text{gr}}[1 - \alpha_{\text{gr}}\xi(\alpha_{\text{gr}})]}{4g} \ln \Lambda + \text{R.T.}$$

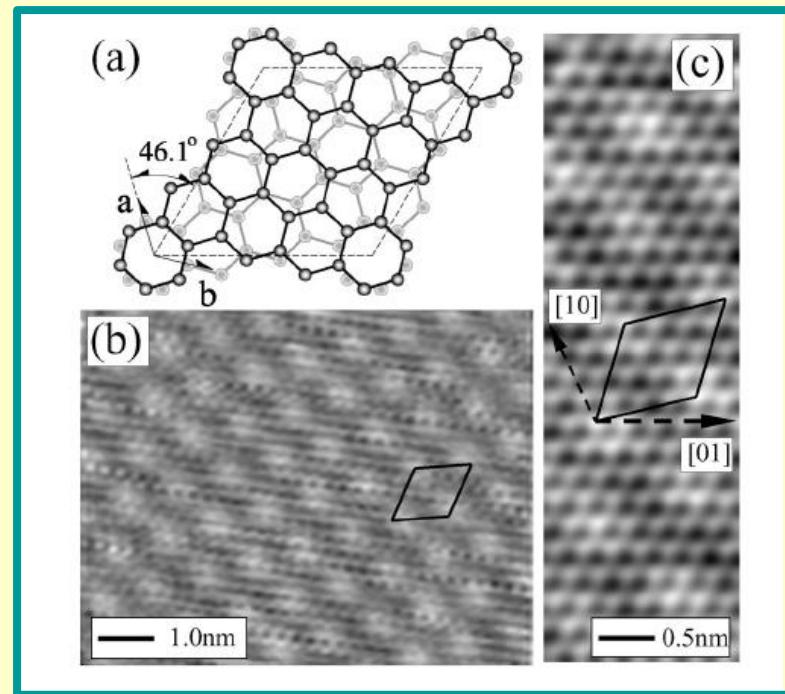
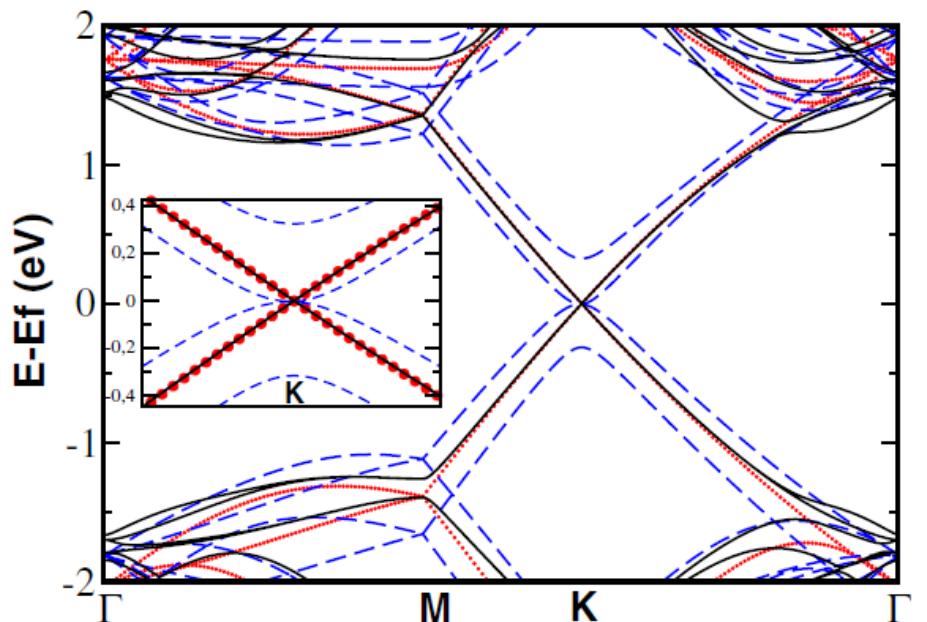
$$\frac{1}{\chi_s(T)} = \frac{1}{n\mu_B^2} \frac{\partial^2[f_0(T, \xi) + f_{\text{int}}(T, \xi)]}{\partial \xi^2} \Big|_{\xi=0}$$

$$\begin{aligned} \delta\chi_s^{-1}(T) &= \chi_s^{-1}(T) - \chi_s^{-1}(T = 0) \\ &= \frac{\varepsilon_F \pi^2}{8n\mu_B^2} \left(\frac{T}{T_F}\right)^2 \left[ \frac{g_v}{3} - \eta \ln \Lambda \right] \end{aligned}$$

