TeraHP

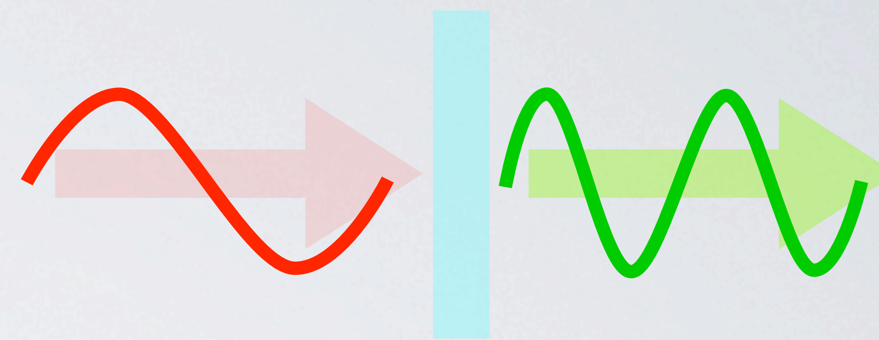
Here !

Quest for Extreme Terahertz Nonlinear Optics : on Dirac Fermions in Graphene

Shuntaro Tani, Kyoto University

Outline

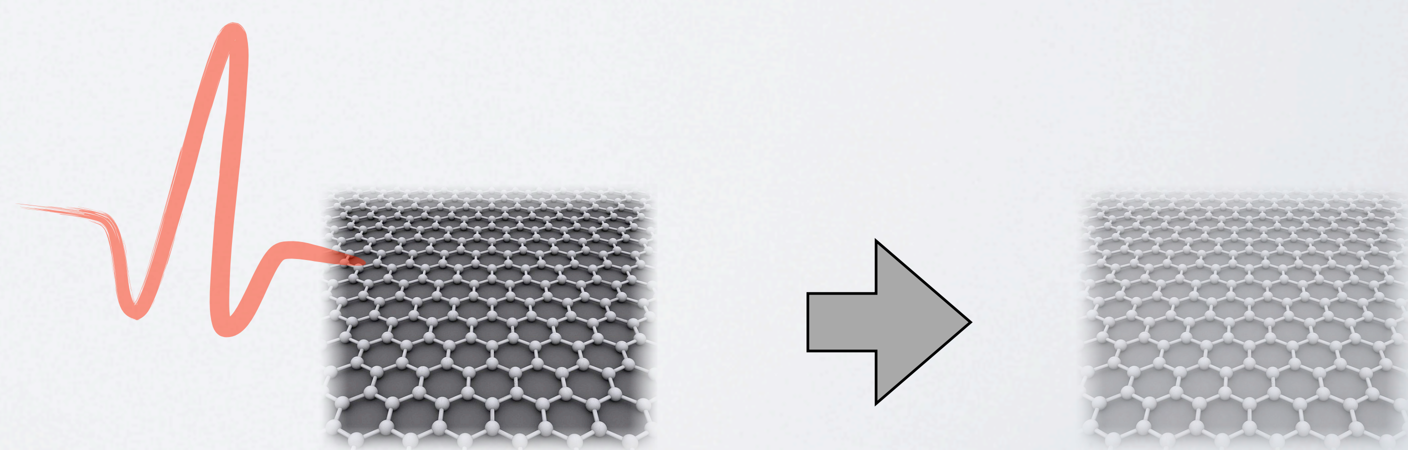
- Nonlinear Optics and Extreme Nonlinear Optics



- Terahertz Nonlinear Optics

$$U_p = \frac{e^2 E^2}{4m\omega^2}$$

- Terahertz Nonlinear Response in Graphene



What is Nonlinear Optics ?

Maxwell equations in medium

$$\nabla \cdot E = -\frac{1}{\epsilon_0}\nabla \cdot P \qquad \nabla \times E = -\frac{\partial B}{\partial t}$$

$$\nabla \cdot B = 0 \qquad c^2\nabla \times B = \frac{\partial E}{\partial t} + \frac{1}{\epsilon_0}\frac{\partial P}{\partial t}$$

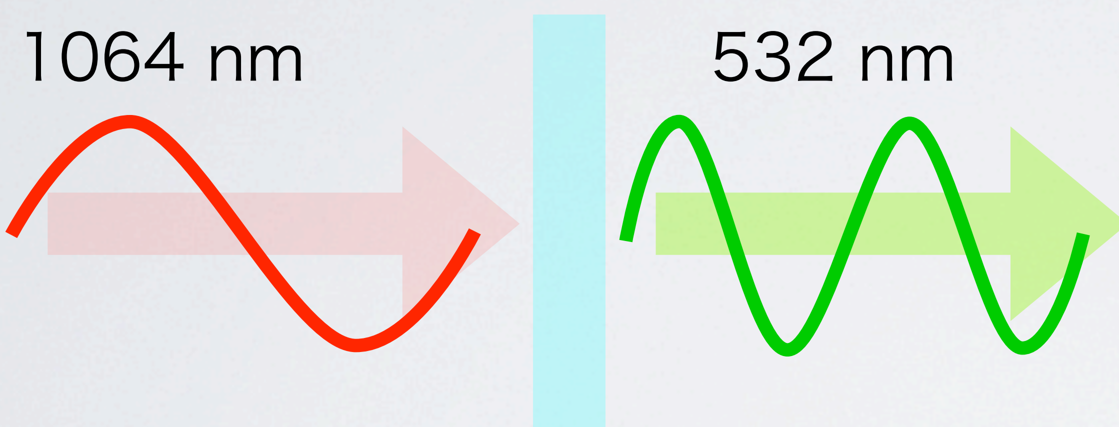
Polarization $P \propto \chi E$ ($E <<$ interatomic electric fields)

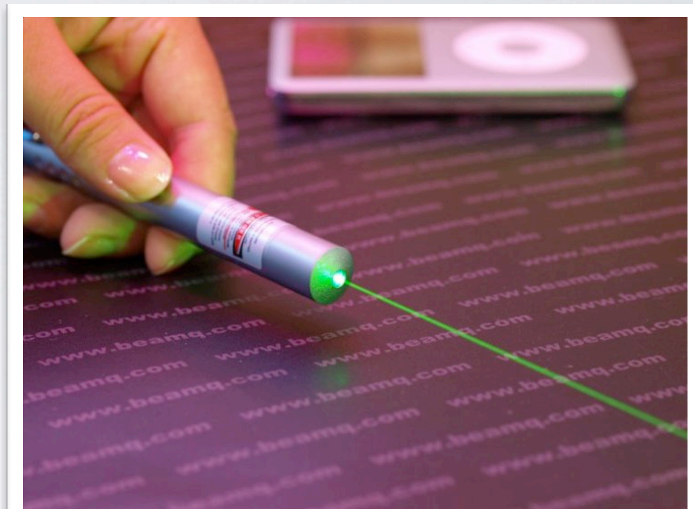
$$P \propto \chi^{(1)}E + \chi^{(2)}EE + \chi^{(3)}EEE + \cdots$$

What is Nonlinear Optics ?

$$P \propto \chi^{(1)} E + \underline{\chi^{(2)} EE} + \chi^{(3)} EEE + \cdots$$

$$E \propto e^{-i\omega t} \quad \rightarrow \quad P \propto e^{-i\,\underline{2\omega} t} \quad \rightarrow \quad E \propto e^{-i\,\underline{2\omega} t}$$





http://www.kalaser.com/

What happens when the “energy” of light becomes comparable or larger than a characteristic energy of the system.

