Microscopic Theory of Ultrafast Dynamics of Photo-excited Carriers in Graphene

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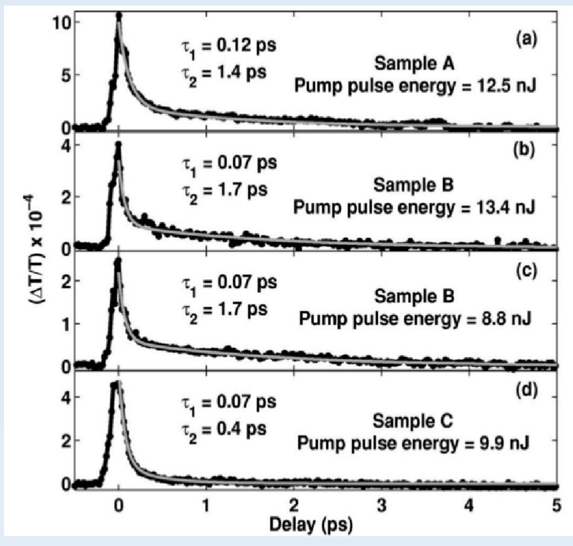
Introduction

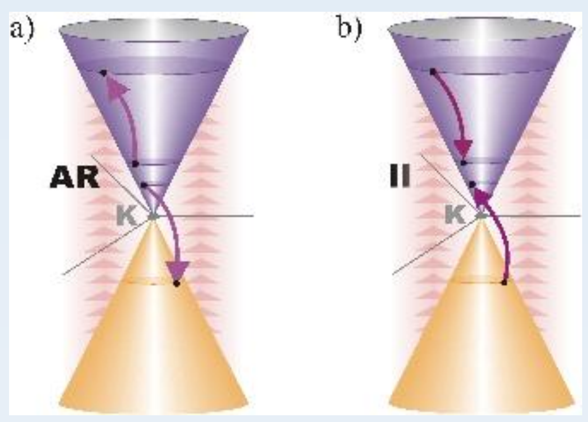
- Pump-probe measurement

  Fast differential transmission (DT) relaxation of several hundred femtoseconds, followed by a slower picosecond one is observed. [Dawlaty *et al.*, APL **92**, 042116 (08); Wang *et al.*, APL **96**, 081917 (10); Ruzicka *et al.*, APL **96**, 173106 (10); Hale *et al.*, PRB **83**, 121404(R) (11)]



[Dawlaty *et al.*, APL **92**, 042116 (08)]

- Auger process was claimed to be important
  - ✓ Under static screening, Auger process without inter-band coherence was claimed to be important. [Winzer *et al.*, Nano Lett. **10**, 4839 (10); Kim *et al.*, PRB **84**, 075449 (11); Winzer and Malic, PRB **85**, 241404(R) (12)]
  - ✓ Under dynamic screening, such process is forbidden. [Müller *et al.*, PRB **78**, 115406 (08)]



[Winzer *et al.*, Nano Lett. **10**, 4839 (10)]

  - ➢ Due to the energy conservation, such process must satisfy $v_F q = |\omega|$.
  - ➢ Corresponding dielectric function under dynamic screening diverges.

Model and Formalism

Effective Hamiltonian in graphene under a linear polarized light

$$H^{\mu}_{\rm eff}(\mathbf{k},t) = v_{\rm F}\{\mu\sigma_x[k_x + eA_x(t)] + \sigma_y[k_y + eA_y(t)]\}$$

$$\mu = 1\,(-1)\ \text{ for K (K}')\text{ valley;}\quad \mathbf{A}(t) = \mathbf{E}_0\sin(\omega t)\exp[-t^2/(2\sigma_t^2)]/\omega$$