# *Outlook*

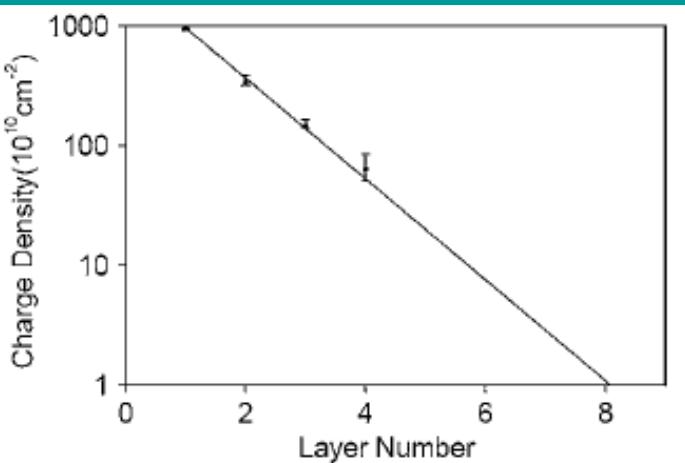
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# *Epitaxial graphene on C-terminated SiC*

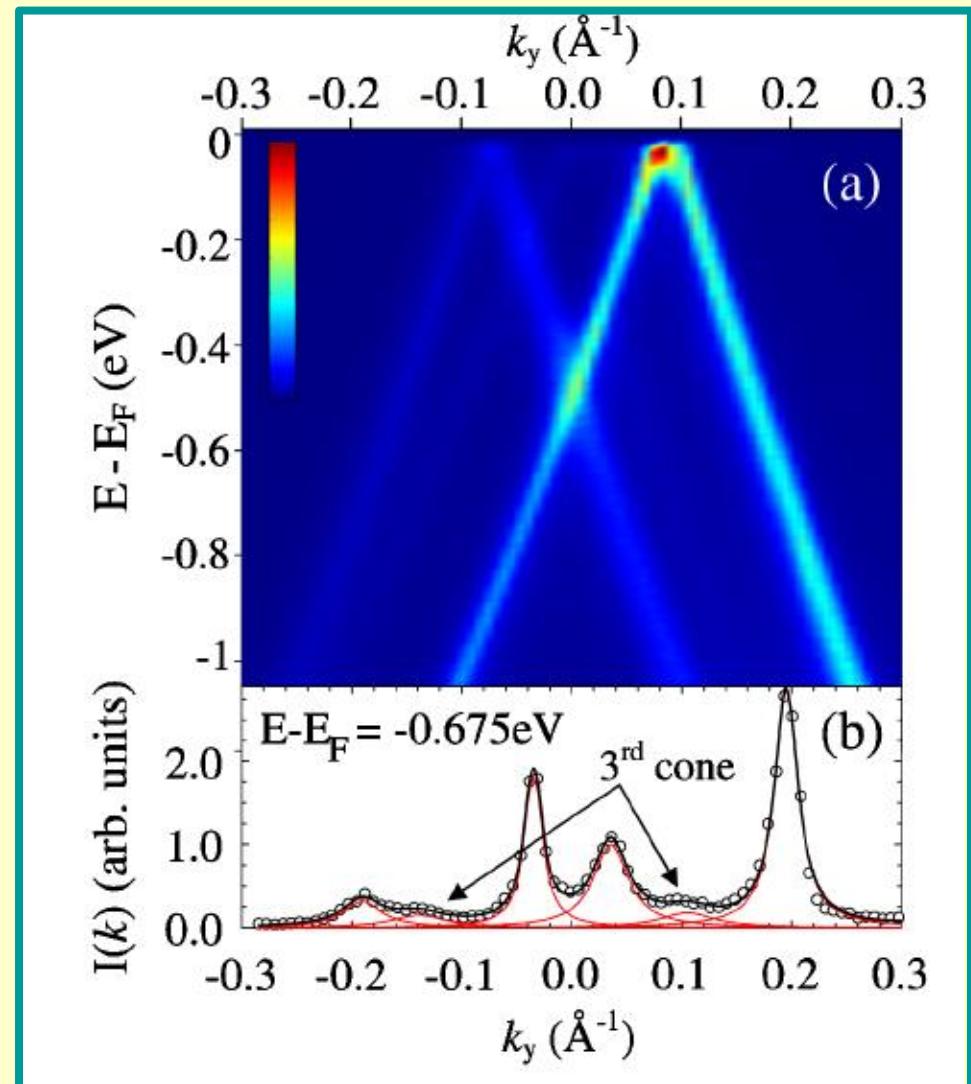


# Epitaxial MLG on C-face SiC

Charge distributions:



Sun, et al Phys. Rev. Lett **104**, 136802 (2010)



Sprinkle, et al Phys. Rev. Lett **103**, 226803 (2009)<sup>32</sup>

# *Electron-electron interactions: Two-layers*

$$\frac{\mathcal{Q}^{el}}{L^2} = \frac{\hbar}{4\pi^3} \int_{-\infty}^{\infty} \omega d\omega \int d\vec{q} |v_{LD,HD}^{sc}|^2 [n_B(T_{LD}) - n_B(T_{HD})] \\ \times \text{Im}\chi_{LD}(q, \omega, T_{LD}) \text{Im}\chi_{HD}(q, \omega, T_{HD})$$

$$v_{LD,HD}^{sc} = v_q / \epsilon^{RPA}(q, \omega, T_{LD}, T_{HD})$$

$$\epsilon^{RPA}(q, \omega, T_{LD}, T_{HD}) = (1 - v_q \chi_{LD}(q, \omega, T_{LD})) (1 - v_q \chi_{HD}(q, \omega, T_{HD})) \\ - v_q^2 e^{-2qd} \chi_{LD}(q, \omega, T_{LD}) \chi_{HD}(q, \omega, T_{HD})$$