Characteristic Energies

The energy associated to the light intensity

Ponderomotive energy	$U_p \propto I$
Rabi energy	$\hbar\Omega_R \propto \sqrt{I}$
Bloch energy	$\hbar\Omega_B \propto \sqrt{I}$
Cyclotron energy	$\hbar\omega_c \propto \sqrt{I}$
Tunneling energy	$\hbar\Omega_{tun} \propto \sqrt{I}$

The characteristic energy of the system

Carrier photon energy	$\hbar\omega_0$
Binding energy	E_b
Rest Energy	m_0c^2

Characteristic Energies

The energy associated to the light intensity

Ponderomotive energy $U_p \propto I$

- An average kinetic energy of a electron in the oscillating electric field

$$U_p = \frac{e^2 E^2}{4m\omega^2} \propto I$$

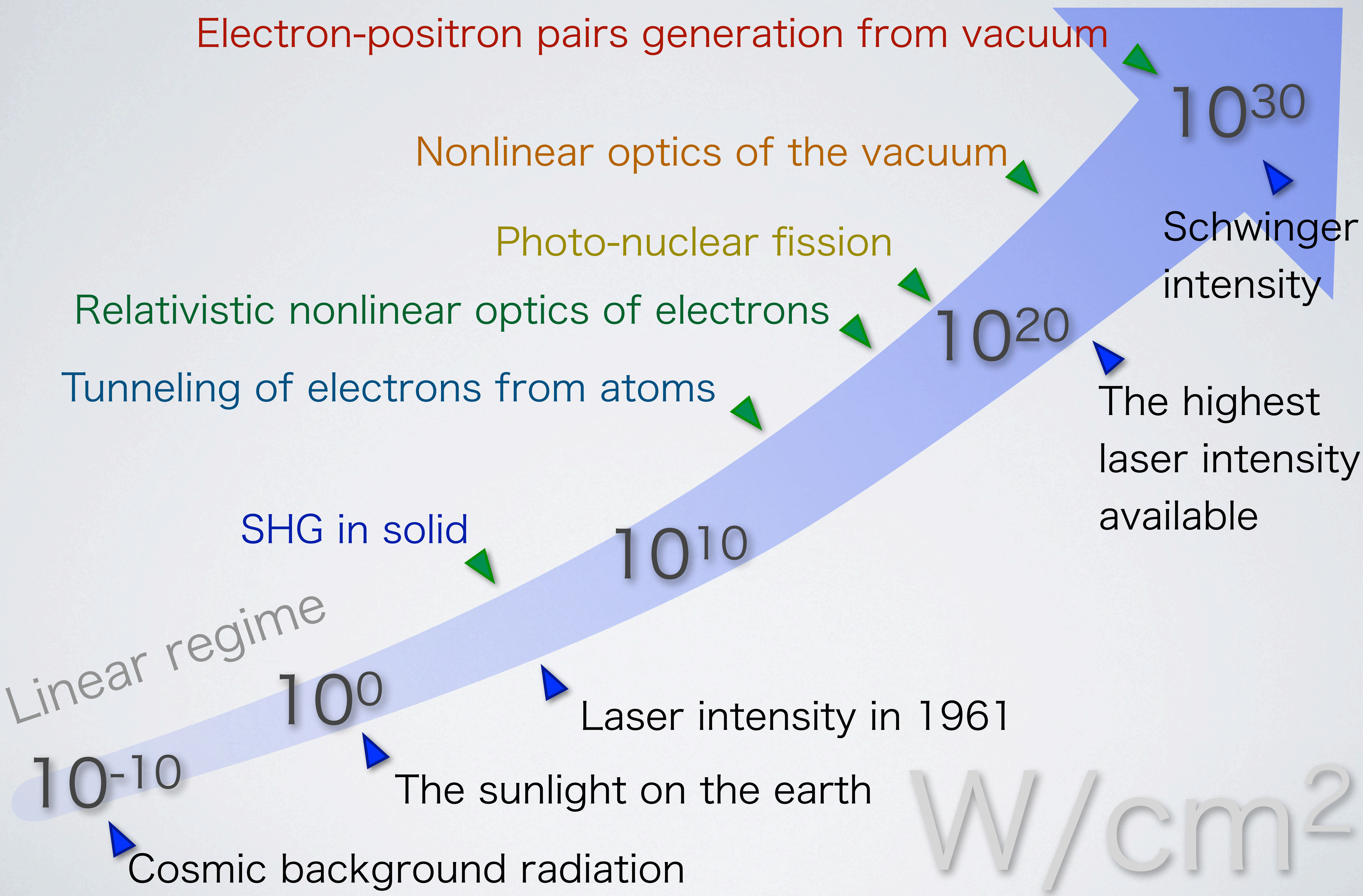
The characteristic energy of the system

Carrier photon energy $\hbar\omega_0$

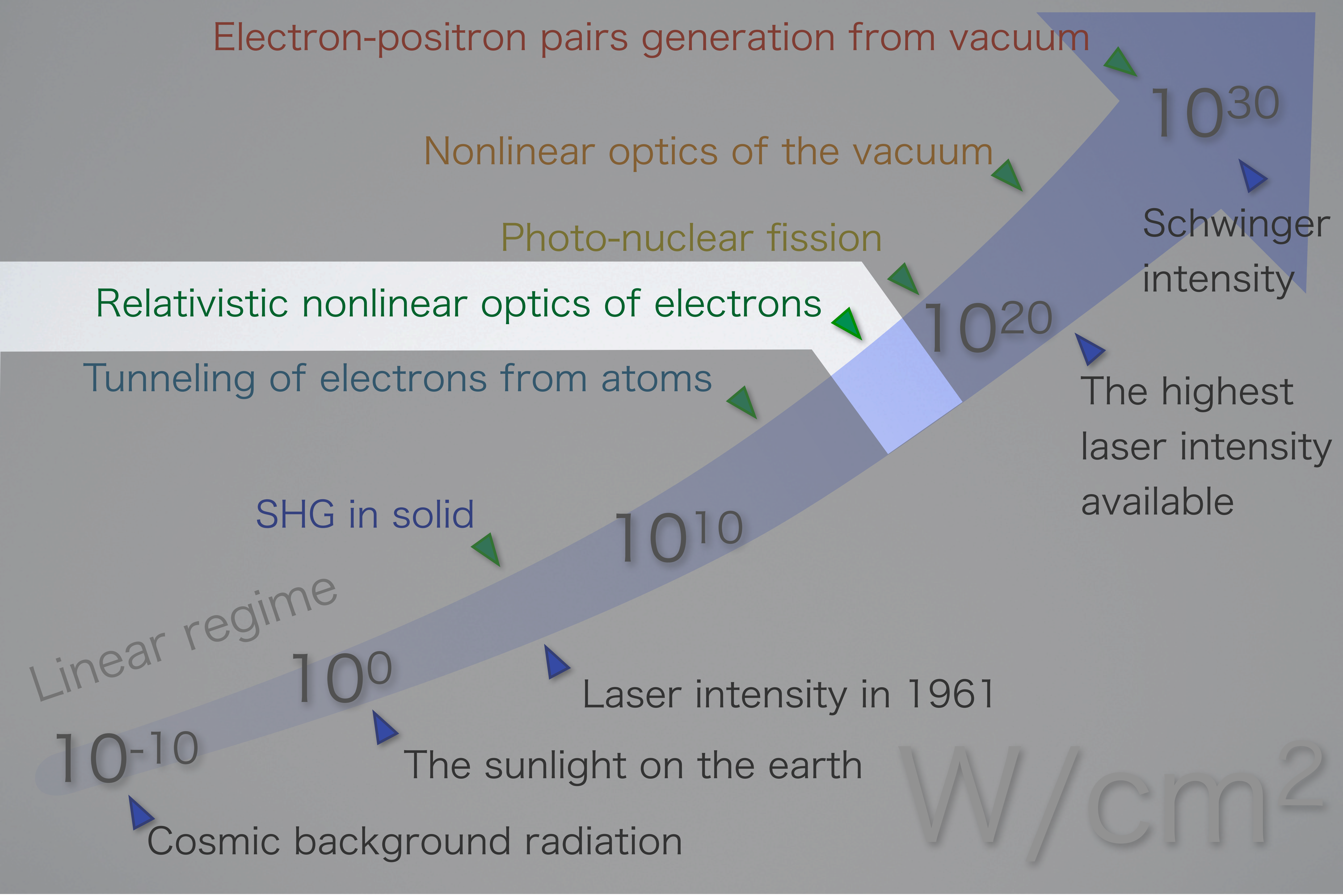
Binding energy E_b

Rest Energy m_0c^2

Nonlinear Optical Phenomena with Visible Laser



Nonlinear Optical Phenomena with Visible Laser



Links Among the Hierarchies