Gauge invariant Green function [Haug and Jauho, *Quantum Kinetics in Transport and Optics of Semiconductors* (Springer, Berlin, 1998)]

$$G^{\mu}(\mathbf{k},\tau,T) = \int d^2r \exp\{-i[\mathbf{k} - \int_{-1/2}^{1/2} d\lambda\, |e|\, \mathbf{A}(T+\lambda\tau)]\cdot\mathbf{r}\} G^{\mu}(\tau,T,\mathbf{r})$$

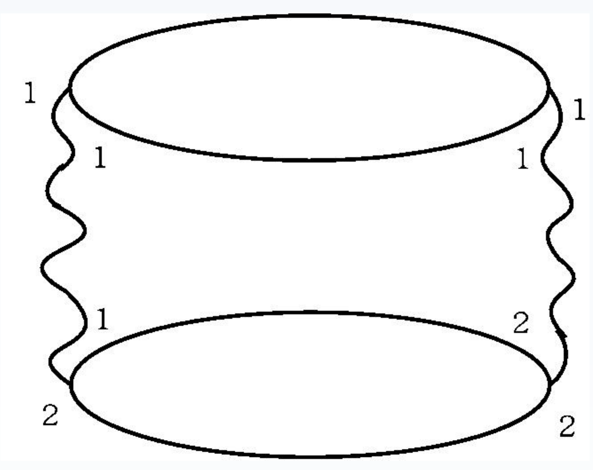
Gauge invariant kinetic equations

$$\partial_t \rho_{\mu\mathbf{k}} = \partial_t \rho_{\mu\mathbf{k}}|_{\rm coh} + \partial_t \rho_{\mu\mathbf{k}}|_{\rm drift} + \partial_t \rho_{\mu\mathbf{k}}|_{\rm scat}$$

$$\partial_t \rho_{\mu\mathbf{k}}|_{\rm coh} = -i[v_F k\sigma_z + \Sigma^{\rm HF}_{\mu\mathbf{k}} - |e|\, v_F \mu A_x \sin\theta_k \sigma_y, \rho_{\mu\mathbf{k}}]$$

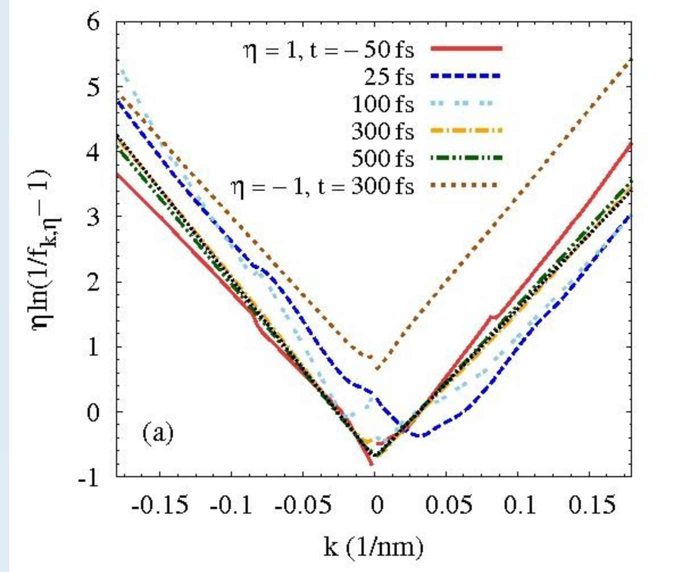
$$\partial_t \rho_{\mu\mathbf{k}}|_{\rm drift} = |e|\,\mathbf{E}\cdot\nabla_{\mathbf{k}}\rho_{\mu\mathbf{k}}$$