# *Electron-electron interactions: two layers*

$$k_{F,LD}d \ll 1 \text{ and } q_{HD}^{TF}/k_{F,LD} \gg 1$$

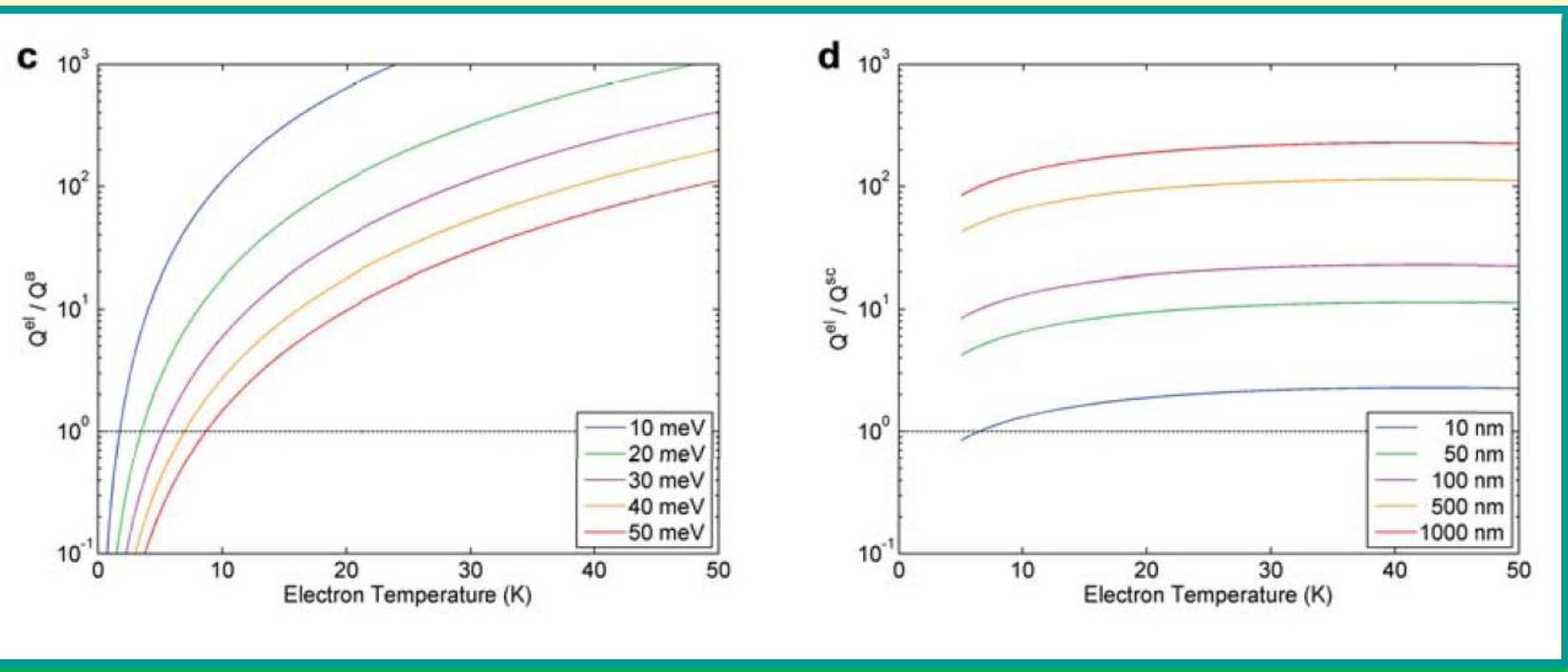
$$q_i^{TF} = qv_q\nu_i$$

$$\frac{\mathcal{Q}^{el}}{L^2} = \frac{E_{F,LD}^4 \nu_{LD}}{2\pi^2 v_F^2 \hbar^3 \nu_{HD}} \int_0^\infty \Omega d\Omega \int_0^\infty Q dQ \left( \frac{1}{e^{\Omega/t} - 1} \right) \left( \frac{\Omega}{Q} \right)^2$$

$$\frac{\mathcal{Q}^{el}}{L^2} = -\frac{E_{F,LD}^4 \pi^2 \nu_{LD}}{15 v_F^2 \hbar^3 \nu_{HD}} t^4 \ln t.$$

