VIII-th International Conference "SOLITONS, COLLAPSES AND TURBULENCE: Achievements, Developments and Perspectives" (SCT-17) in honor of Evgeny Kuznetsov's 70th birthday

Optics of graphene in strong THz field

I.V. Oladyshkin, S.B. Bodrov, Yu.A. Sergeev, M.D. Tokman, A.N. Stepanov, A.I. Korytin

Institute of Applied Physics of the Russian Academy of Sciences (IAP RAS)
Nizhny Novgorod, Russia

May, 21-25, 2017 Chernogolovka, Russia

Outline

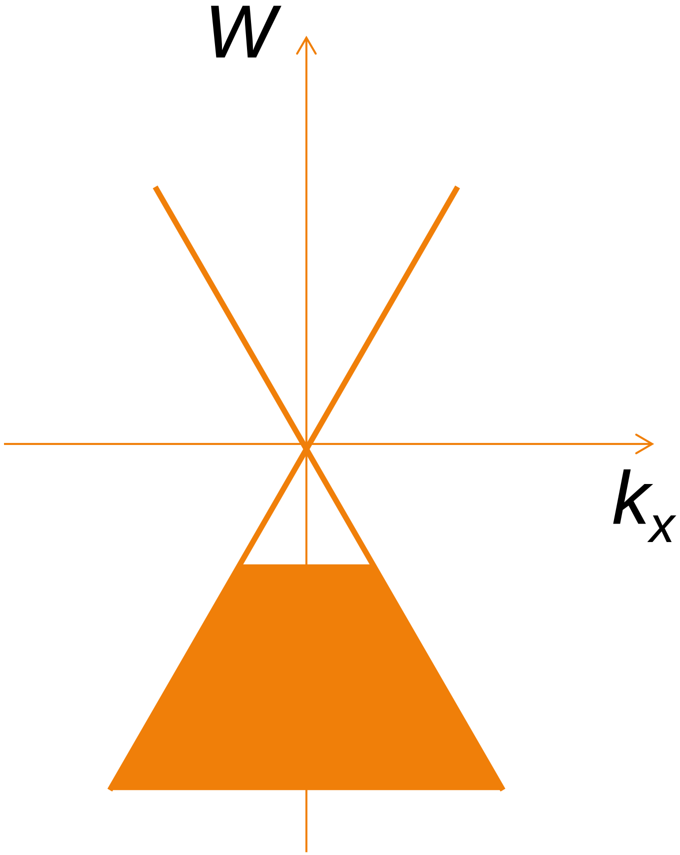
- Introduction: ultrafast carrier dynamics in graphene
- Graphene “luminescence” in THz field
  - idea: electron-hole pair production
  - experiment
- Second harmonic generation in THz field
  - experimental data
  - possible theoretical model
- Future plans

Introduction

Linear dispersion relation in graphene

<u>strong nonlinear effects?</u>

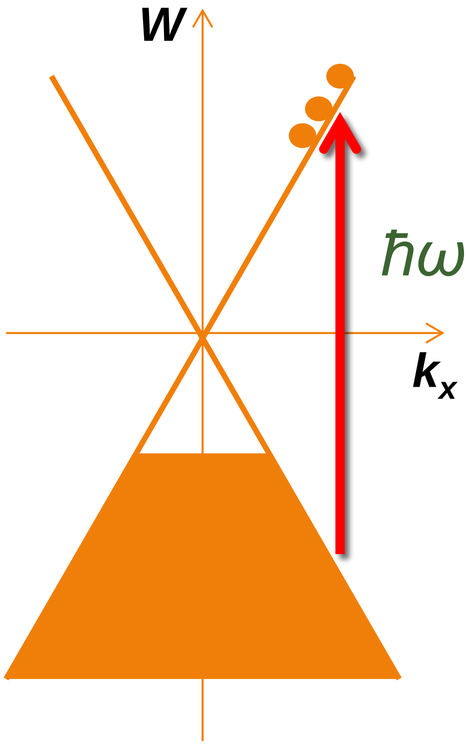
$$W_{c,\upsilon} = \pm \hbar \upsilon_F \sqrt{k_x^2 + k_y^2}$$



Introduction

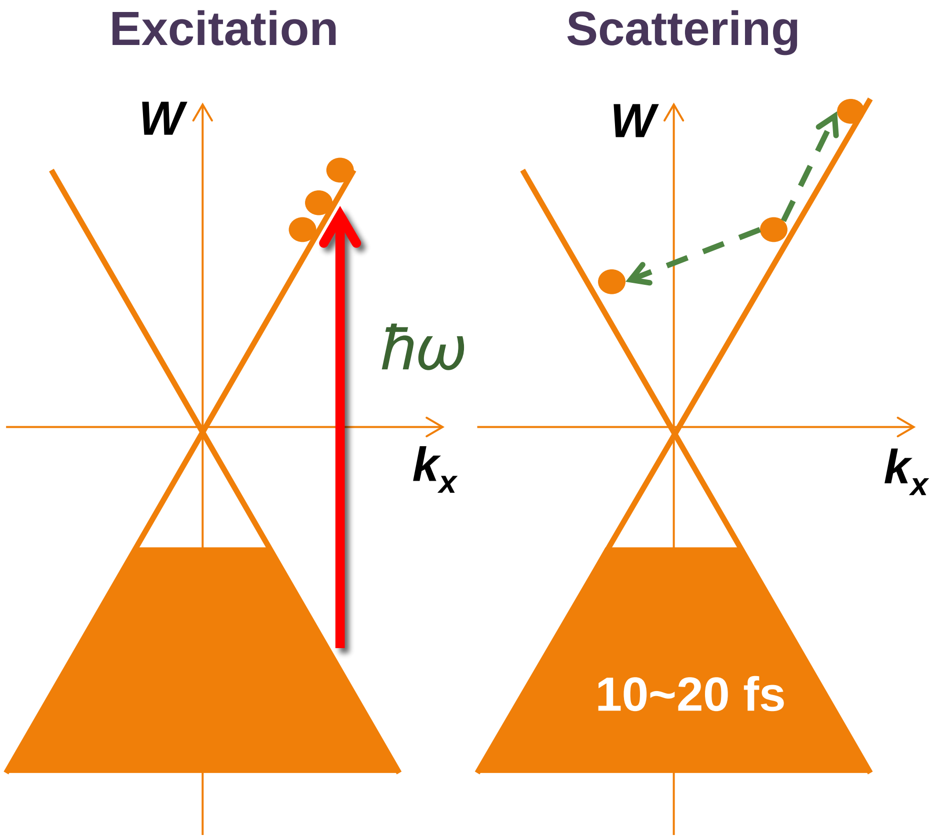
- Experiments on carrier dynamics in graphene