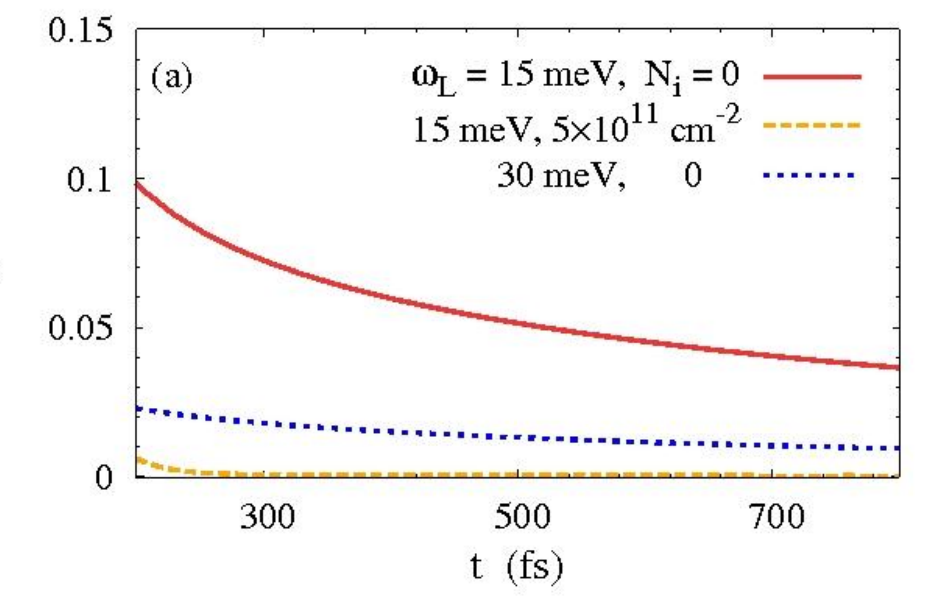
Auger process with inter-band coherence

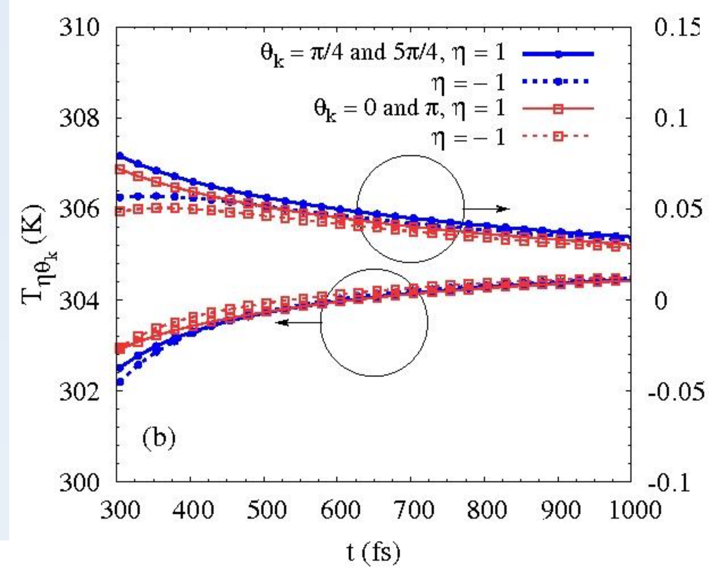


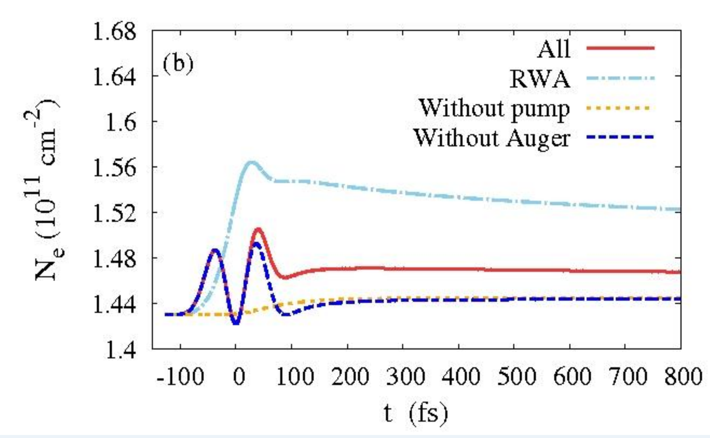
$$-\pi\sum_{\mathbf{q}}\sum_{\mathbf{k}_1\mu'} S^{\mu}_{\mathbf{k},\mathbf{k}-\mathbf{q},2,1}\rho^{>}_{\mu\mathbf{k}-\mathbf{q},1,2} S^{\mu}_{\mathbf{k}-\mathbf{q},\mathbf{k},2,2}\rho^{<}_{\mu\mathbf{k},2,2}$$
$$\times V^{r}(\mathbf{q},\varepsilon_{1,\mathbf{k}_1+\mathbf{q}} - \varepsilon_{1,\mathbf{k}_1}) V^{a}(\mathbf{q},\varepsilon_{1,\mathbf{k}_1+\mathbf{q}} - \varepsilon_{1,\mathbf{k}_1})$$
$$\times S^{\mu'}_{\mathbf{k}_1,\mathbf{k}_1+\mathbf{q},1,1}\rho^{>}_{\mu'\mathbf{k}_1+\mathbf{q},1,1} S^{\mu'}_{\mathbf{k}_1+\mathbf{q},\mathbf{k}_1,1,1}\rho^{<}_{\mu'\mathbf{k}_1,1,1} e^{-i\omega t}$$
$$\times \delta(\varepsilon_{2\mathbf{k}-\mathbf{q}} - \varepsilon_{2\mathbf{k}} + \varepsilon_{1\mathbf{k}_1+\mathbf{q}} - \varepsilon_{1\mathbf{k}_1})$$