Vacuum → “Vacuum” in a semiconductor

Electron-positron pairs → Electron-hole pairs

Visible Laser → Terahertz-wave

Terahertz Wave

1 THz ~ 4.14 meV << visible light (~ 2 eV)

Elementary excitation in solid

- Exciton binding energy
- Lattice vibration energy
- Superconductor gap

etc...

Temporal waveform measurement is possible!

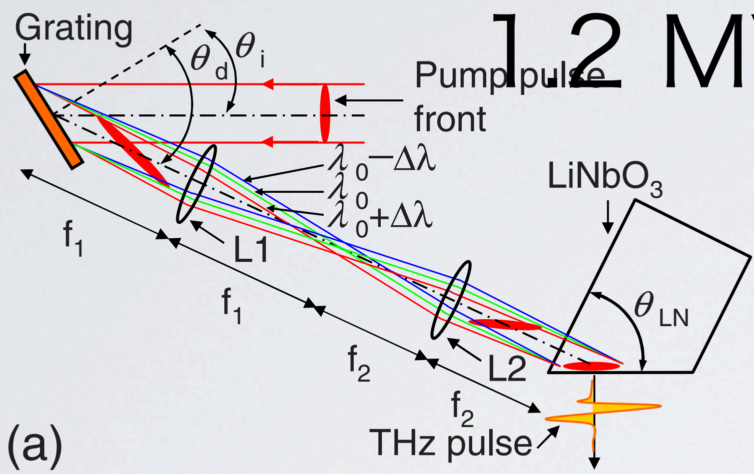
Coherent single cycle pulse is available!

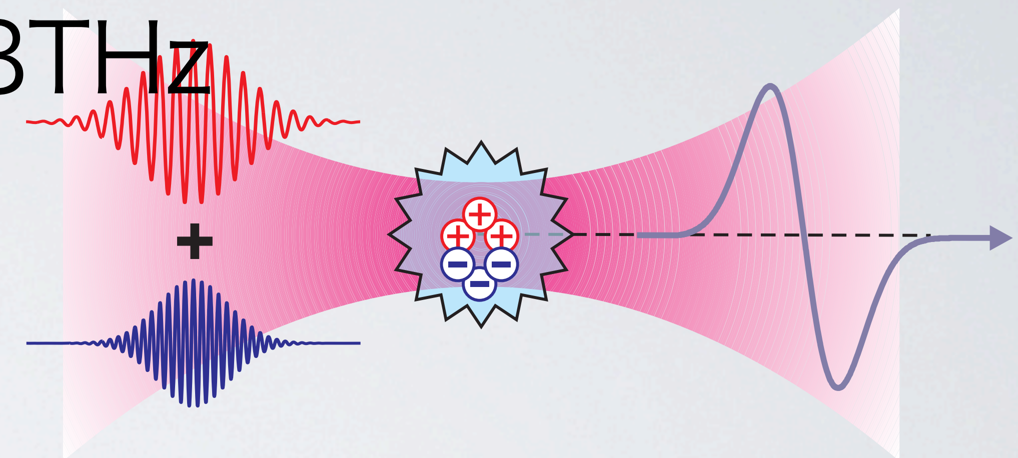
High Power Terahertz-wave Generation

Tilted wavefront method

Air plasma method

1.2 MV/cm @ 0.8THz





H.Hirori, K.Tanaka, et al., APL **98**, 091106 (2011)

M.D.Thomson, et al., Laser & Photon. Rev. **1**, 349 (2007)

Terahertz Nonlinear Phenomena

- Coherent control of intra-excitonic state

S.Leinß, et al., Phys. Rev. Lett. **101**, 246401 (2008)

- Light-induced superconductivity in cuprate

D.Fausti, et al., Science **331**, 189 (2011)

- Extraordinary carrier multiplication

H.Hirori, K.Tanaka, et al., Nature Comm. **2**, 594 (2011)