Excitation



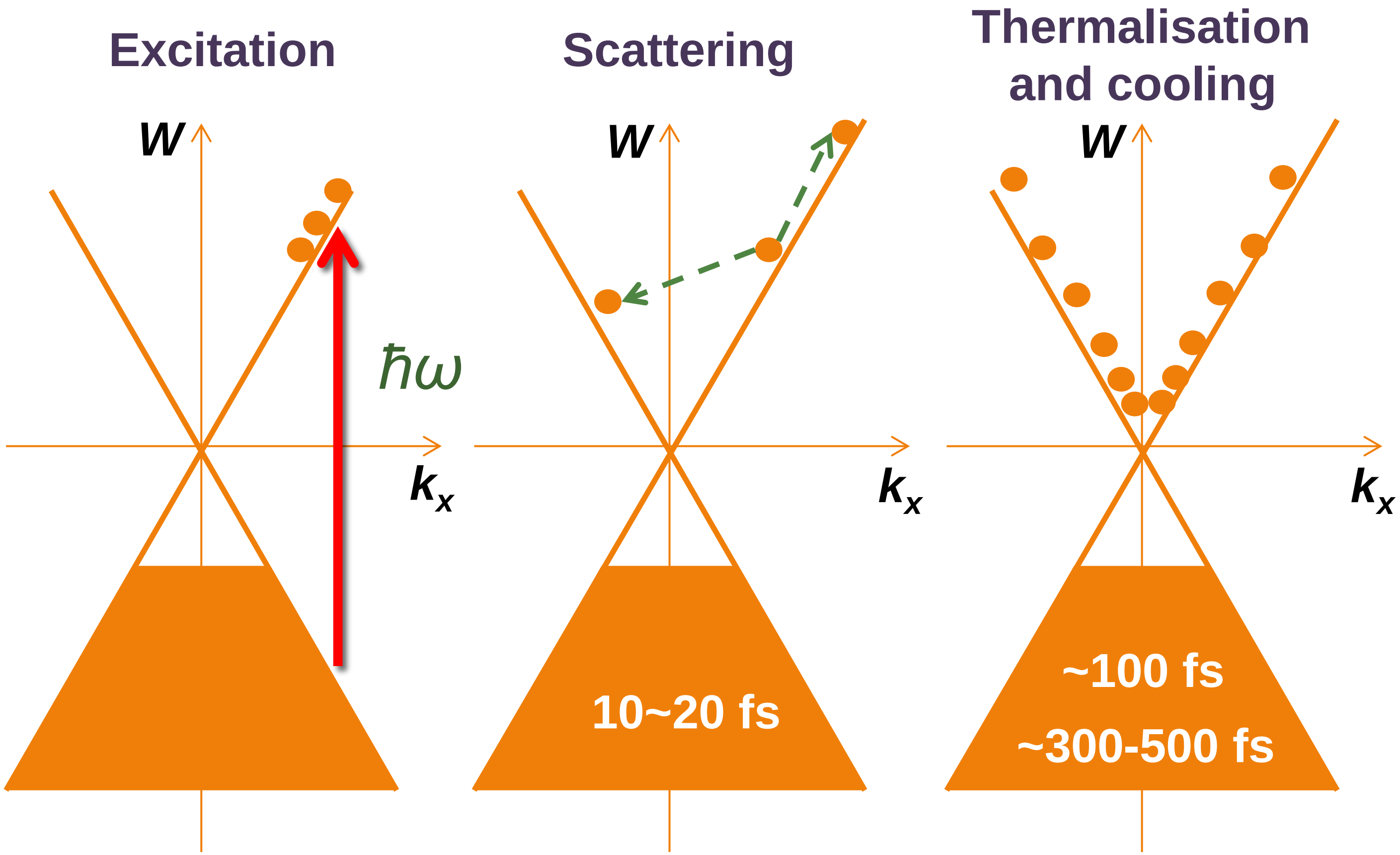
Introduction

- Experiments on carrier dynamics in graphene



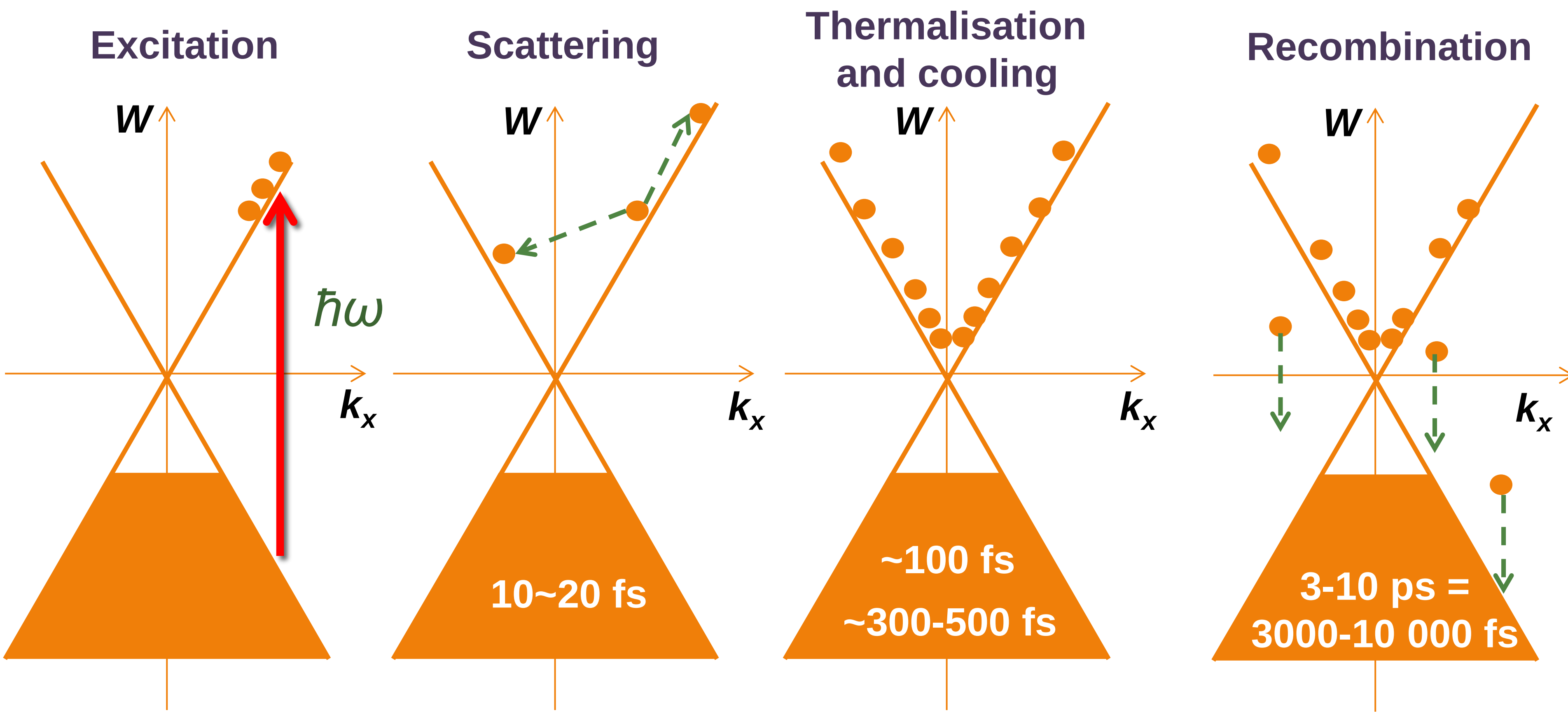
Introduction

- Experiments on carrier dynamics in graphene



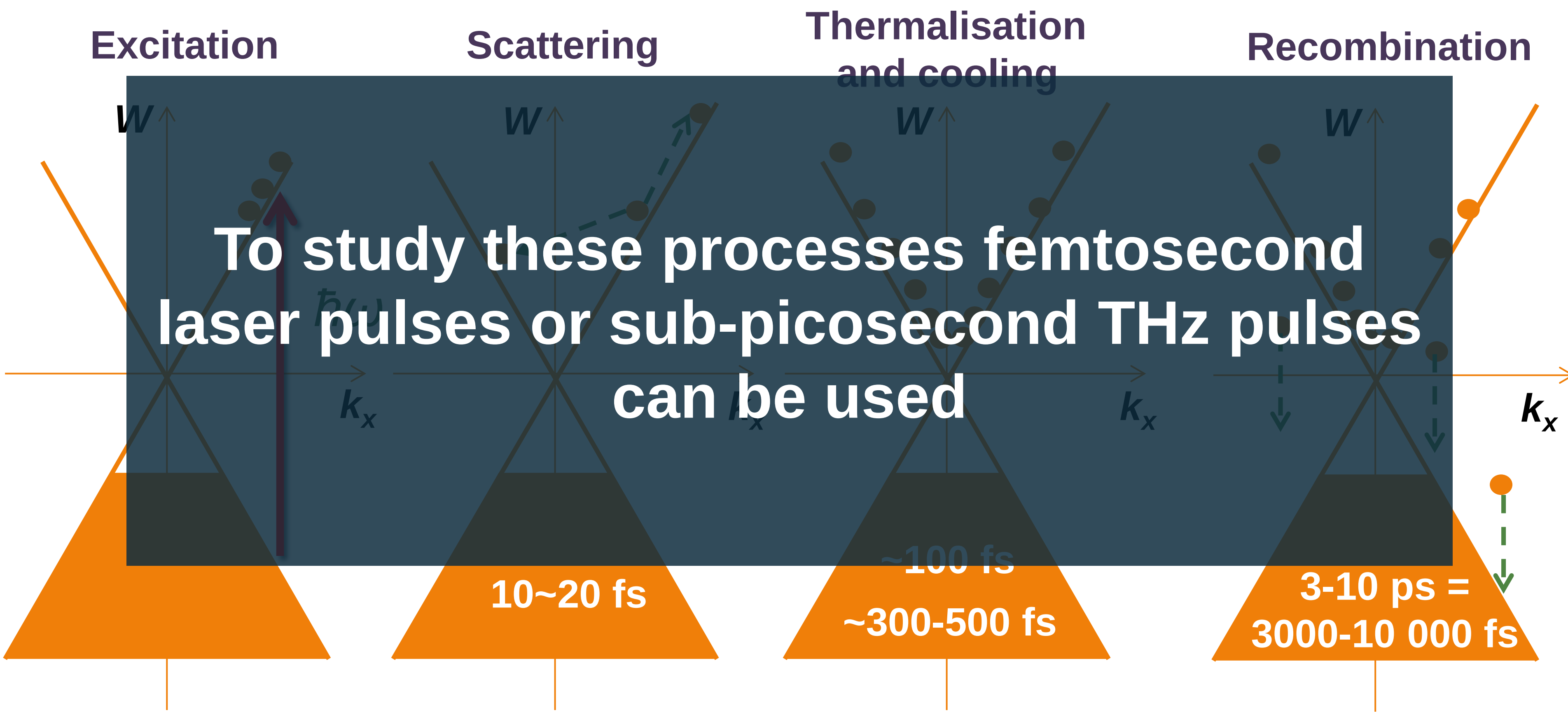
Introduction

- Experiments on carrier dynamics in graphene



Introduction

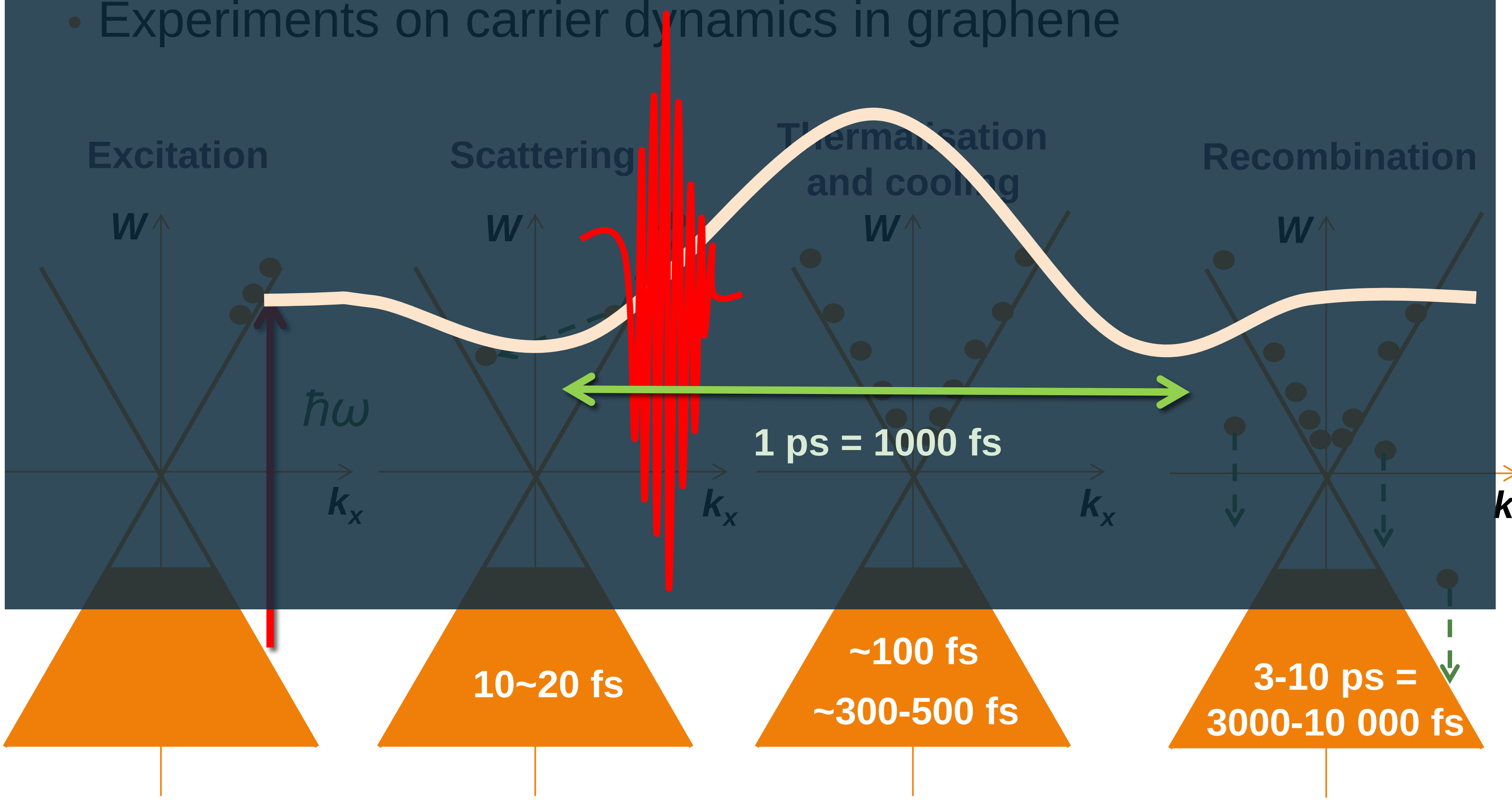
- Experiments on carrier dynamics in graphene



To study these processes femtosecond laser pulses or sub-picosecond THz pulses can be used

Introduction

- Experiments on carrier dynamics in graphene

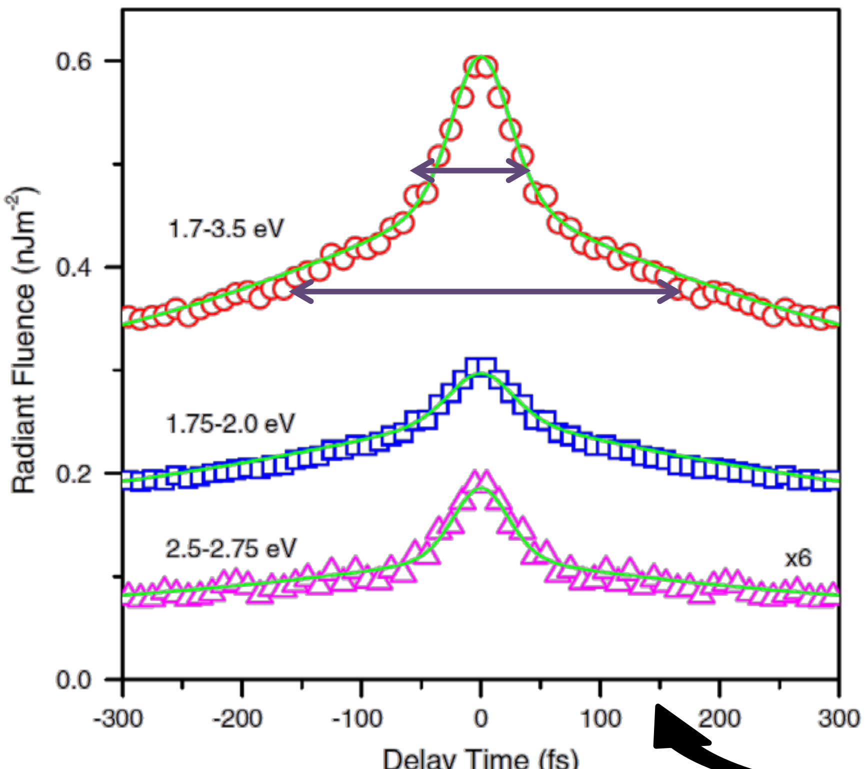


Introduction: photoluminescence

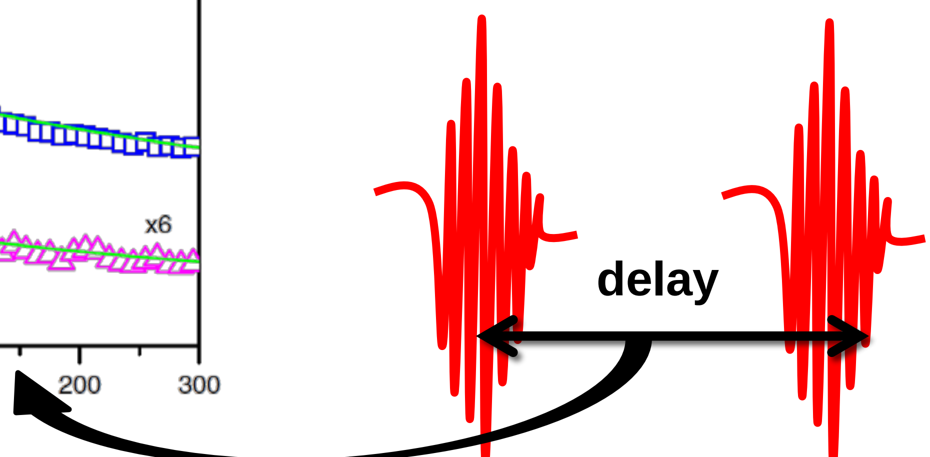
PRL 105, 127404 (2010) PHYSICAL REVIEW LETTERS week ending 17 SEPTEMBER 2010

Ultrafast Photoluminescence from Graphene

Chun Hung Lui (呂振鴻),[1] Kin Fai Mak,[1] Jie Shan,[2] and Tony F. Heinz[1,*]