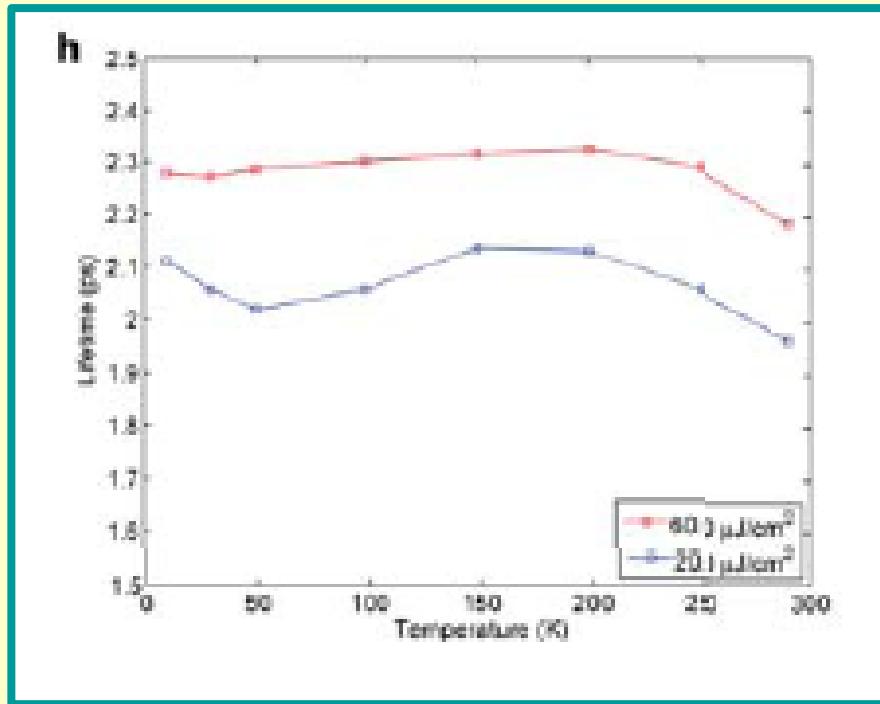
$$t = k_B T / E_{F,LD}$$

# *Electron-electron interactions: Interlayer*



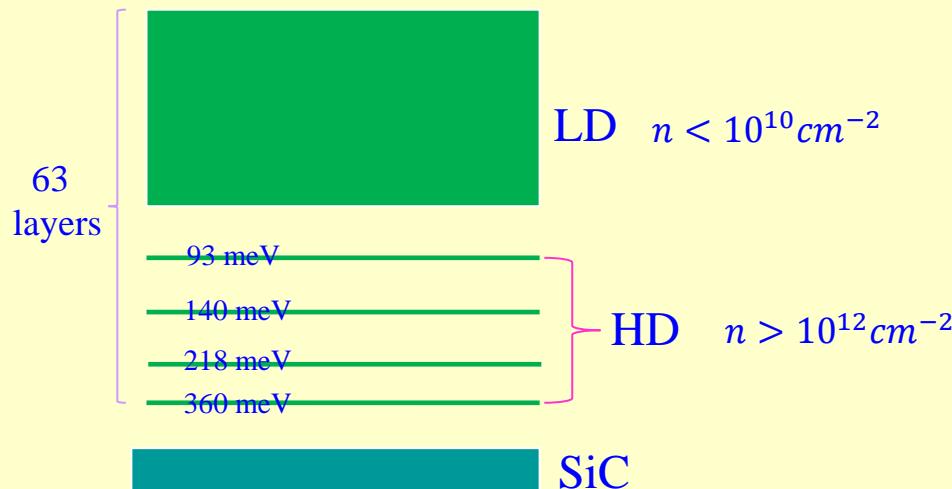
M. Mihney, J. Tolsma, C. David, D. Sun, R. Asgari, M. Polini, C. Berger, W. A. de Heer, A. H. MacDonald, T. Norris, Nature Commun. (2015)

# *Cooling lifetime in MEG*



$$\partial_t T_i = \left( \mathcal{Q}_i^{ph}(T_i) + \sum_{j \neq i} \mathcal{Q}_{ij}^{el}(T_i, T_j) \right) / C_i$$

# Power rate in MEG



$$\begin{aligned} Q_{ij}^{el} = & \frac{\hbar}{\pi} \int_{-\infty}^{\infty} d\omega \omega \sum_{\vec{q}} |v_{ij}^{sc}|^2 \\ & \times [n_B(\hbar\omega/T_i) - n_B(\hbar\omega/T_j)] \\ & \times \text{Im}\chi_i(q, \omega, T_i) \text{Im}\chi_j(q, \omega, T_j) \end{aligned}$$

$$\mathbf{v}_{ij}^{sc} = (\mathbf{v}^{-1} - \delta_{i,j} \chi_i(q, \omega, T_i))_{ij}^{-1}$$

# Cooling in multilayer: approximation

$$\begin{aligned}
 \mathcal{Q}_{ij}^{el} = & \frac{\hbar}{\pi} \int_{-\infty}^{\infty} \omega d\omega \sum_{\vec{q}} |v_{ij}^{sc}|^2 \\
 & \times \frac{\hbar\omega}{4T_L^2} \frac{\delta T}{\sinh^2(\hbar\omega/2T_L)} \\
 & \times \text{Im}\chi_i(q, \omega, T_L) \text{Im}\chi_j(q, \omega, T_L).
 \end{aligned}$$

$$\epsilon^{MEG}(q, \omega) = 1 - \frac{2\pi e^2}{\kappa q} \sum_{j \in HD} \chi_j(q, \omega, T_j).$$

$$\partial_t T_{LD} = \left( \sum_{j \in HD} \mathcal{Q}_{LD,j}^{el} (T_{HD} = T_L, T_{LD}, d_{j,LD}) \right) / \mathcal{C}_{LD}$$