Links Among the Hierarchies

Ponderomotive energy

$$U_p = \frac{e^2 E^2}{4m\omega^2}$$

Visible $\rightarrow$ THz

10^6 times larger

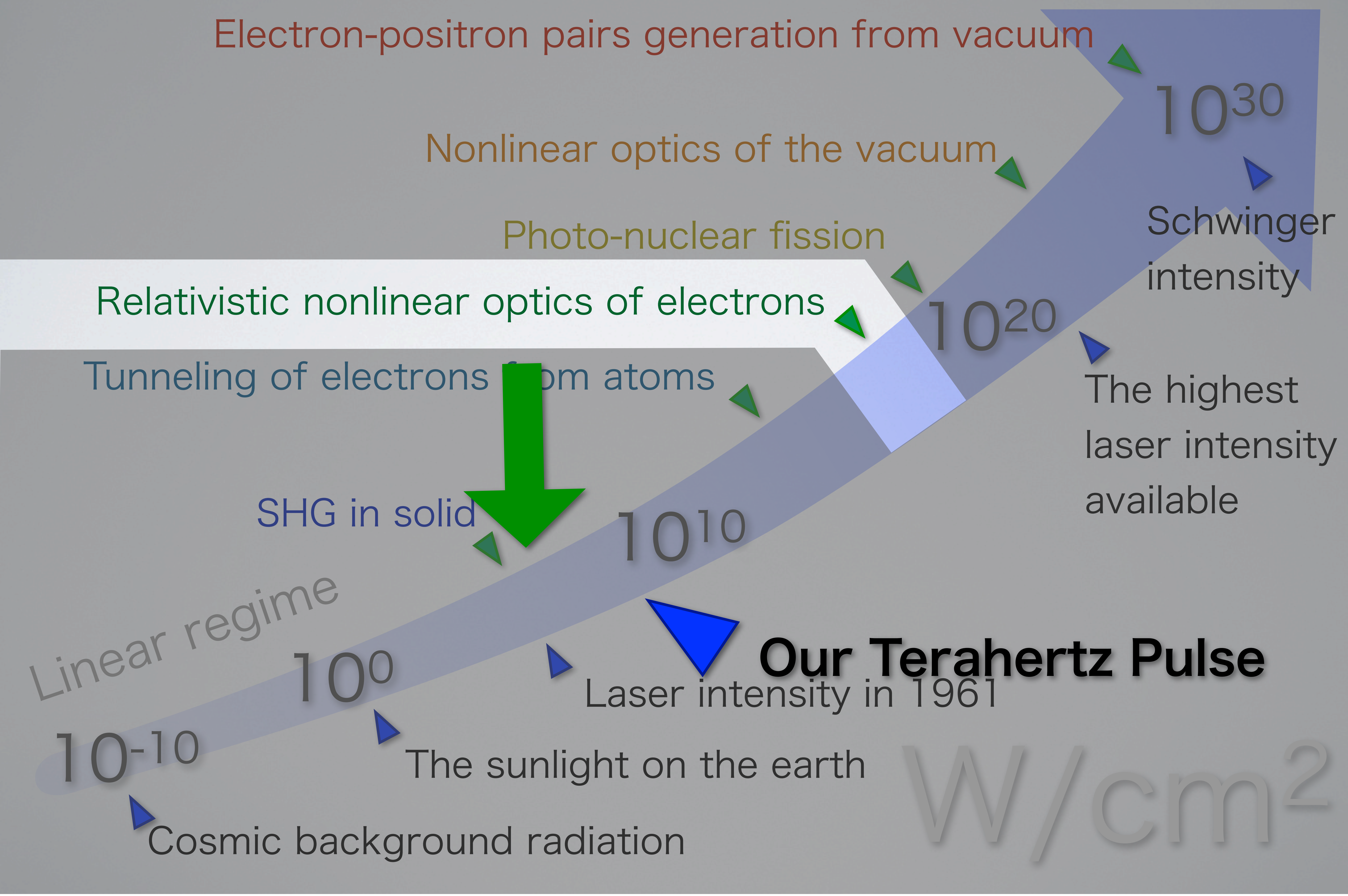
10 eV with 1 MV/cm
@ 1 THz

Carrier photon energy $\hbar\omega_0$ — 10^3 times smaller

Binding energy E_b — 10^3 times smaller

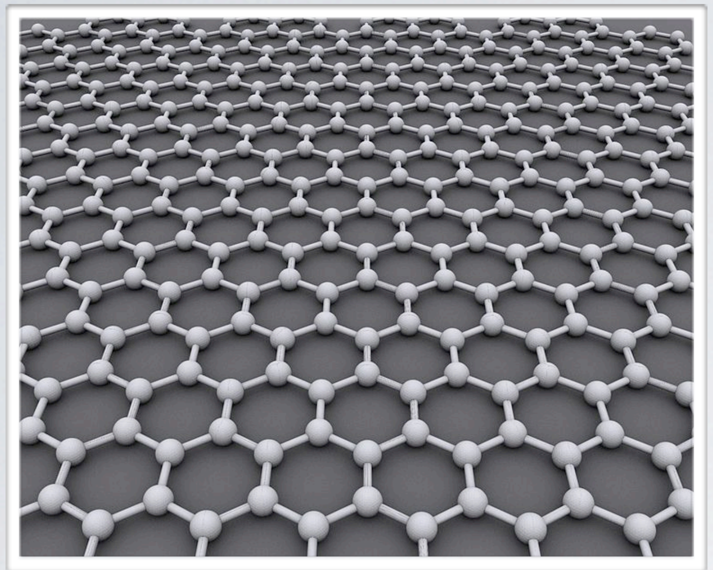
Rest Energy $m_0 c^2$ — 10^5 times smaller