Graphene luminescence after the action of two 30-fs laser pulses

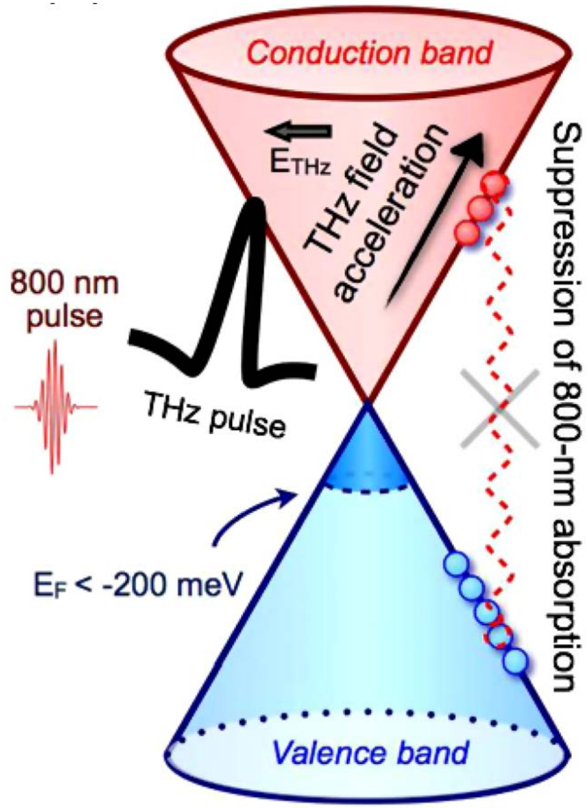


Introduction: THz action

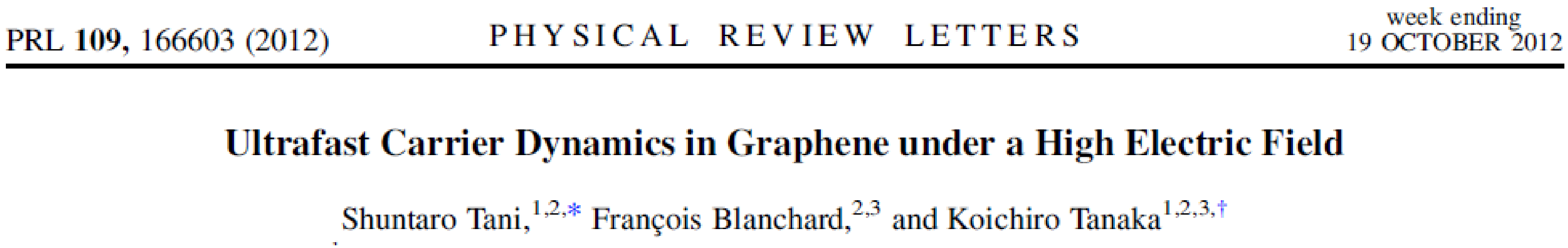
PRL 109, 166603 (2012) PHYSICAL REVIEW LETTERS week ending 19 OCTOBER 2012

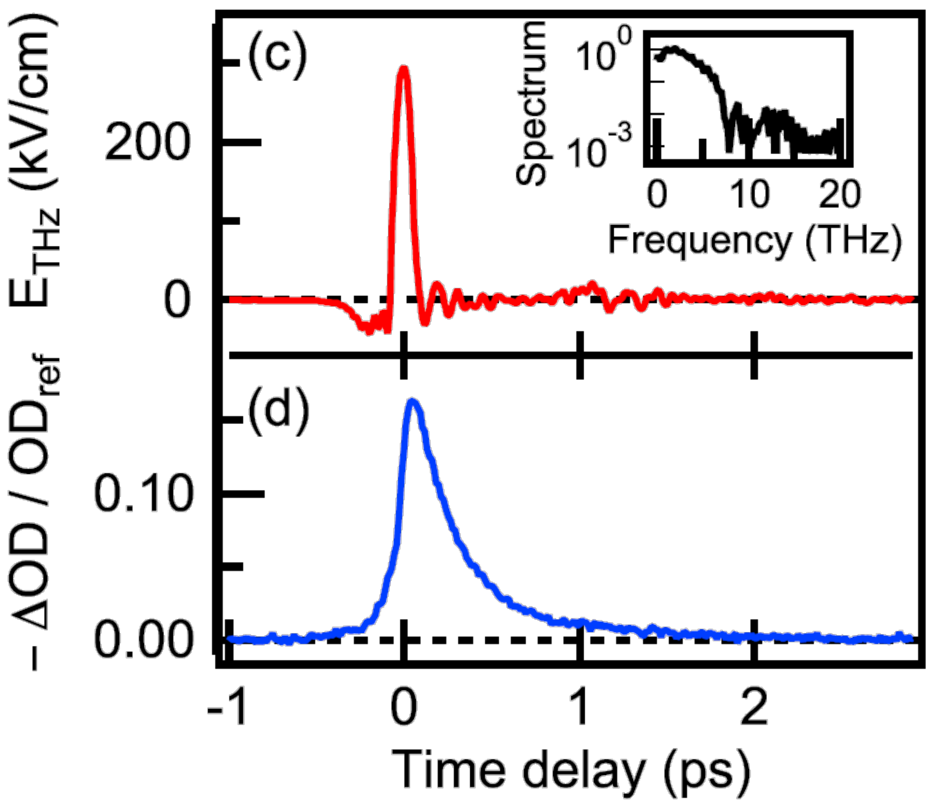
Ultrafast Carrier Dynamics in Graphene under a High Electric Field

Shuntaro Tani,[1,2,*] François Blanchard,[2,3] and Koichiro Tanaka[1,2,3,†]



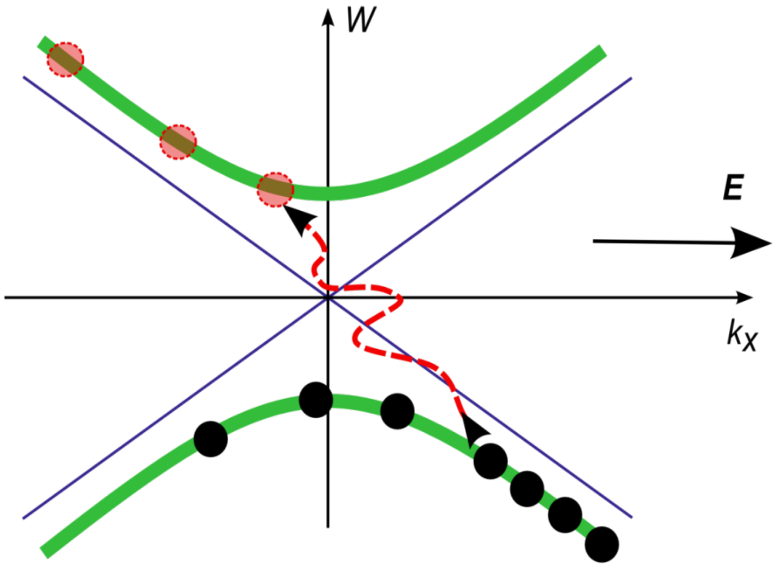
Introduction: THz action





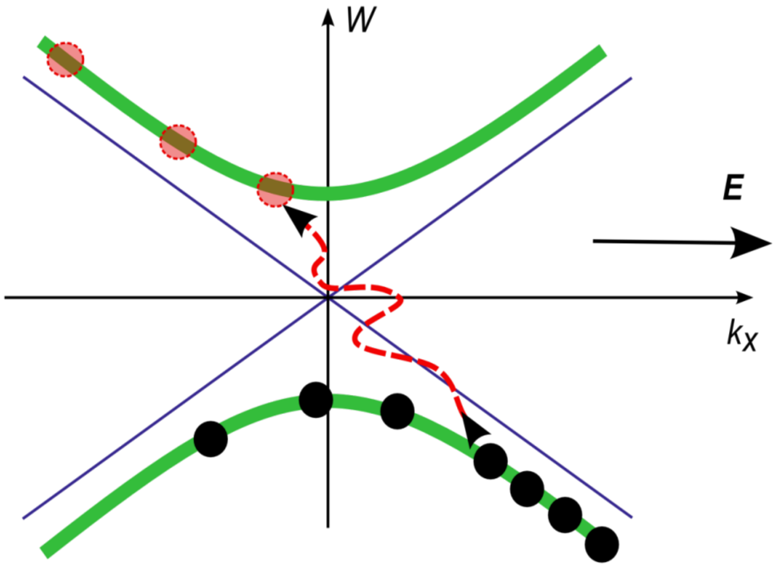
Number of electrons in the upper band was up to 10 times grater than the initial number of holes

“New” idea: Landau-Zener transitions



$$\hbar \dot{k}_x = -eE$$

"New" idea: Landau-Zener transitions

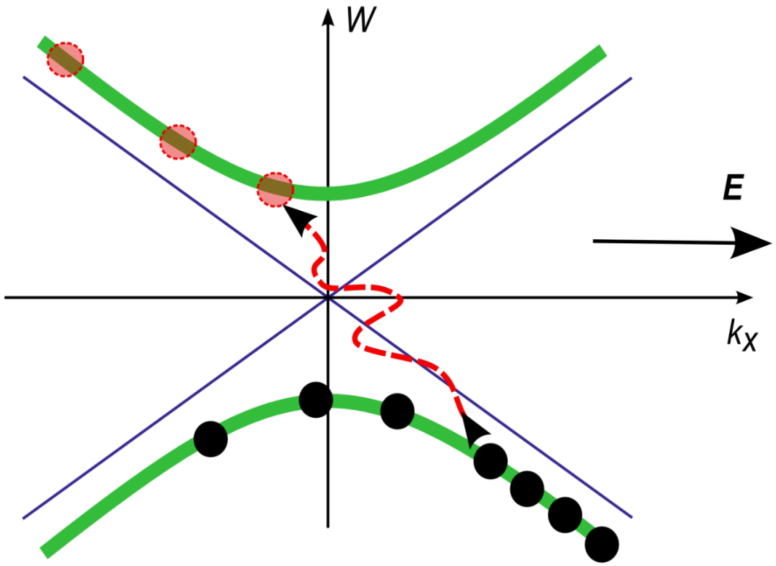


$$\hbar \dot{k}_x = -eE$$

$$P_{LZ} = \exp\left(-2\pi\Gamma\right)$$

$$\Gamma = \hbar^{-1}\left(\Delta W_{min}/2\right)^2 \left|\partial(W_+ - W_-)/\partial t\right|^{-1}$$

"New" idea: Landau-Zener transitions



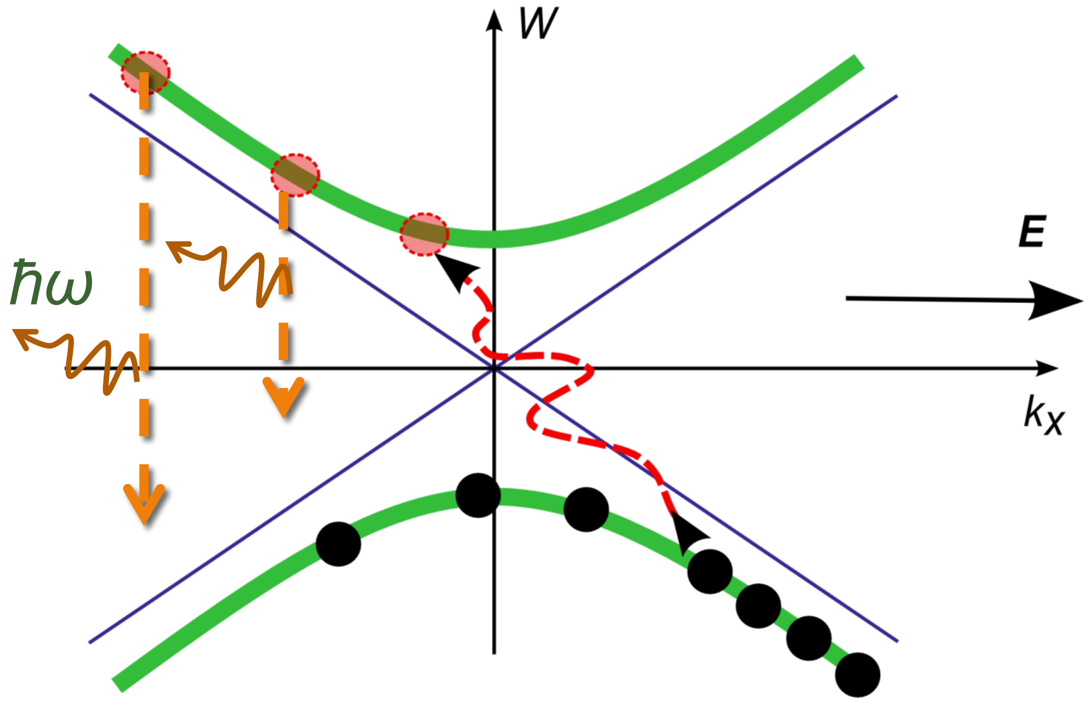
$$\hbar \dot{k}_x = -eE$$

$$P_{LZ} = \exp\left(-2\pi\Gamma\right)$$

$$\Gamma = \hbar^{-1}\left(\Delta W_{min}/2\right)^2 \left|\partial(W_+ - W_-)/\partial t\right|^{-1}$$