Low Probe-Photon Energy



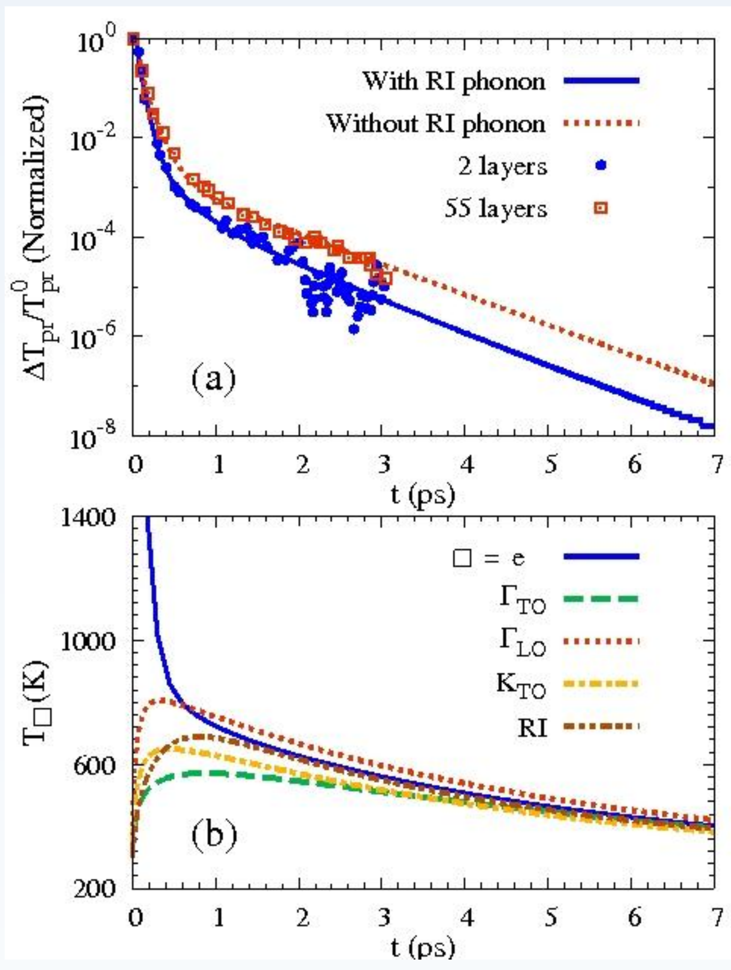




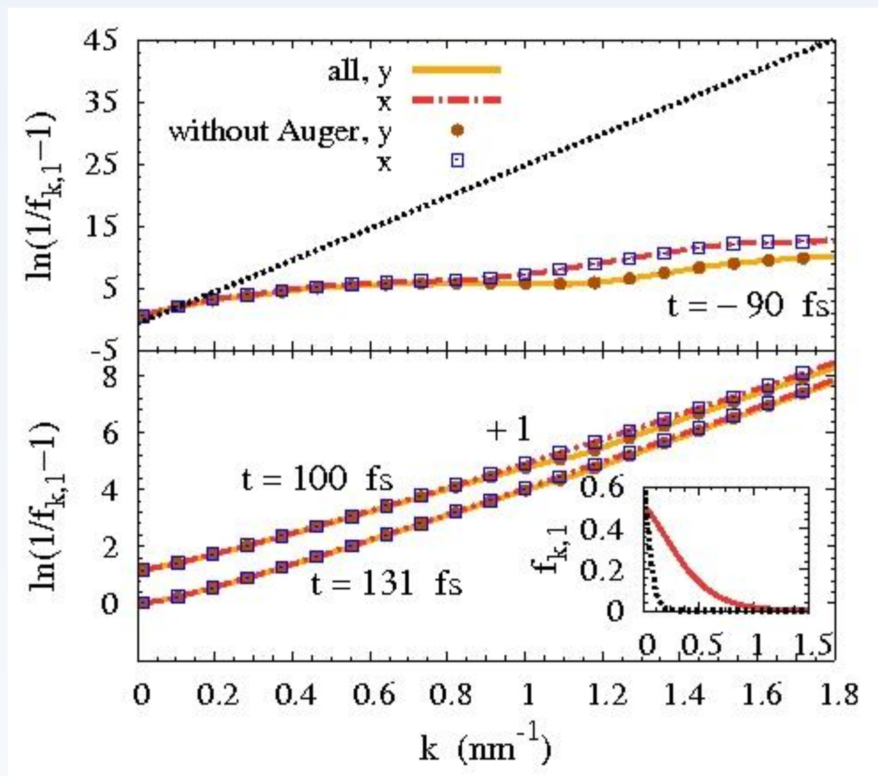


- Due to the strong Coulomb scattering, the drifted Fermi distribution established after pulse is
  $$f_{\mathbf{k}\eta} = \{\exp[\beta_\eta(\eta v_F k - \mathbf{u}_\eta\cdot\mathbf{k} - \mu_\eta)] + 1\}^{-1}$$
- This is different from the one established under static electric field: [Y. Zhou and M. W. Wu, PRB **82**, 085304 (10)]
  $$f_{\mathbf{k}\eta} = [\exp[\beta_\eta(\eta v_F|\mathbf{k} - \mathbf{u}_\eta| - \mu_\eta) + 1]^{-1}$$
- Auger process involving inter-band coherence contributes markedly.
- Rotating-wave approximation (RWA) is no longer valid.
- Drift term contributes to the excitation of electrons from conduction band to valence band.