Nonlinear Optical Phenomena with Visible Laser

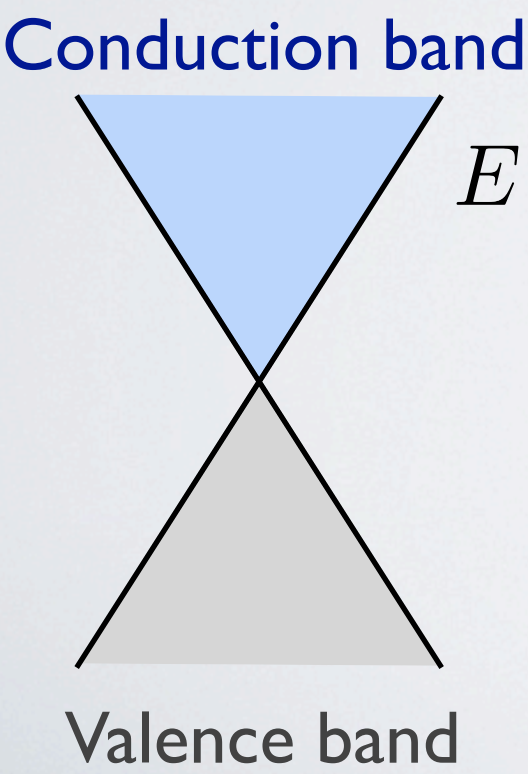


Relativistic Massless Electrons in Solid : Graphene

Graphene



Monoatomic layer carbon material

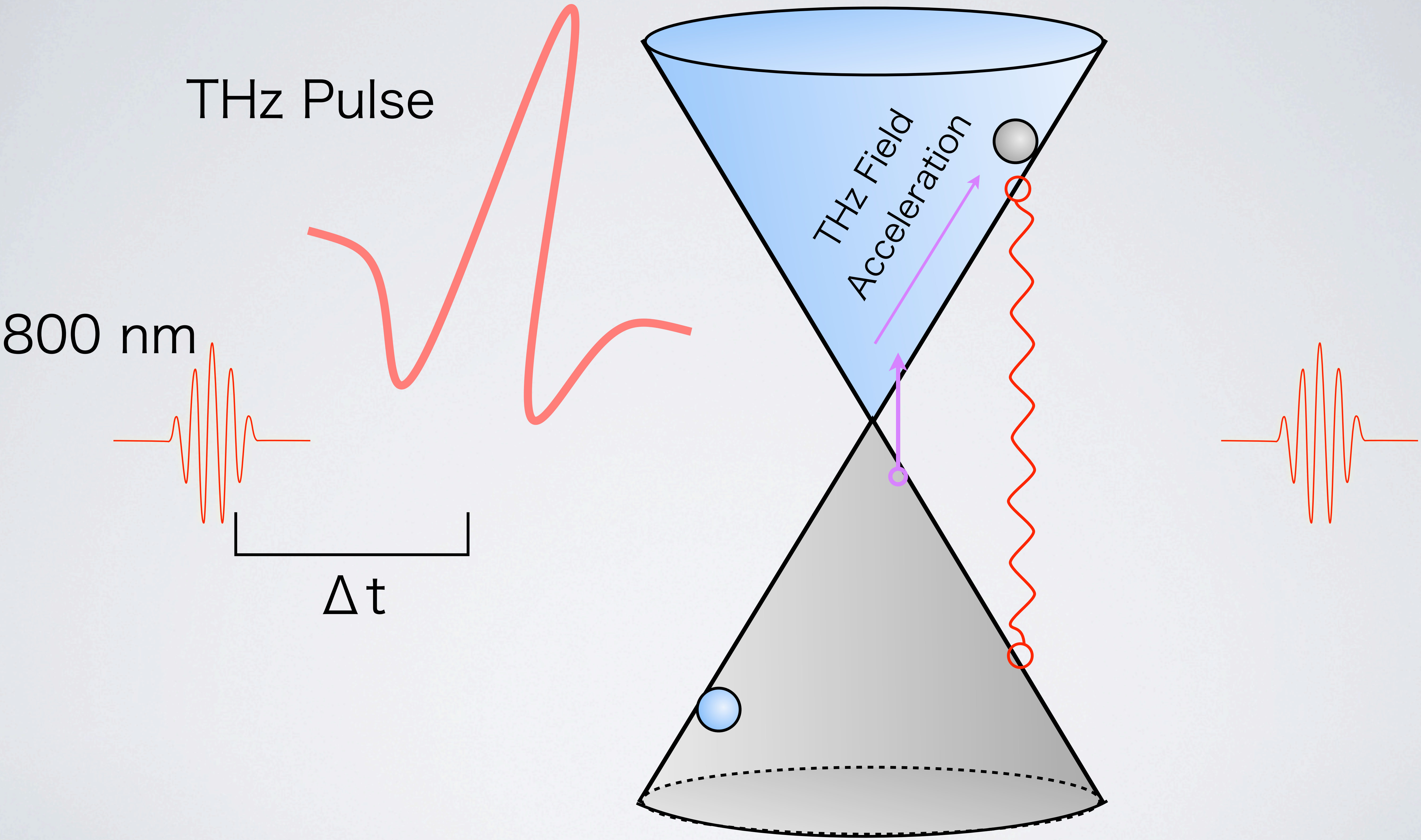


$$E = \pm \hbar v_F k$$

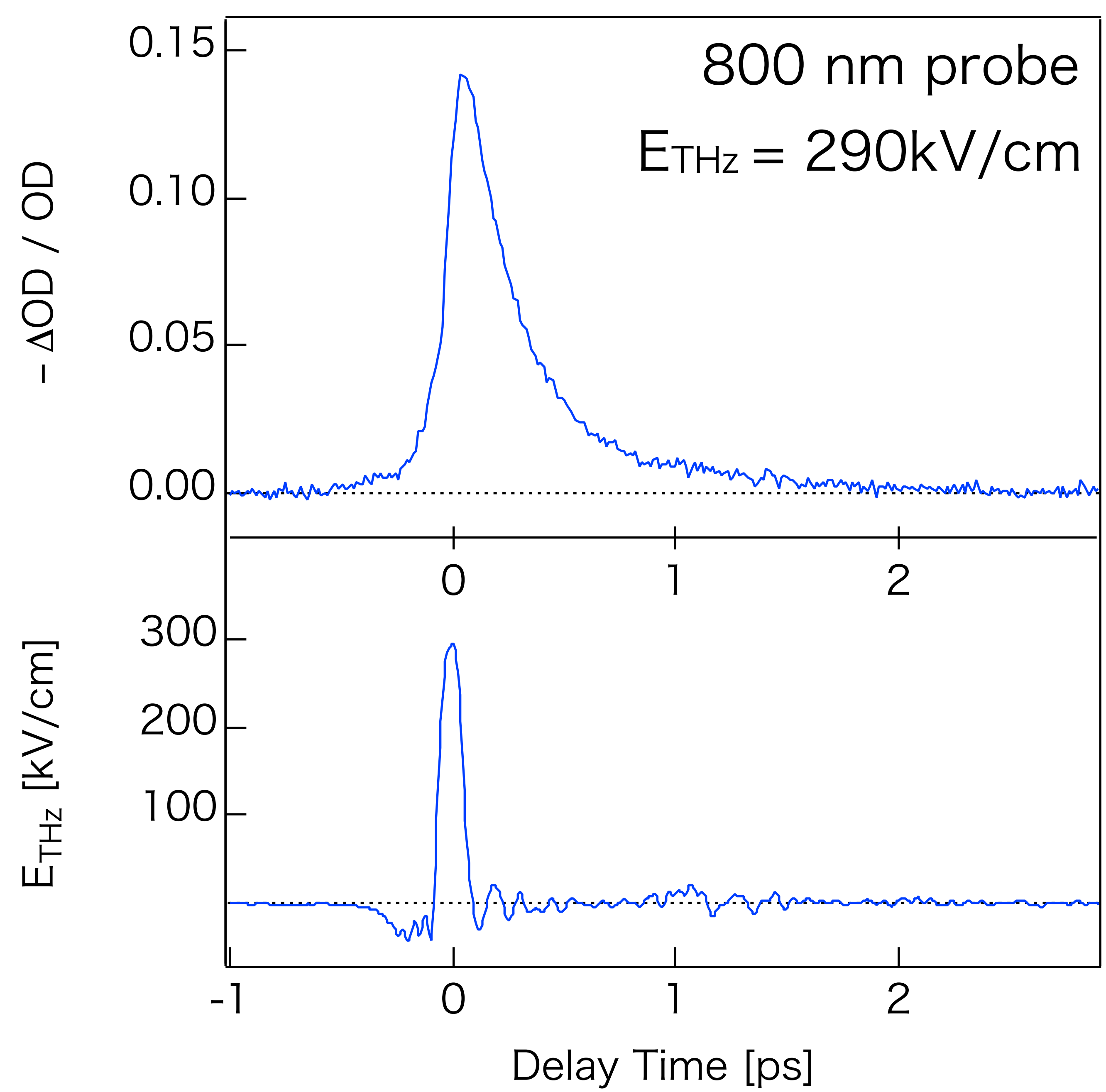
Dirac cone

- Linear dispersion
- Relativistic massless electrons

Experimental Scheme