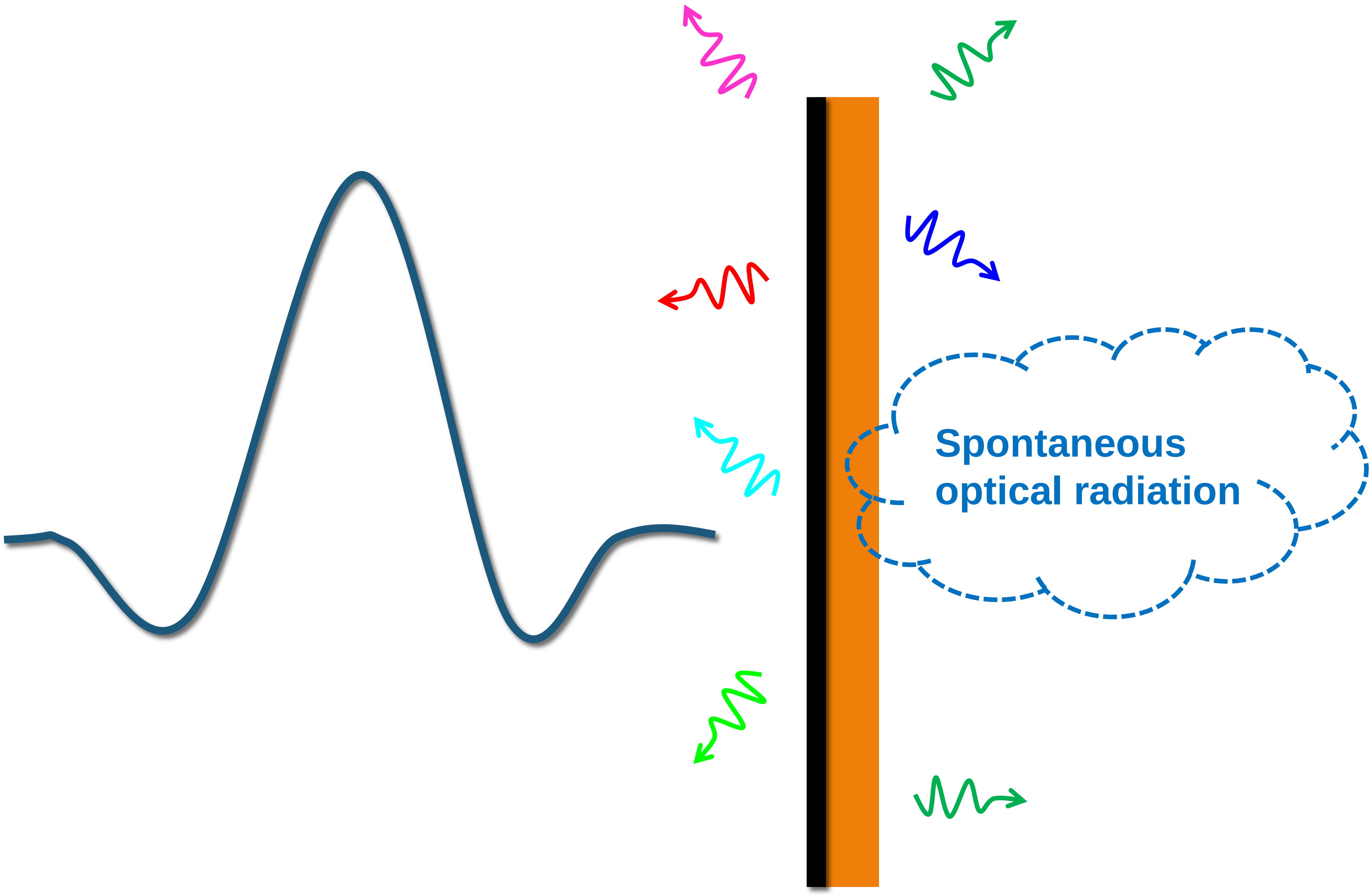
$$\Rightarrow \quad P_{LZ} = \exp\left(-\pi\hbar\upsilon_F k_y^2 / |eE|\right)$$

“New” idea: Landau-Zener transitions

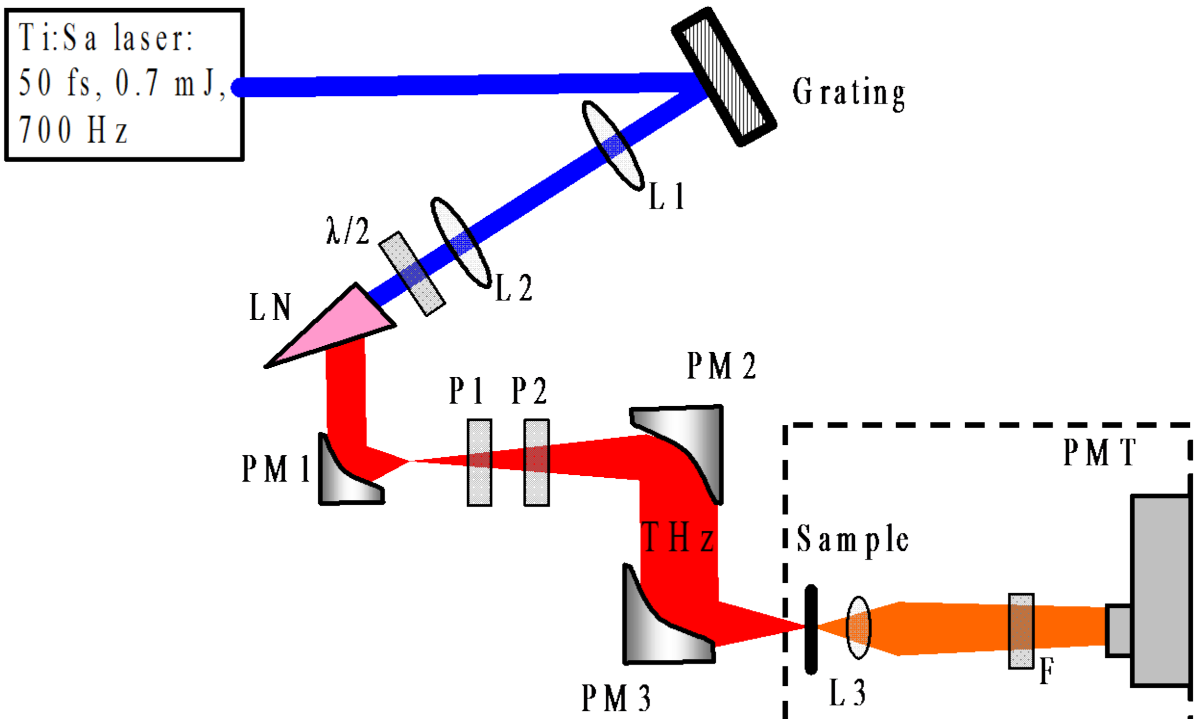


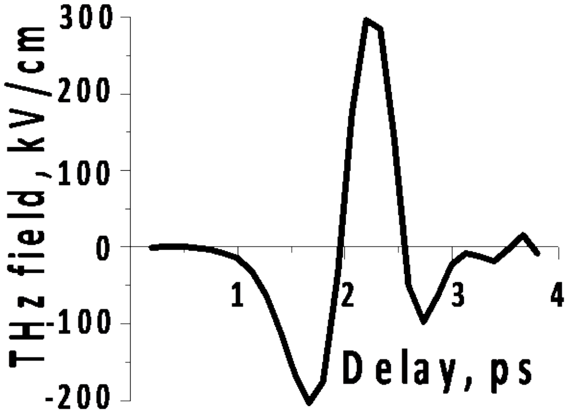
THz-induced population inversion
=> spontaneous emission

Experiment idea



Experimental setup





Detection range: from 350 nm to 600 nm

Spontaneous optical emission

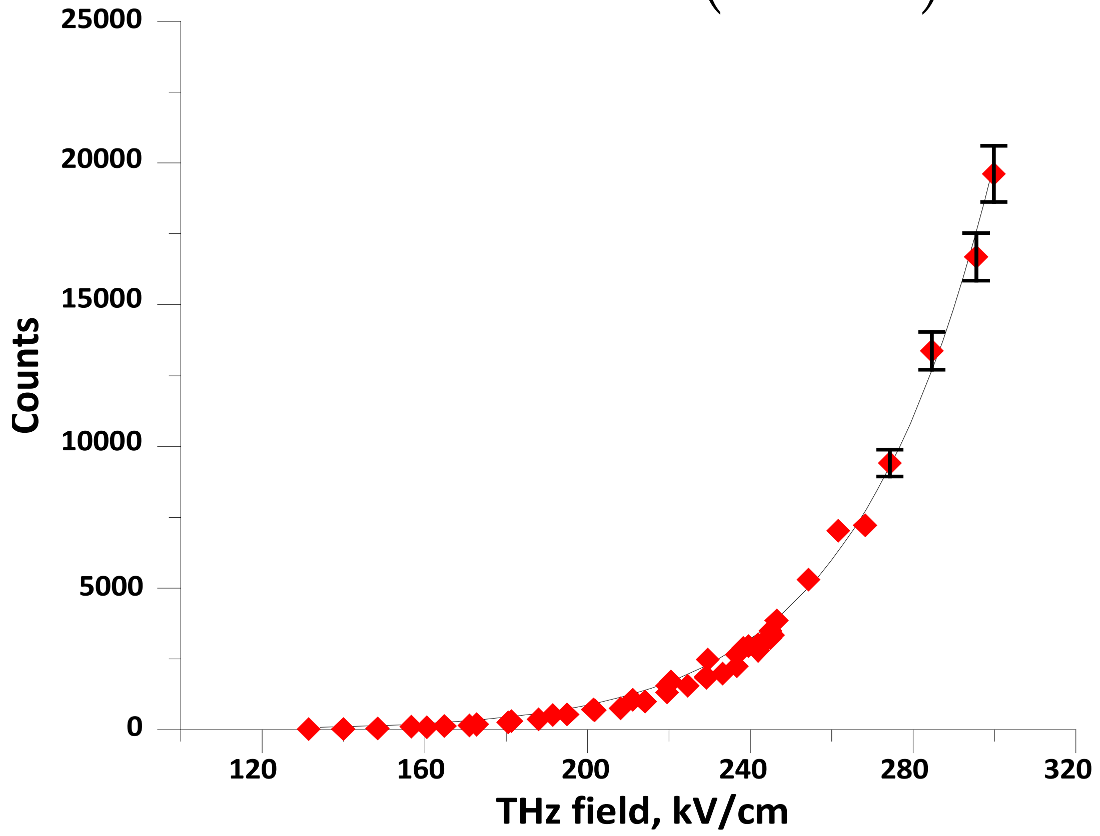
- Photons up to 3.5 eV observed
- Radiation pattern is almost spherical

* Photons are partially polarized

  – because the electron distribution is asymmetric?