High Probe-Photon Energy

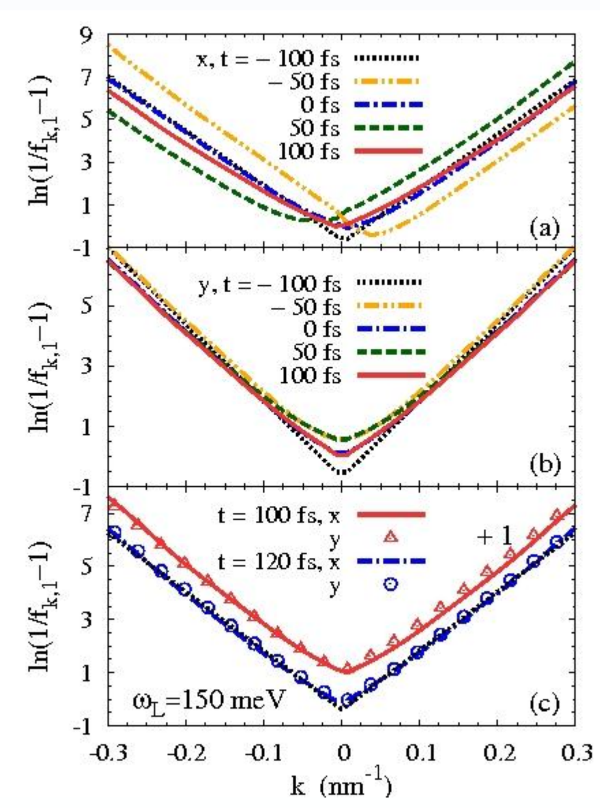


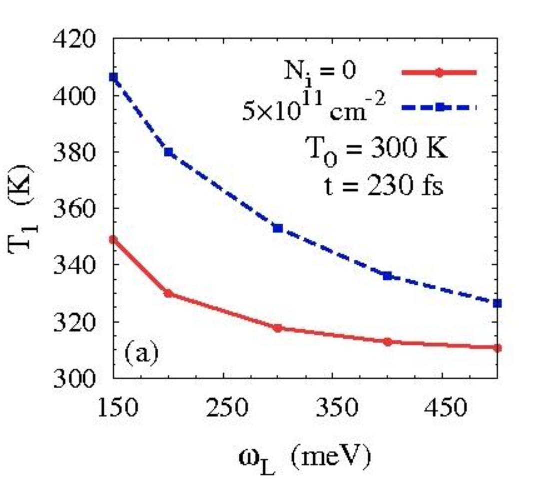
[Wang et al., APL **96**, 081917 (10)]

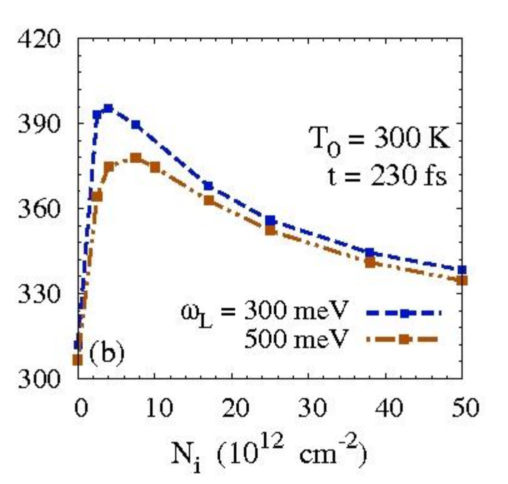


- Electrons are photoexcited anisotropically.
- Under the scattering, isotropic hot-electron Fermi distribution is found to be established within t = 131 fs.
- Fast relaxation of DT is due to the rapid equilibration of the carrier-phonon system and the slow one is from the slow hot-phonon decay.
- RI phonons are important only when the number of the graphene layers is small and they are responsible for the difference between the DTs in the graphene samples with few and many layers.

Influence of Drift Term







- Pump process is switched off.
- Influence of the drift term decreases with the increase of the pump photon energy and it is negligible when the pump-photon energy is high enough.

Summary

- Auger process investigated in the literature which only involves the diagonal terms of density matrices is forbidden by the dynamic screening. However, Auger process involving the inter-band coherence contributes to the dynamics of carriers, when the pump-photon energy is low.
- When the pump-photon energy is low, a large net momentum is transferred to electrons through the drift term and a drifted Fermi distribution different from the one under static field is established.
- The experimental observed fast relaxation of DT comes from the rapid equilibration of the carrier-phonon system and the slow one is due to the slow hot-phonon decay.
- The rotating-wave approximation is no longer valid when the pump-photon energy is low.

Reference:
B. Y. Sun, Y. Zhou, and M. W. Wu, Phys. Rev. B **85**, 125413 (2012).
B. Y. Sun and M. W. Wu, arXiv:1302.3677.