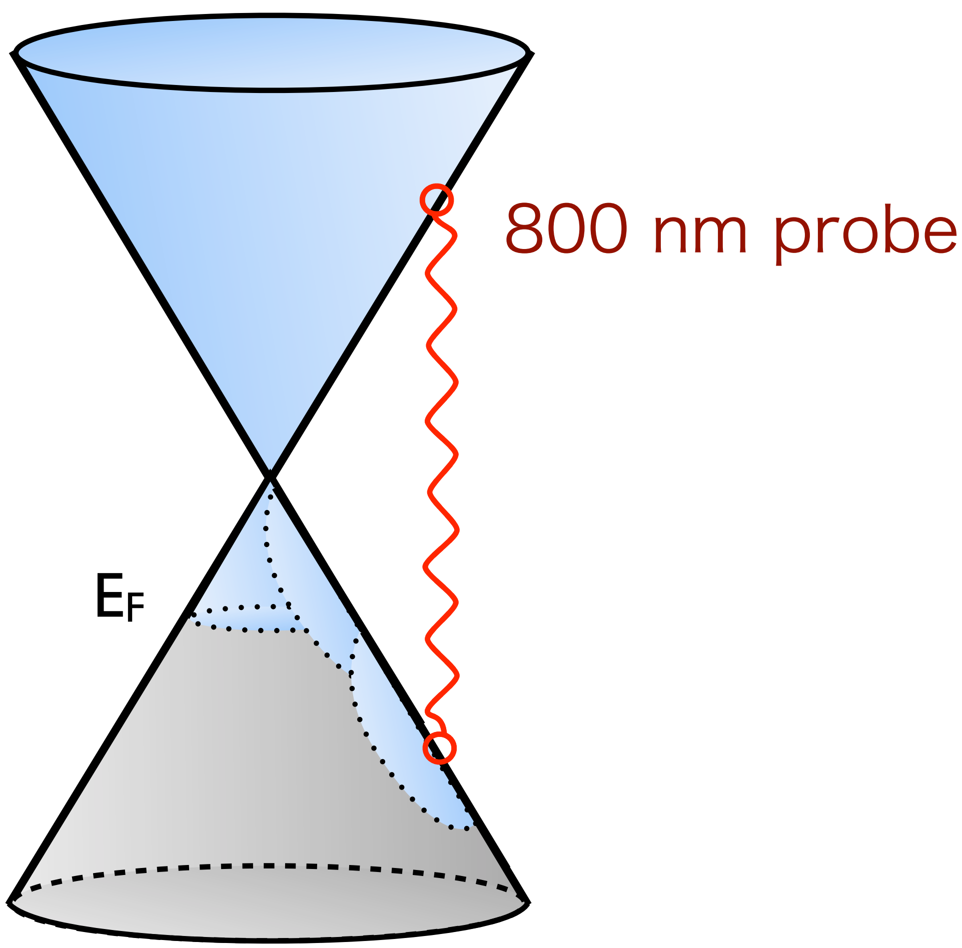


Results : Typical THz-induced Transparency in Near-infrared Region

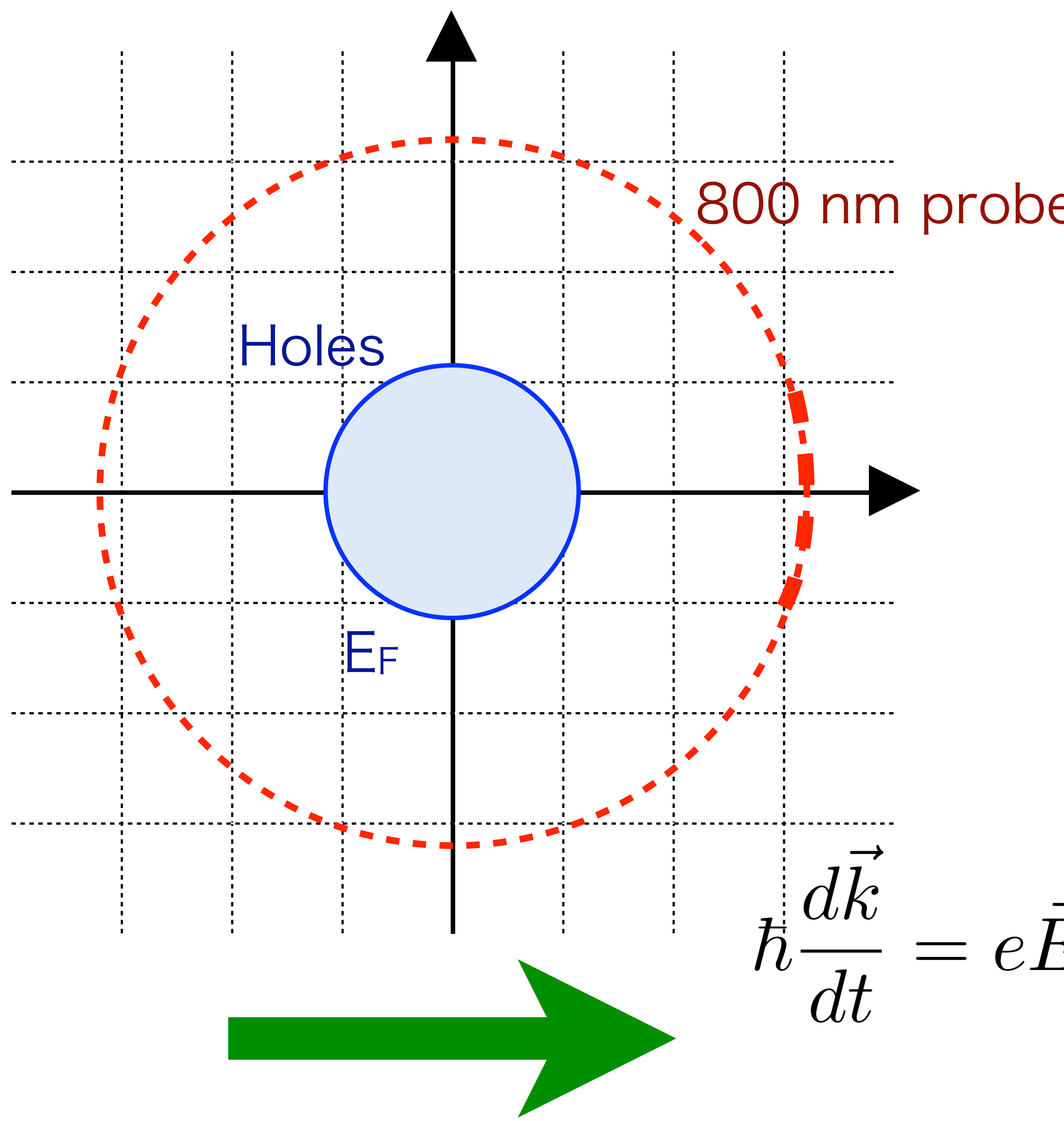


THz induced transparency over 14% at 800 nm

Origin of THz Induced Transparency

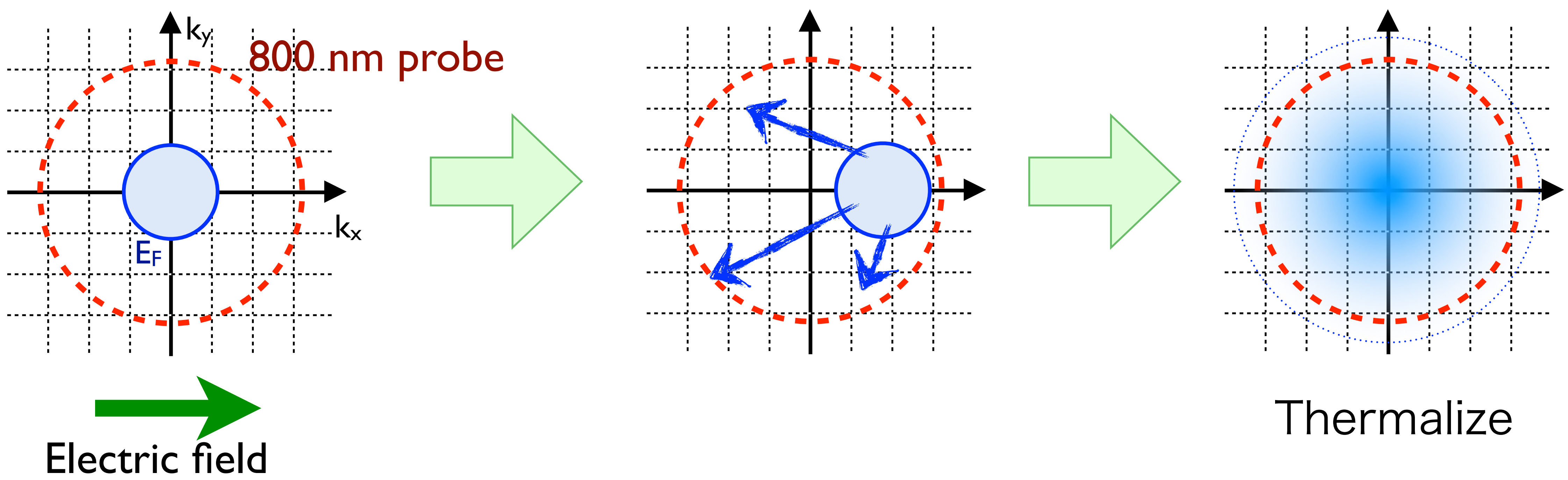


Hole distribution in the **momentum space**



$$\hbar \frac{d\vec{k}}{dt} = e\vec{E}$$

Carrier Scattering Process