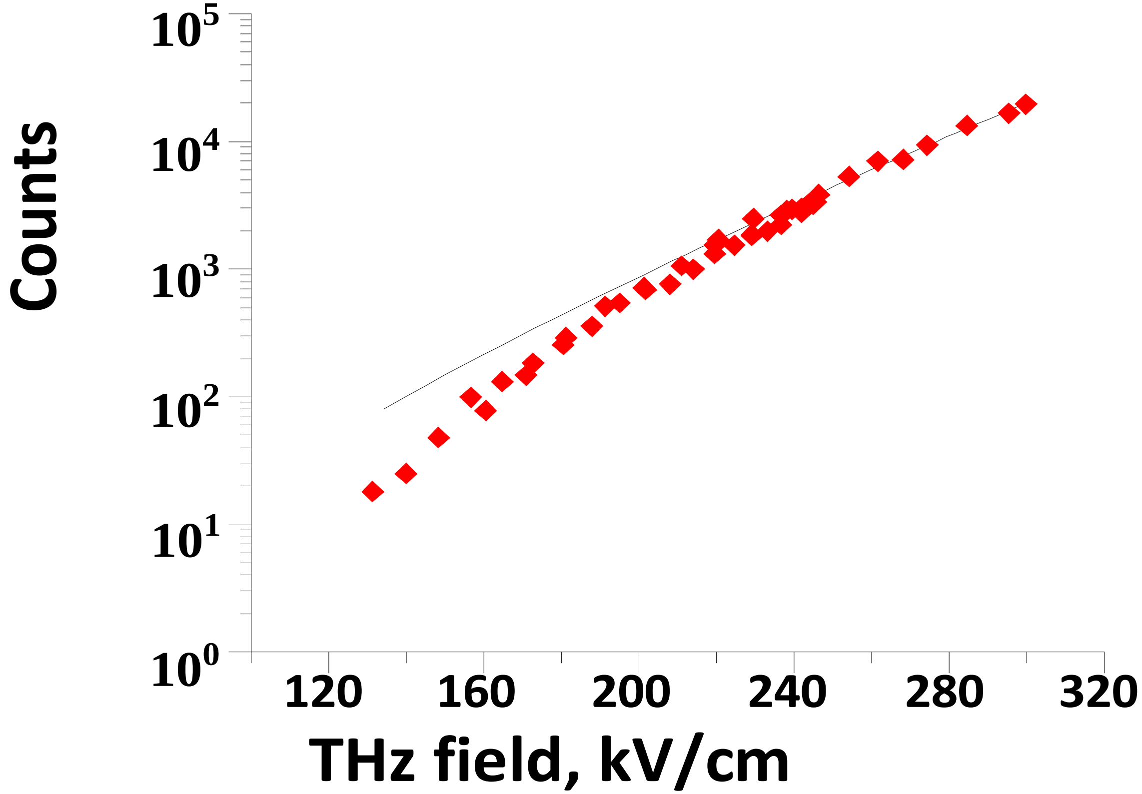
Spontaneous optical emission

$$N_{\text{detected}} \propto \exp\left(E_{THz}{}^{3/4}\right)$$

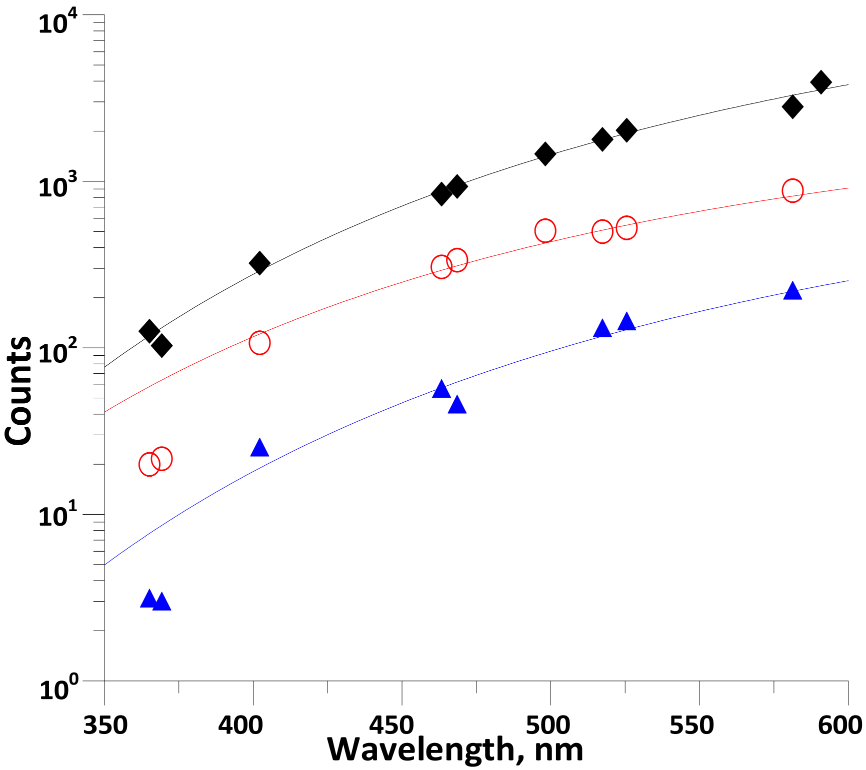


Spontaneous optical emission

$$N_{\mathrm{detected}} \propto \exp\left(E_{THz}{}^{3/4}\right)$$



Spontaneous optical emission: spectrum



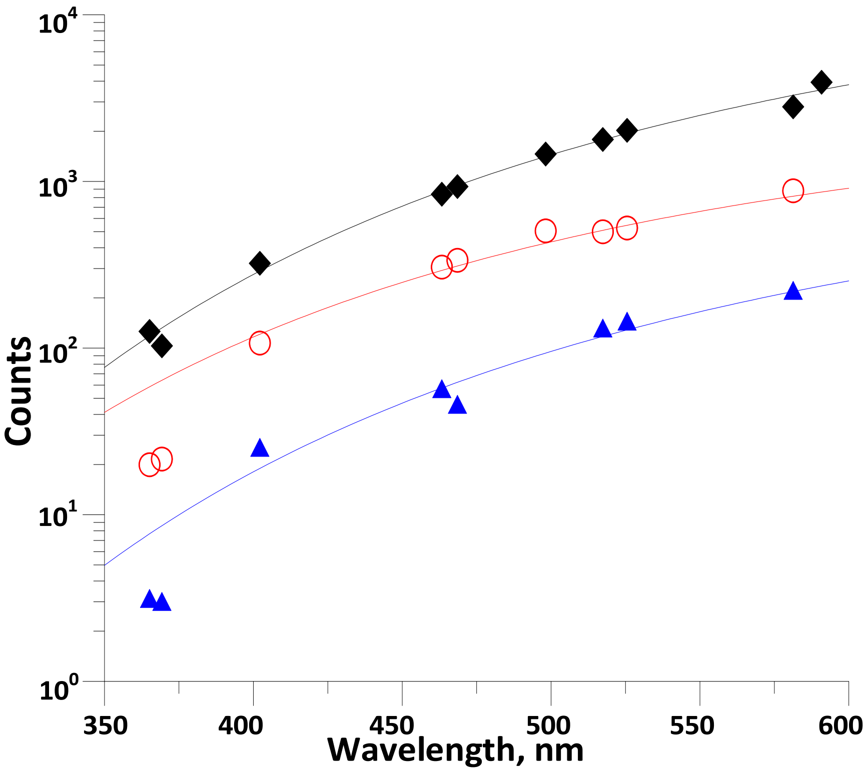
Black: E_{THz} = 300 kV/cm, T = 0.25 eV

Red: E_{THz} = 250 kV/cm, T = 0.28 eV

Blue: E_{THz} = 206 kV/cm, T = 0.25 eV

$$P_0(\omega, T) \propto \omega^2 \exp(-\hbar\omega/T)$$

Spontaneous optical emission: spectrum



$$P_0(\omega, T) \propto \omega^2 \exp(-\hbar\omega/T)$$

Black: E_{THz} = 300 kV/cm, T = 0.25 eV

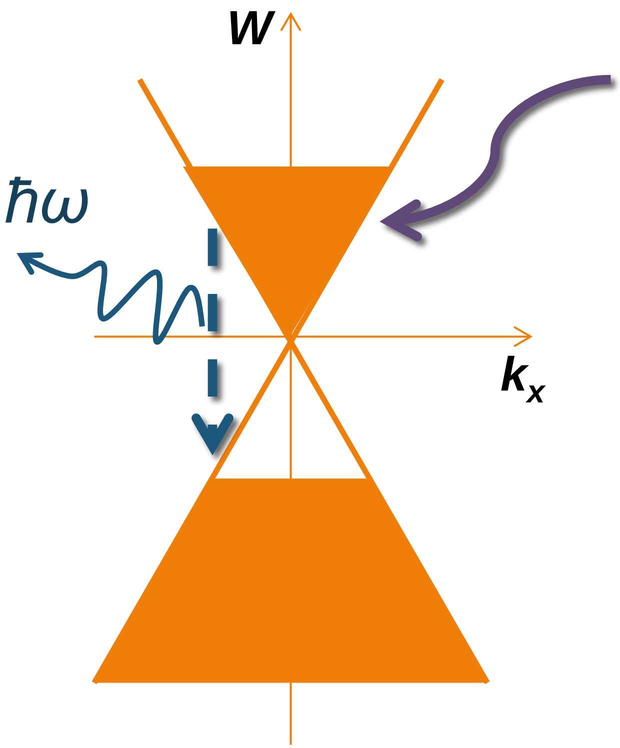
Red: E_{THz} = 250 kV/cm, T = 0.28 eV

Blue: E_{THz} = 206 kV/cm, T = 0.25 eV

This is not a black-body radiation

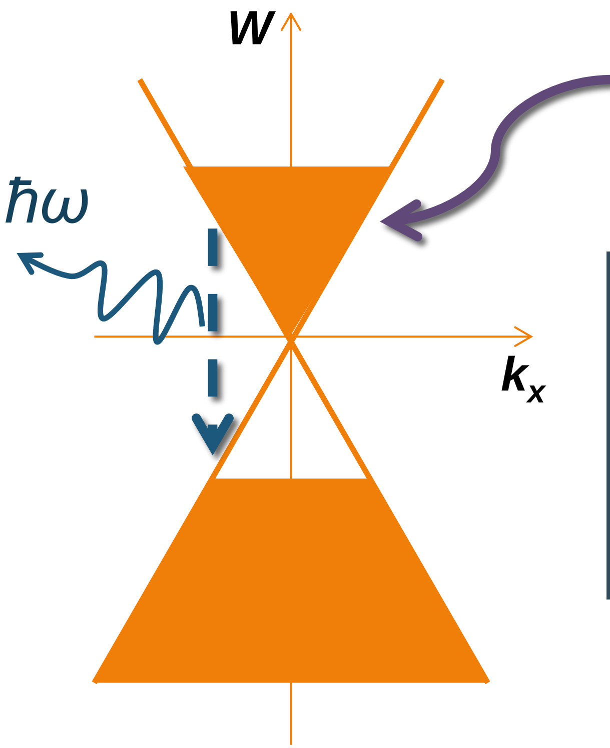
THz action is not simply heating

Spontaneous emission: bi-Fermi model



$$N_{LZ-electrons} \approx \pi^{-2} \upsilon_F^{-1/2} \left| \hbar^{-1} e E_{THz} \right|^{3/2} \Delta t_{\text{eff}}$$

Spontaneous emission: bi-Fermi model



$$N_{LZ-electrons} \approx \pi^{-2} \upsilon_F^{-1/2} \left| \hbar^{-1} e E_{THz} \right|^{3/2} \Delta t_{\mathrm{eff}}$$

Spontaneous optical emission of this system seemed to be suitable for experiment interpretation

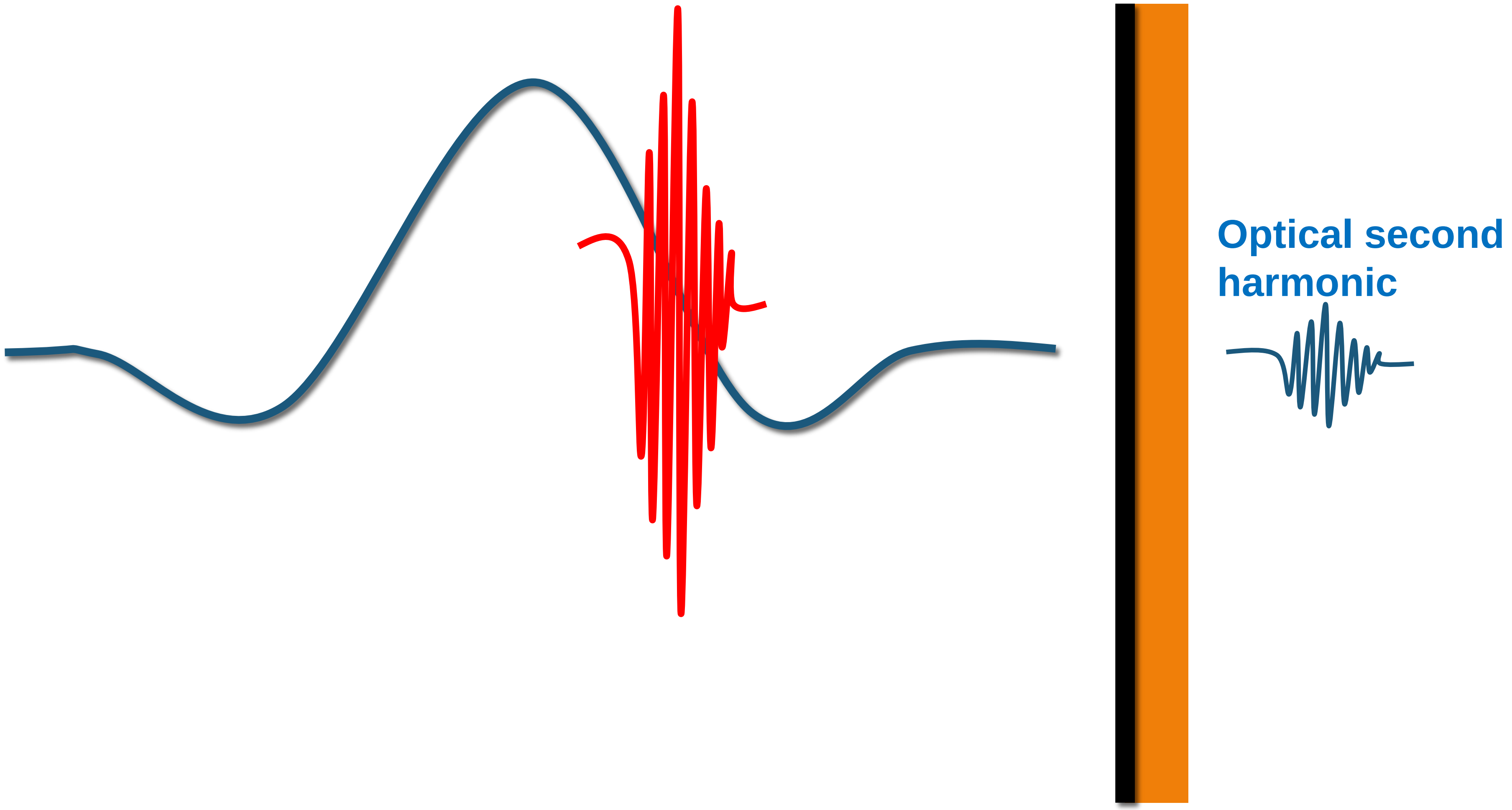
$$N_{\mathrm{detected}} \propto \exp\left(E_{THz}^{\ 3/4} \right)$$

arXiv:1704.01551 [cond-mat.mes-hall]

I.V. Oladyshkin, S.B. Bodrov, Yu.A. Sergeev, A.I.Korytin, M.D Tokman, A.N. Stepanov Optical emission of graphene and electron-hole pair production induced by a strong THz field