$$\mathcal{C}_{LD} = 18\zeta(3)T_{LD}^2 / (\pi v_F^2)$$

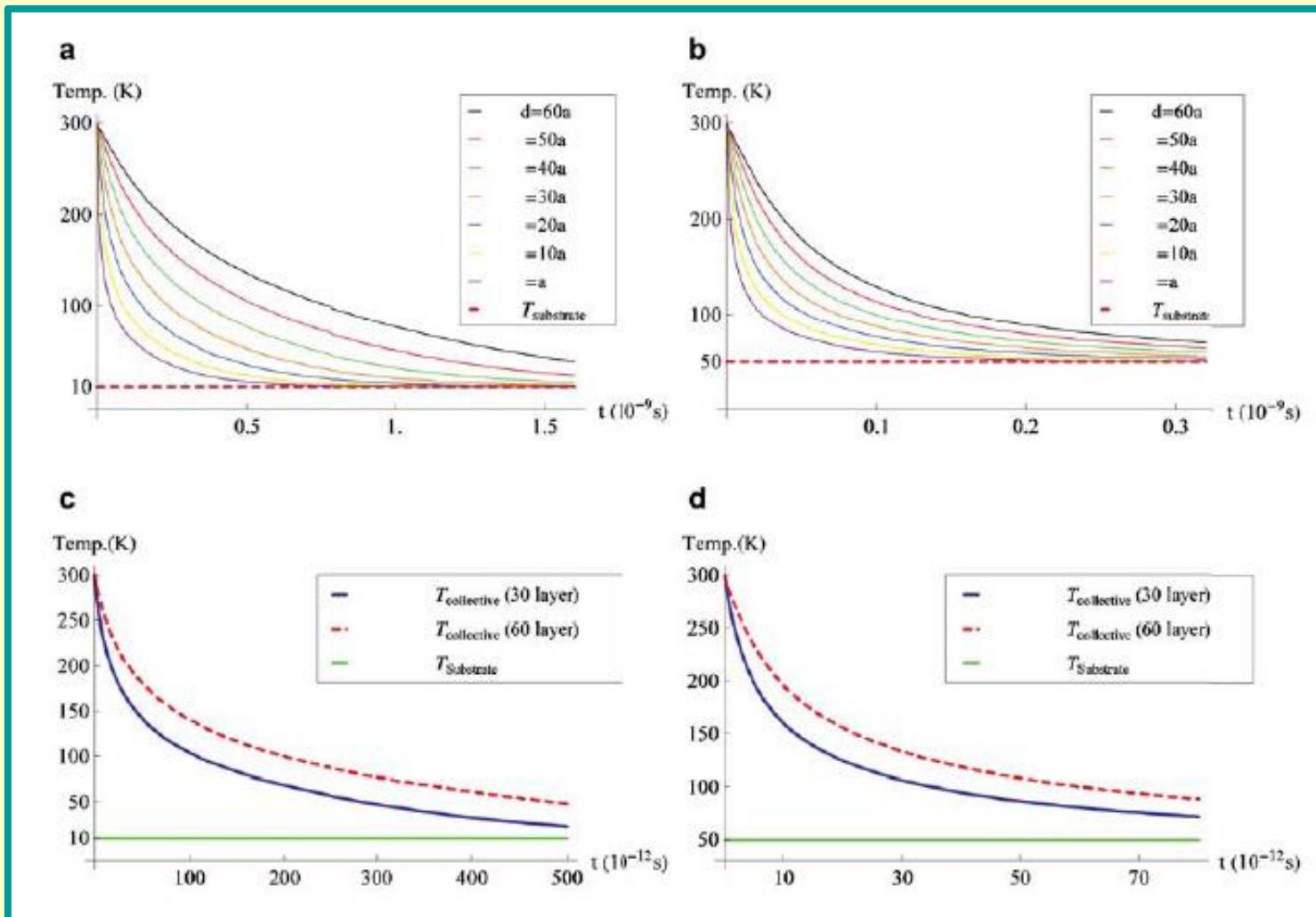
## *T(t) in MEG*

$$\partial_t T_{LD} = \left( \sum_{j \in HD} \mathcal{Q}_{LD,j}^{el}(T_{HD} = T_L, T_{LD}, d_{j,LD}) \right) / \mathcal{C}_{LD}$$