Boltzmann equation for $f(\vec{k},t)$

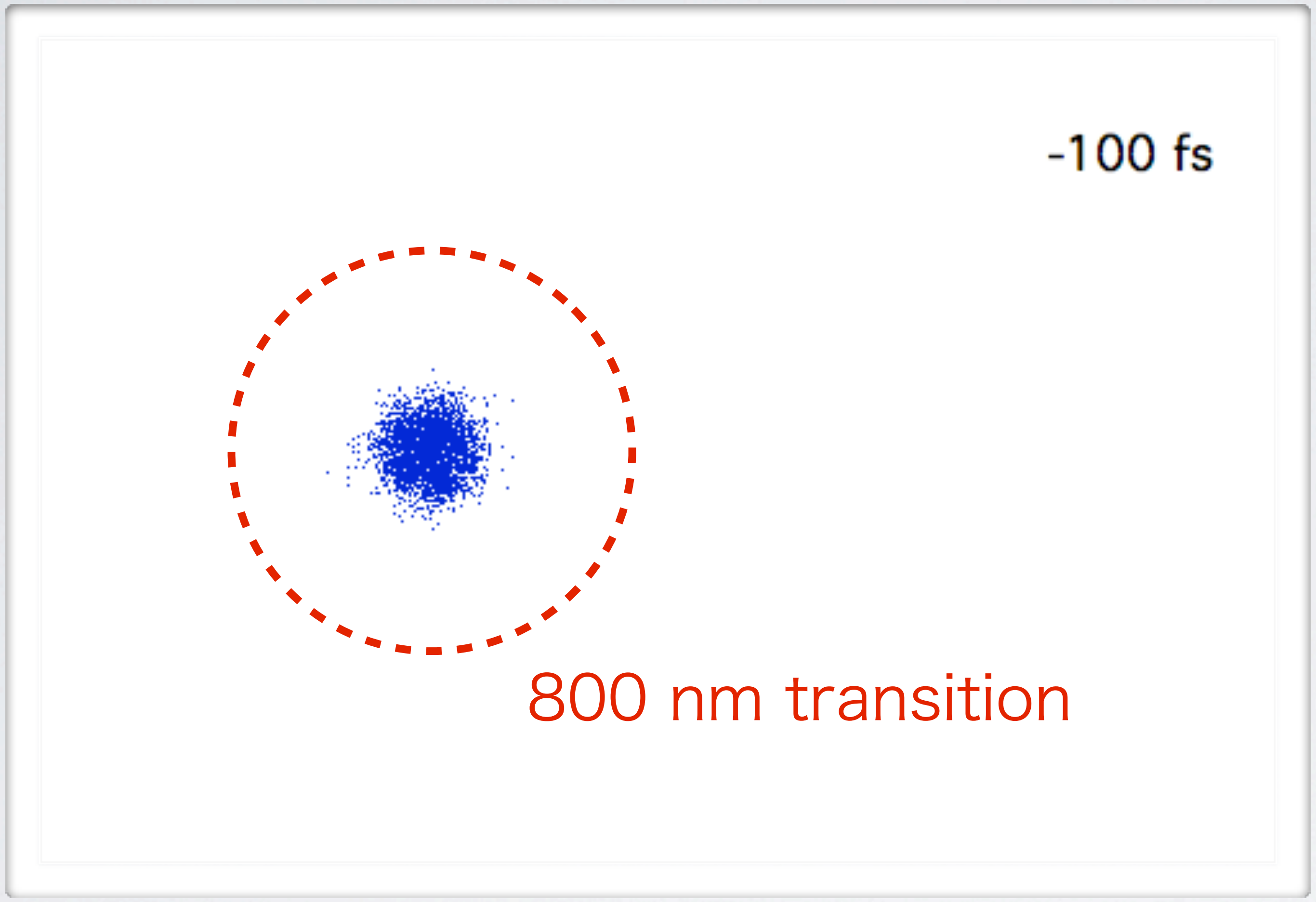
$$\frac{\partial f}{\partial t} + \frac{e\vec{E}}{\hbar}\cdot\frac{\partial f}{\partial \vec{k}} = \left.\frac{\partial f}{\partial t}\right|_{col}$$

$$= \left.\frac{\partial f}{\partial t}\right|_{e-e} + \left.\frac{\partial f}{\partial t}\right|_{h-h} + \left.\frac{\partial f}{\partial t}\right|_{OPE}$$