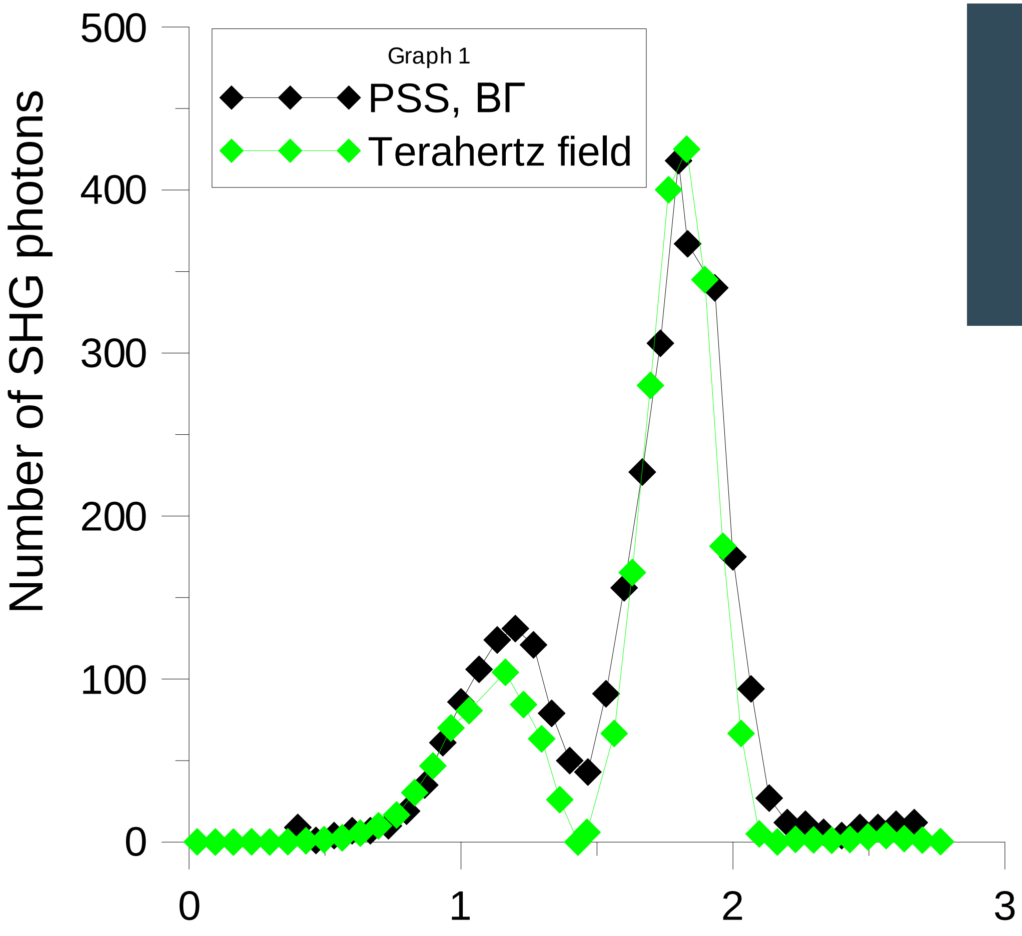
Second harmonic generation

SHG from graphene in the presence of THz pulse



Second harmonic generation

SHG from graphene in the presence of THz pulse

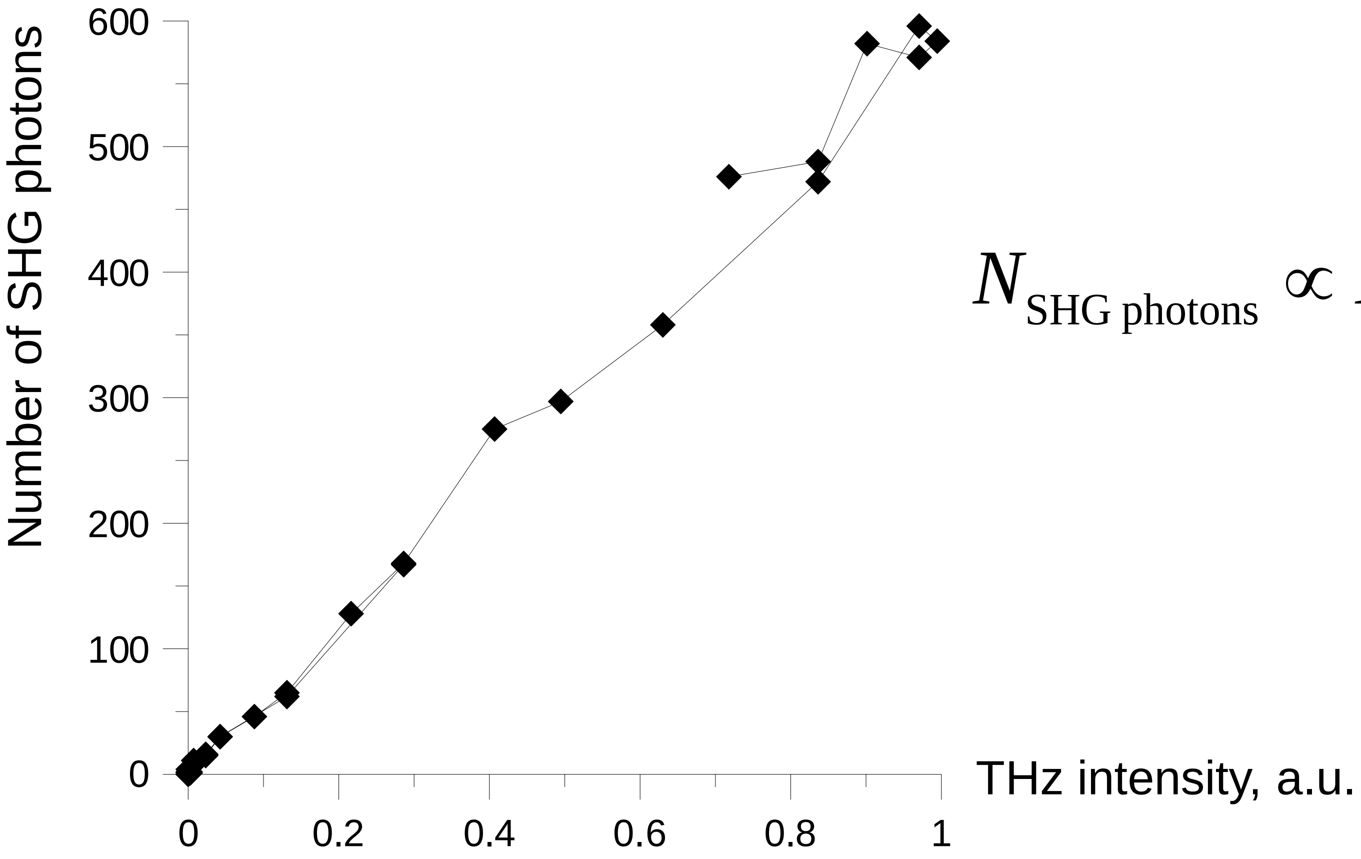


SHG depends on THz field almost locally

Time delay between laser and THz pulses, ps

Second harmonic generation

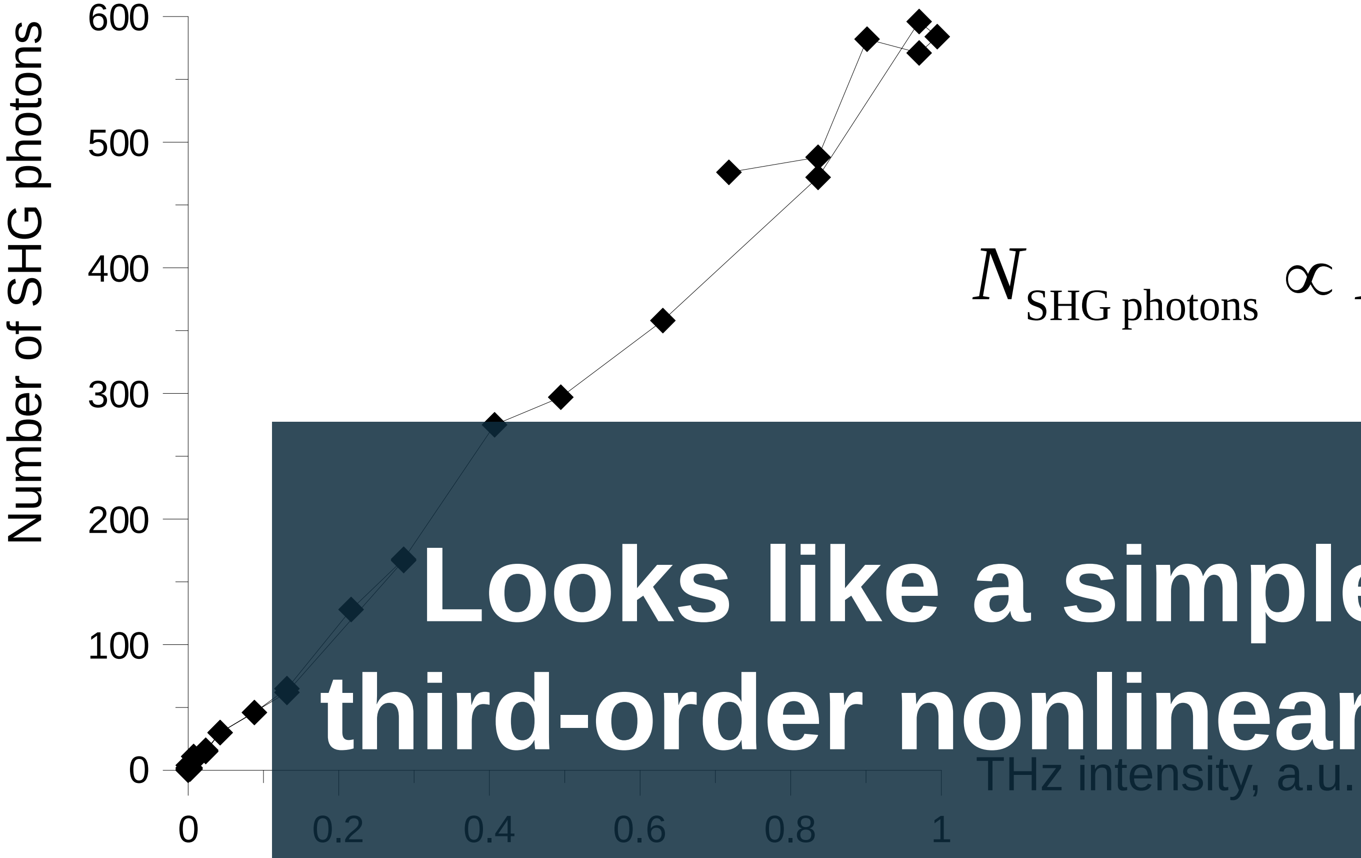
SHG from graphene in the presence of THz pulse