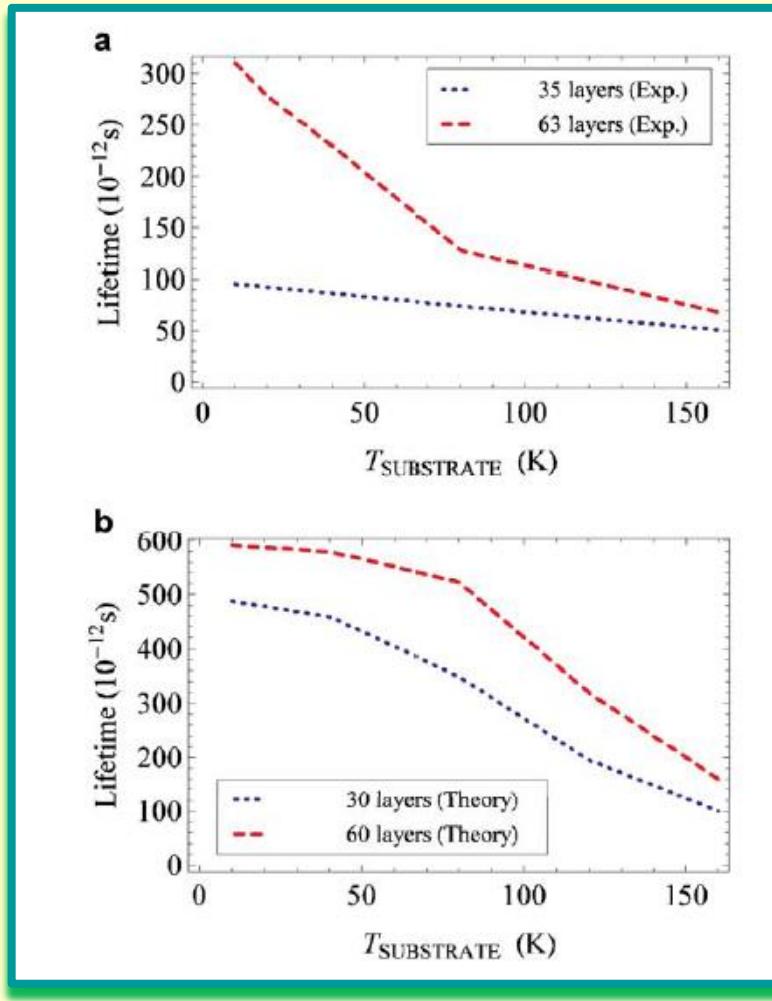
$$\partial_t T_c = \left( \sum_{i \in LD} \sum_{j \in HD} \mathcal{Q}_{ij}^{el}(T_L, T_{LD} \rightarrow T_c, d_{ij}) \right) / N \mathcal{C}_{LD}$$

$$\mathcal{C}_{LD} = 18\zeta(3)T_{LD}^2 / (\pi v_F^2)$$

# $T(t)$ in MEG



# *Cooling lifetime in MEG*



M. Mihney, J. Tolsma, C. David, D. Sun. R. Asgari, M. Polini, C. Berger, W. A. de Heer, A. H. MacDonald, T. Norris, Nature Commun. (2015)

# *Conclusions*

- Electron cooling mechanisms: e-ph, e-imp and e-e interactions  
 $\delta = 2+d$  ?
- Epitaxial graphene: C-face multilayer graphene on SiC substrate electronic cooling times ranging from a few to hundreds of picoseconds that strongly depend on the lattice temperature and the number of epitaxial graphene layers
- Developed a theory of hot-carrier equilibration based on interlayer energy transfer via screened Coulomb interactions
- Energy transfer between the LD layers is much stronger than between LD and HD layers
- The theoretically calculated thermal equilibration times are free of any fitting parameters, compare closely with the experimental relaxation times

# Thanks for your attention