$$N_{\text{SHG photons}} \propto I_{THz} \times I_{opt}^{\;2}$$

Second harmonic generation

SHG from graphene in the presence of THz pulse

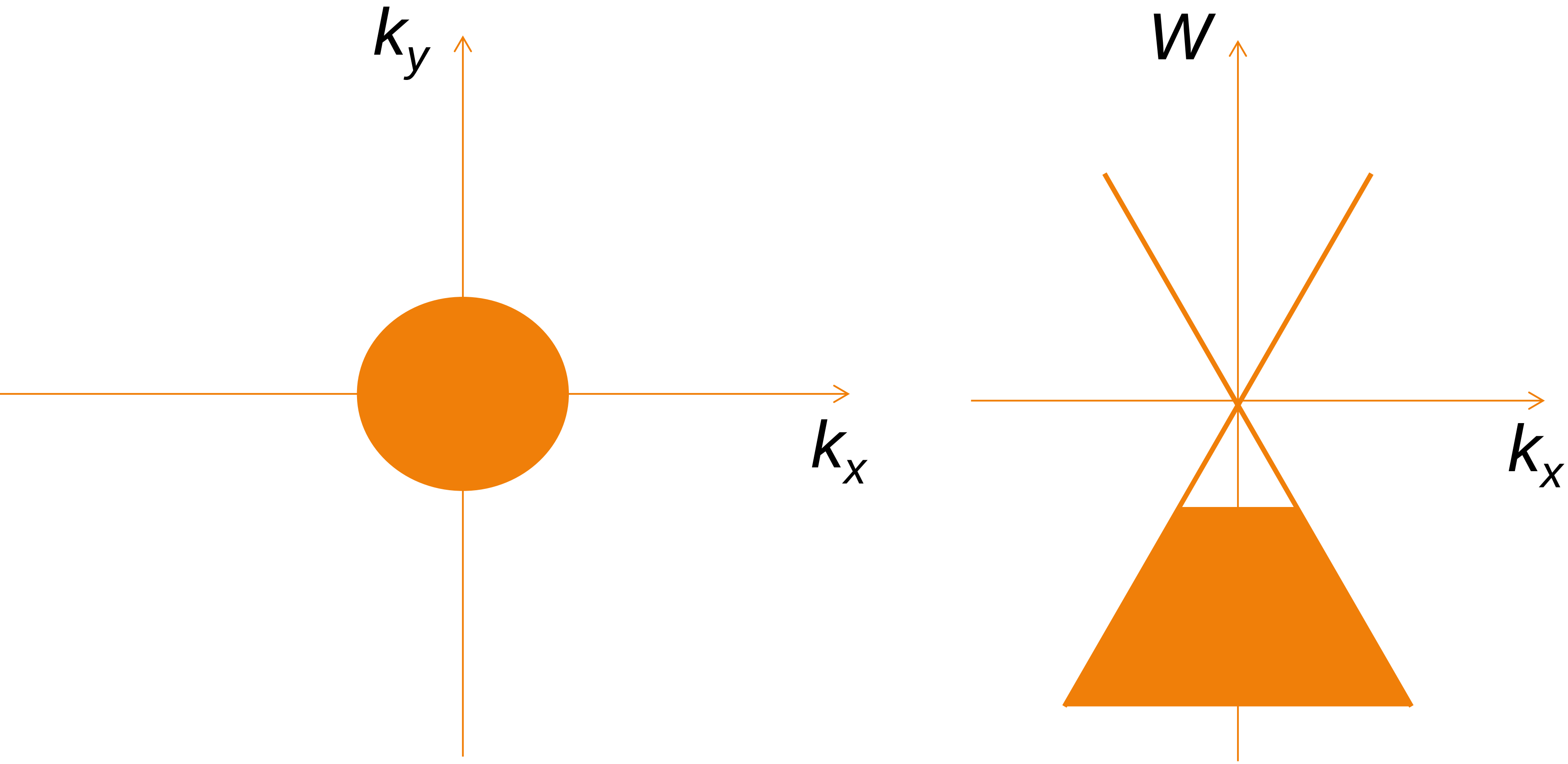


$$N_{\text{SHG photons}} \propto I_{THz} \times I_{opt}{}^{2}$$

Looks like a simple third-order nonlinearity

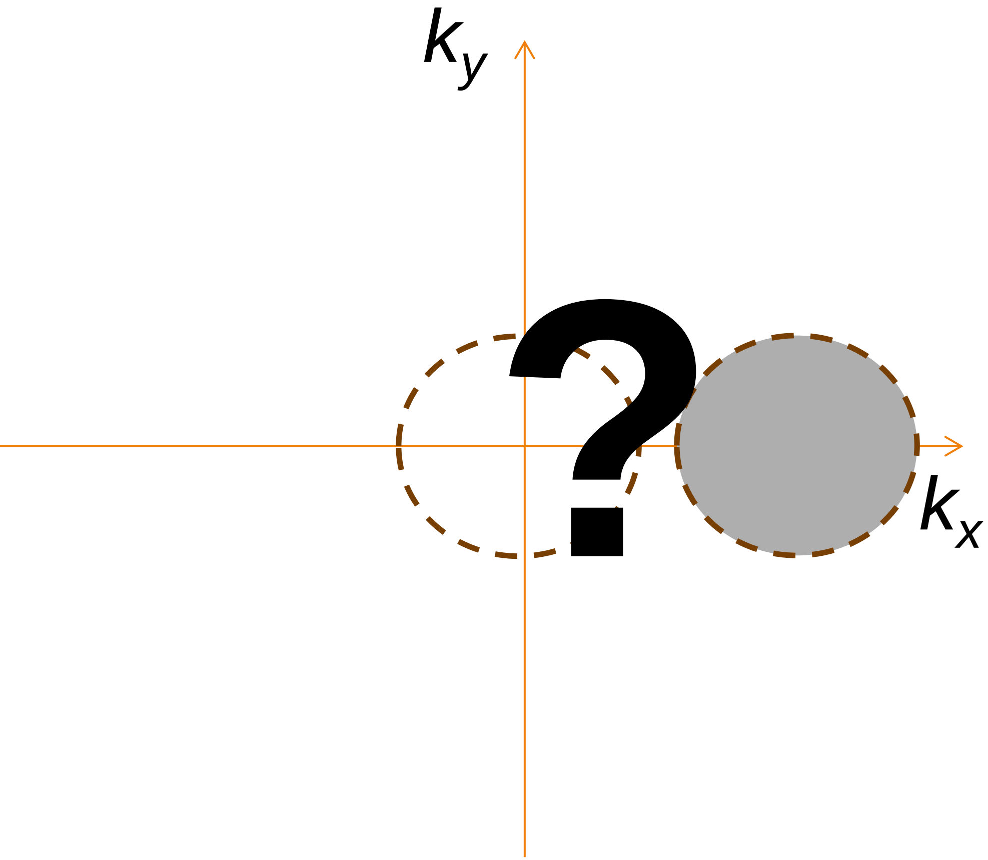
Second harmonic generation

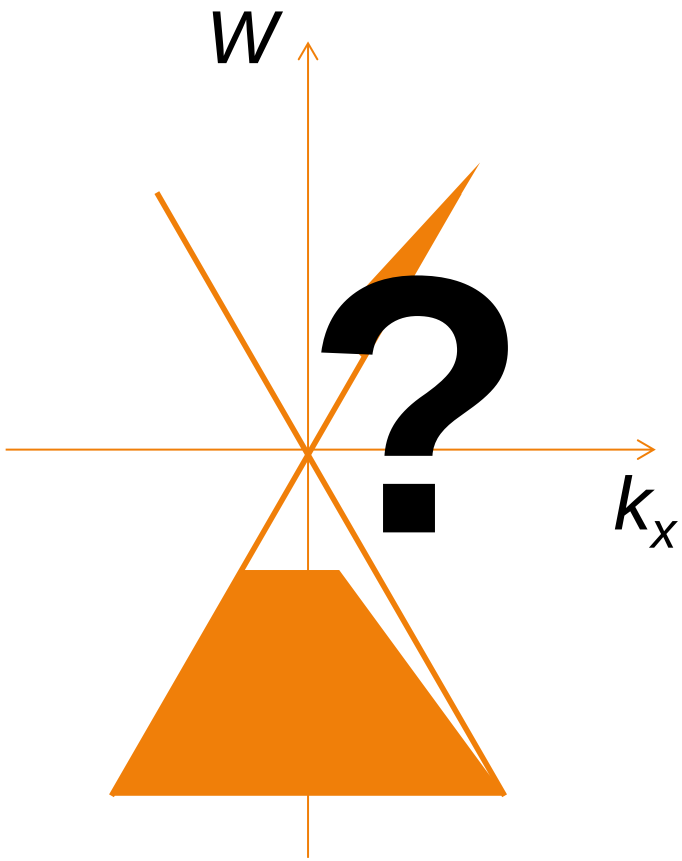
No field



Second harmonic generation

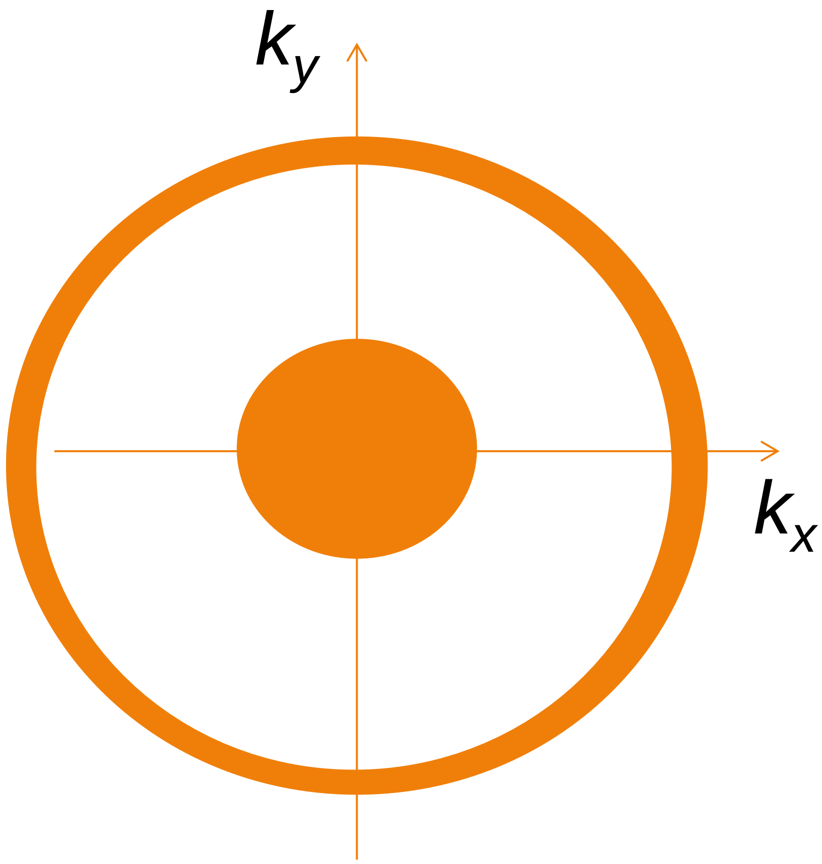
THz field is very strong
The perturbation theory is inapplicable

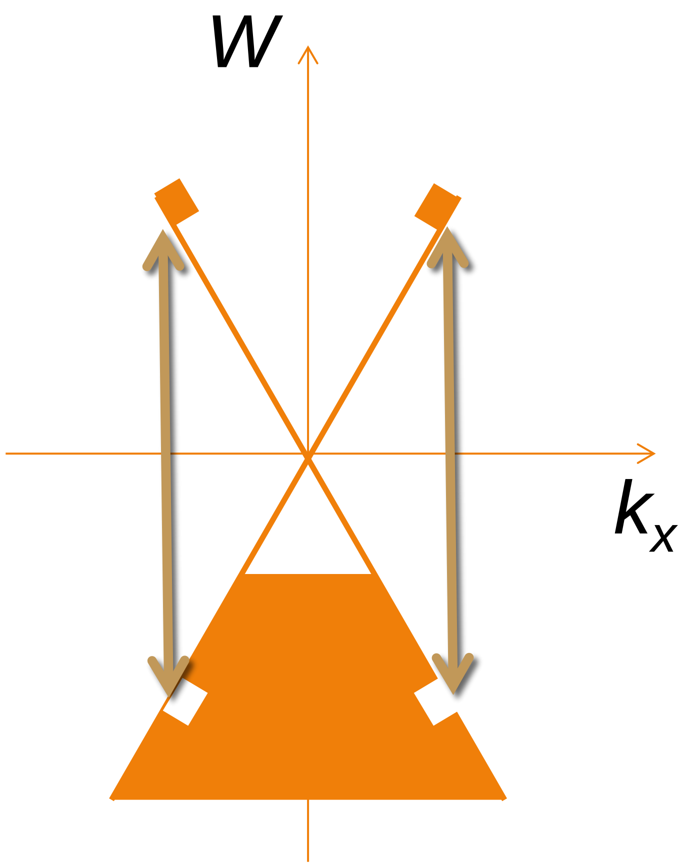




Second harmonic generation

Only optical field

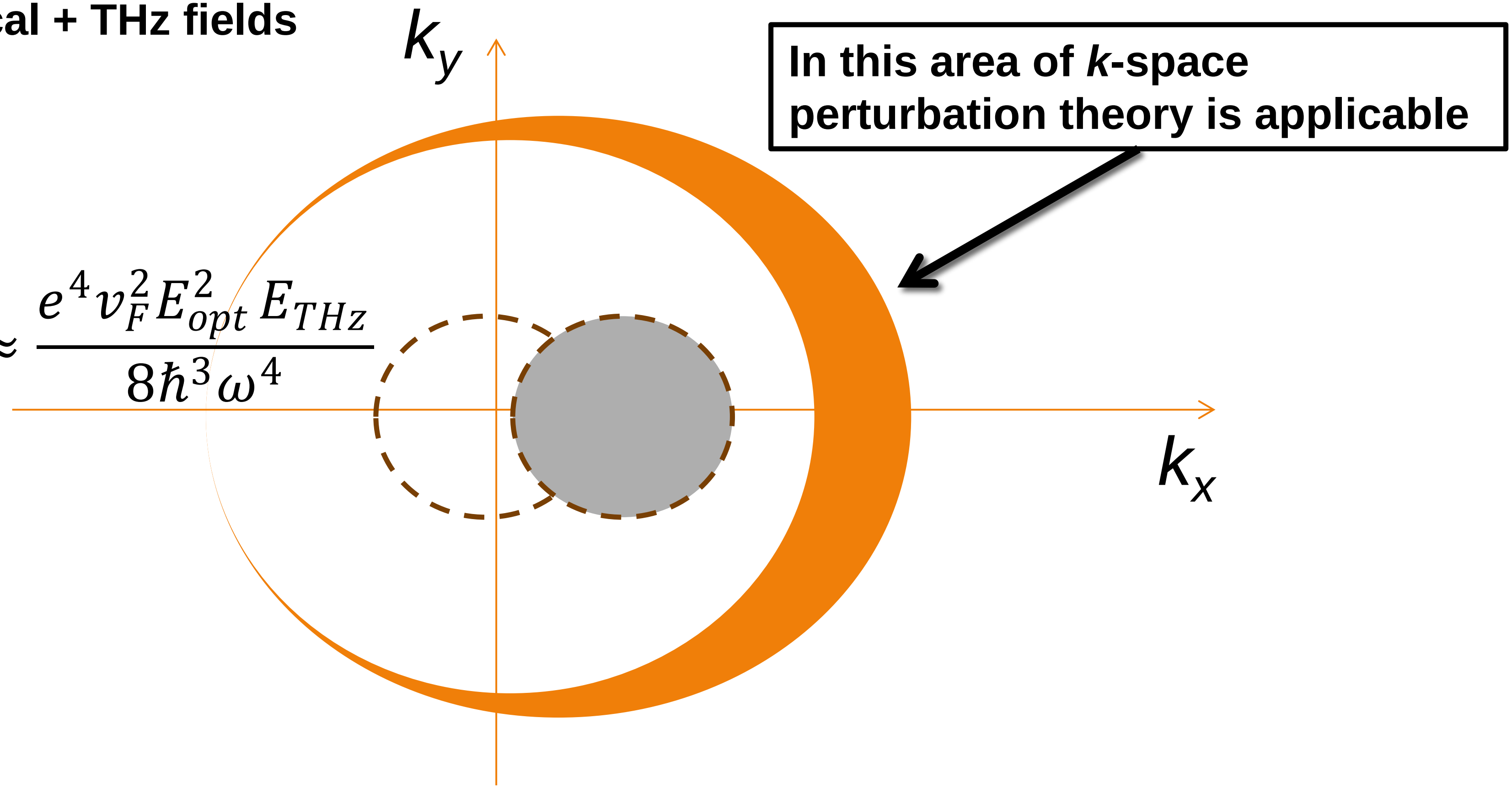




Second harmonic generation

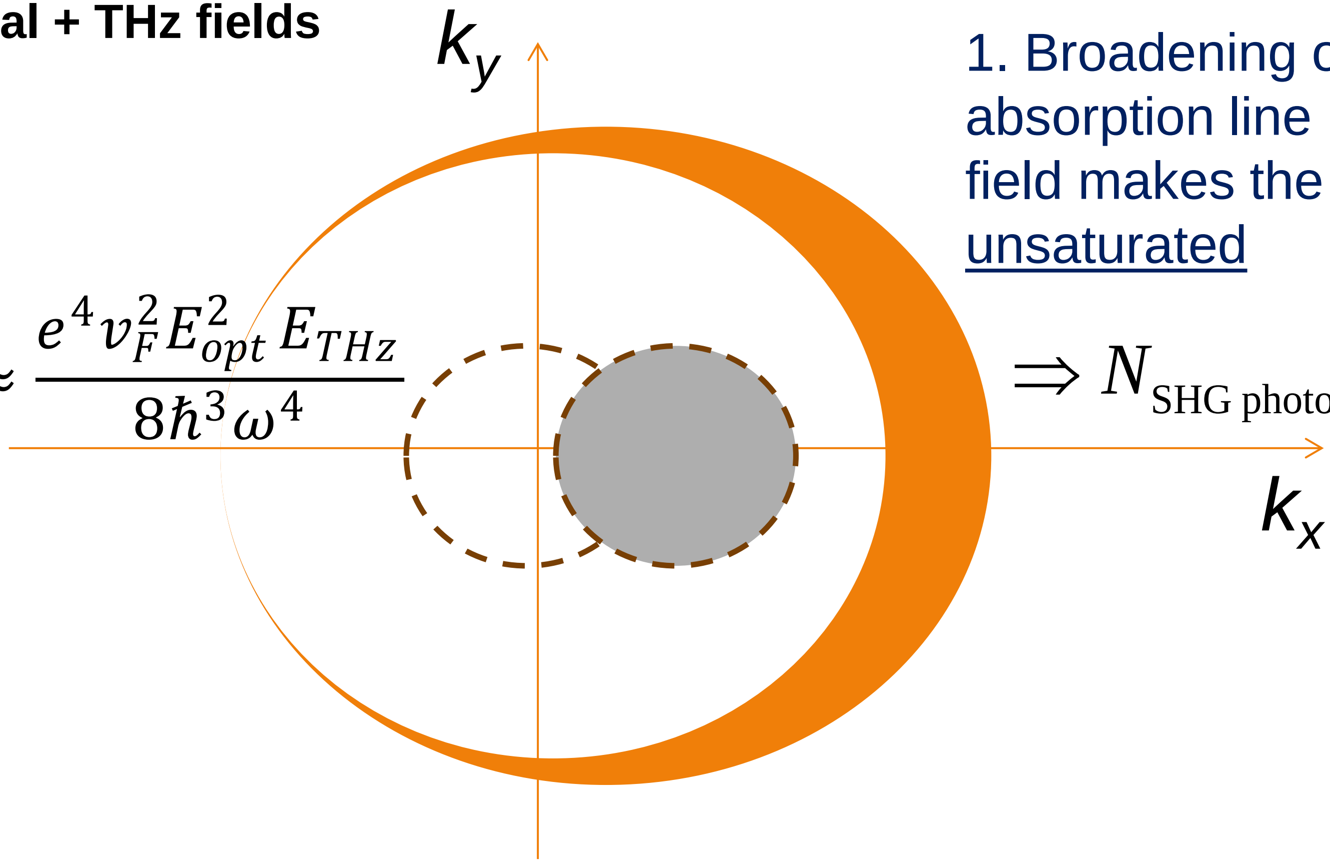
Optical + THz fields

$$j_{x2\omega} \approx \frac{e^4 v_F^2 E_{opt}^2 E_{THz}}{8\hbar^3 \omega^4}$$



Second harmonic generation

Optical + THz fields



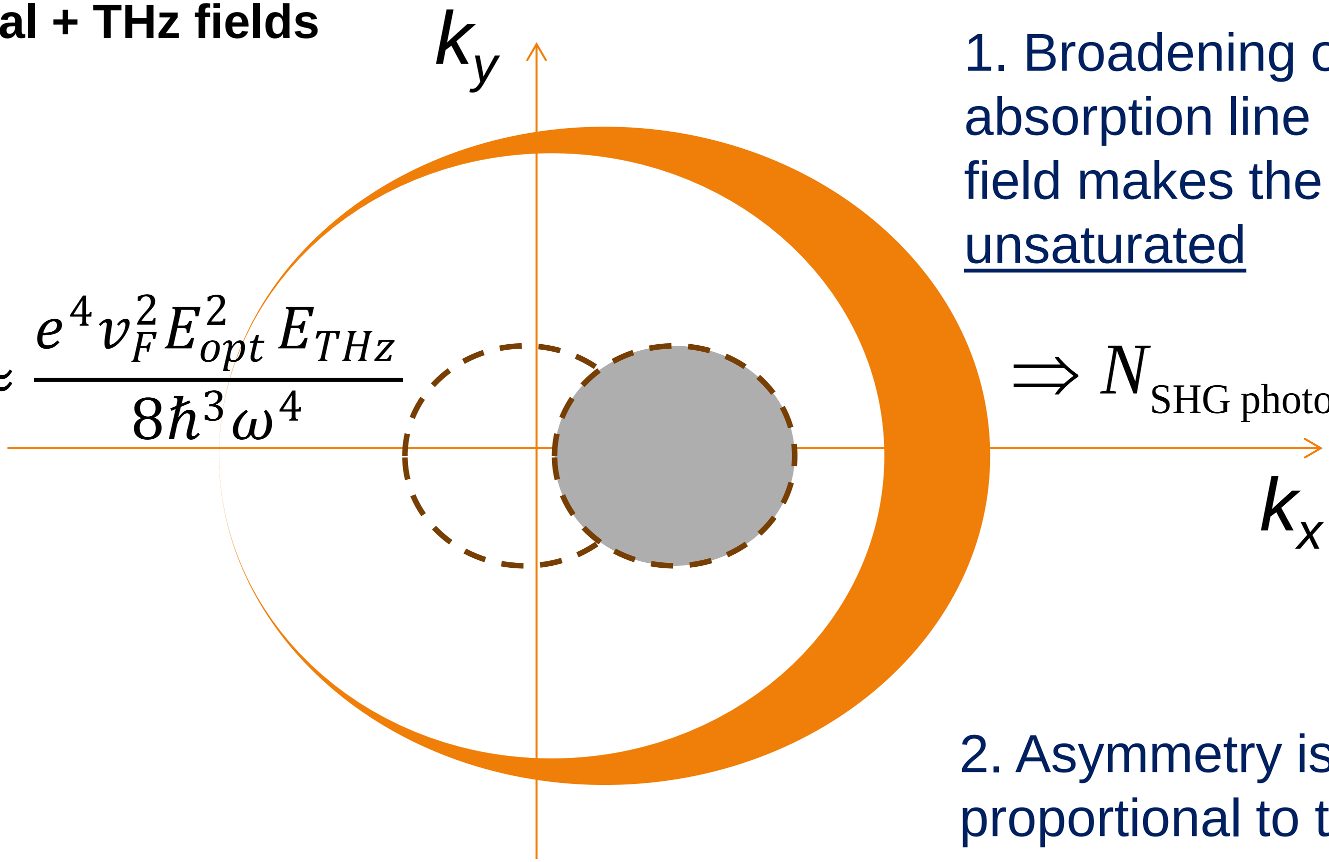
$$j_{x2\omega} \approx \frac{e^4 v_F^2 E_{opt}^2 E_{THz}}{8\hbar^3\omega^4}$$

1. Broadening of the absorption line by THz field makes the process unsaturated

$$\Rightarrow N_{\text{SHG photons}} \propto I_{opt}^{\;2}$$

Second harmonic generation

Optical + THz fields



$$j_{x2\omega} \approx \frac{e^4 v_F^2 E_{opt}^2 E_{THz}}{8\hbar^3 \omega^4}$$

1. Broadening of the absorption line by THz field makes the process <u>unsaturated</u>

$$\Rightarrow N_{\text{SHG photons}} \propto I_{opt}{}^2$$

2. Asymmetry is proportional to the THz field

$$\Rightarrow N_{\text{SHG photons}} \propto I_{THz}$$

Conclusion

- Spontaneous emission of graphene in strong THz field was observed
- The Landau-Zener transitions are the may mechanism of electron-hole pair production

Conclusion

- Spontaneous emission of graphene in strong THz field was observed
- The Landau-Zener transitions are the may mechanism of electron-hole pair production

- SHG from graphene in strong THz field was measured
- Broadening of the absorption line by THz field is the principle effect
- The experimental data can be interpreted taking into account electron distribution asymmetry in the resonant region

Plans / questions

- Is it possible to observe the Dicke effect in graphene?
    - we need less phonons

- What is different if there is a strong magnetic field?

- Can we extract any data on graphene from these measurements?

Density matrix equations

$$\hat{H} = \upsilon_F \hat{\boldsymbol{\sigma}}\left(\hat{\boldsymbol{p}} + e\boldsymbol{A}/c\right) - e\phi$$

$$\left(\frac{\partial}{\partial t} - \frac{e}{\hbar}E\left(t\right)\cdot\frac{\partial}{\partial k_x}\right)\cdot\rho_{\boldsymbol{k}} + i\omega_k\rho_{\boldsymbol{k}} = -i\frac{\Omega_{\boldsymbol{k}}\left(t\right)}{2}\Delta_{\boldsymbol{k}} \qquad \Omega_{\boldsymbol{k}}\left(t\right) = \frac{\sin\theta_{\boldsymbol{k}} eE\left(t\right)}{k\hbar}$$

$$\left(\frac{\partial}{\partial t} - \frac{e}{\hbar}E\left(t\right)\cdot\frac{\partial}{\partial k_x}\right)\cdot\Delta_{\boldsymbol{k}} = -i\Omega_{\boldsymbol{k}}\left(t\right)\cdot\left(\rho_{\boldsymbol{k}} - \rho_{\boldsymbol{k}}^*\right)$$

$$\left(\frac{\partial}{\partial t} - \frac{e}{\hbar}E\left(t\right)\cdot\frac{\partial}{\partial k_x}\right)\cdot n_{\Sigma\boldsymbol{k}} = 0$$

Density matrix equations: SHG

$$\rho_{\omega\boldsymbol{k}}(t) \approx -\frac{i}{4}\mathrm{e}^{i\frac{(\omega-\omega_k)^2}{4\cos\theta_{\boldsymbol{k}}\frac{ev_F E_0}{\hbar}} - \gamma\frac{\omega-\omega_k}{2\cos\theta_{\boldsymbol{k}}\frac{ev_F E_0}{\hbar}}}\Omega_{\omega\boldsymbol{k}}(t_s,t)\Delta_{0\boldsymbol{k}}(t_s,t) \times$$

$$\times\left(\mathrm{e}^{\mp i\pi/4}\sqrt{\frac{\pi}{\left|\cos\theta_{\boldsymbol{k}}\frac{ev_F E_0}{\hbar}\right|}} + \int_0^{\left(\frac{\omega-\omega_k}{2\cos\theta_{\boldsymbol{k}}\frac{v_F e E_0}{\hbar}}\right)} \mathrm{e}^{-i\cos\theta_{\boldsymbol{k}}\frac{ev_F E_0}{\hbar}x^2}dx\right)$$