- No screening
- $\omega_{TO} = 160$ meV

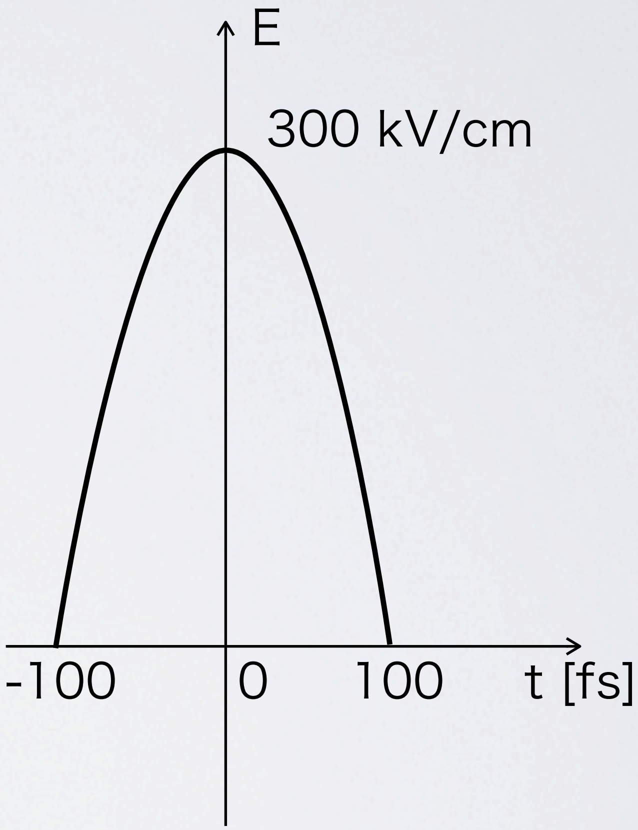
Time Development of Hole Distribution

Hole distribution in the momentum space



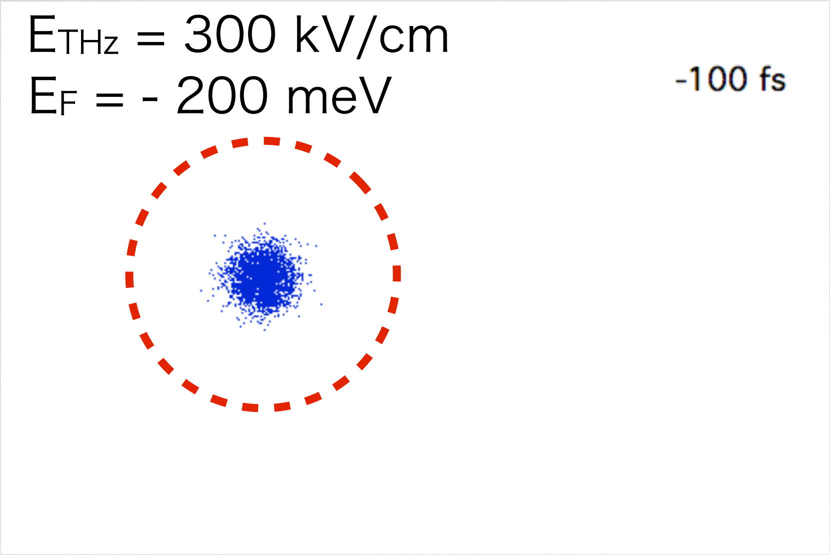
E_{THz} = 300 kV/cm

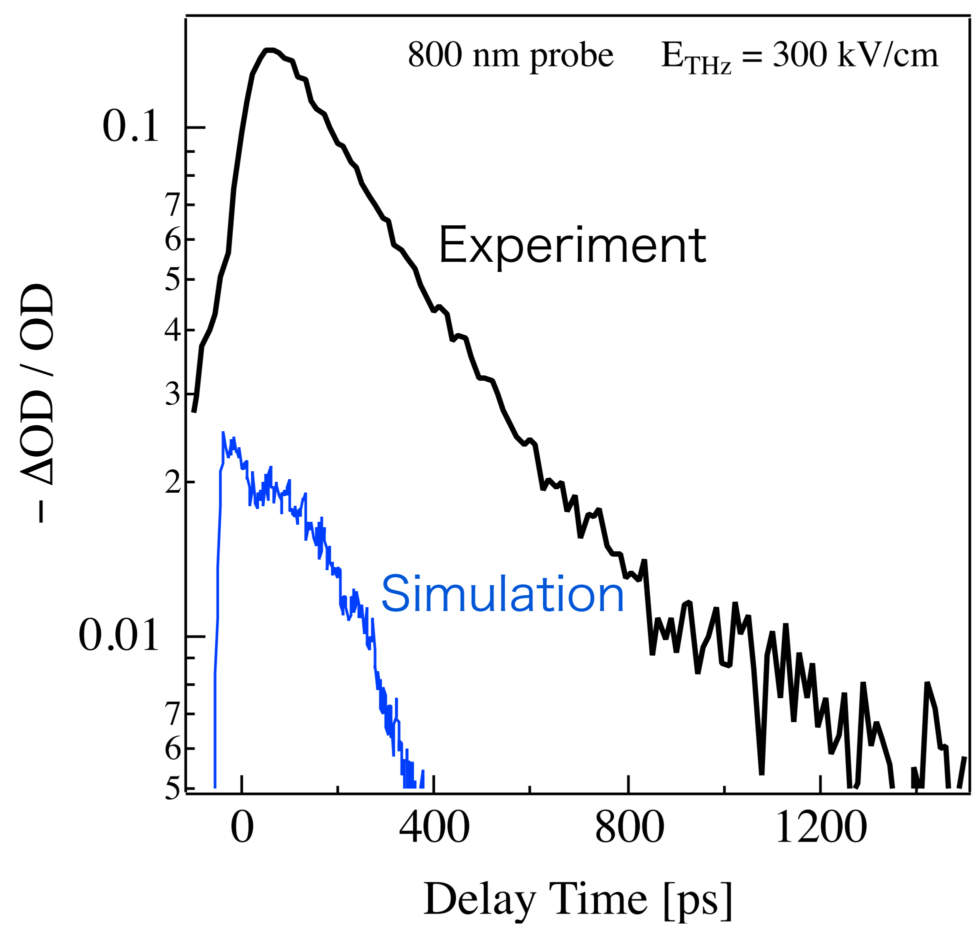
E_F = - 200 meV

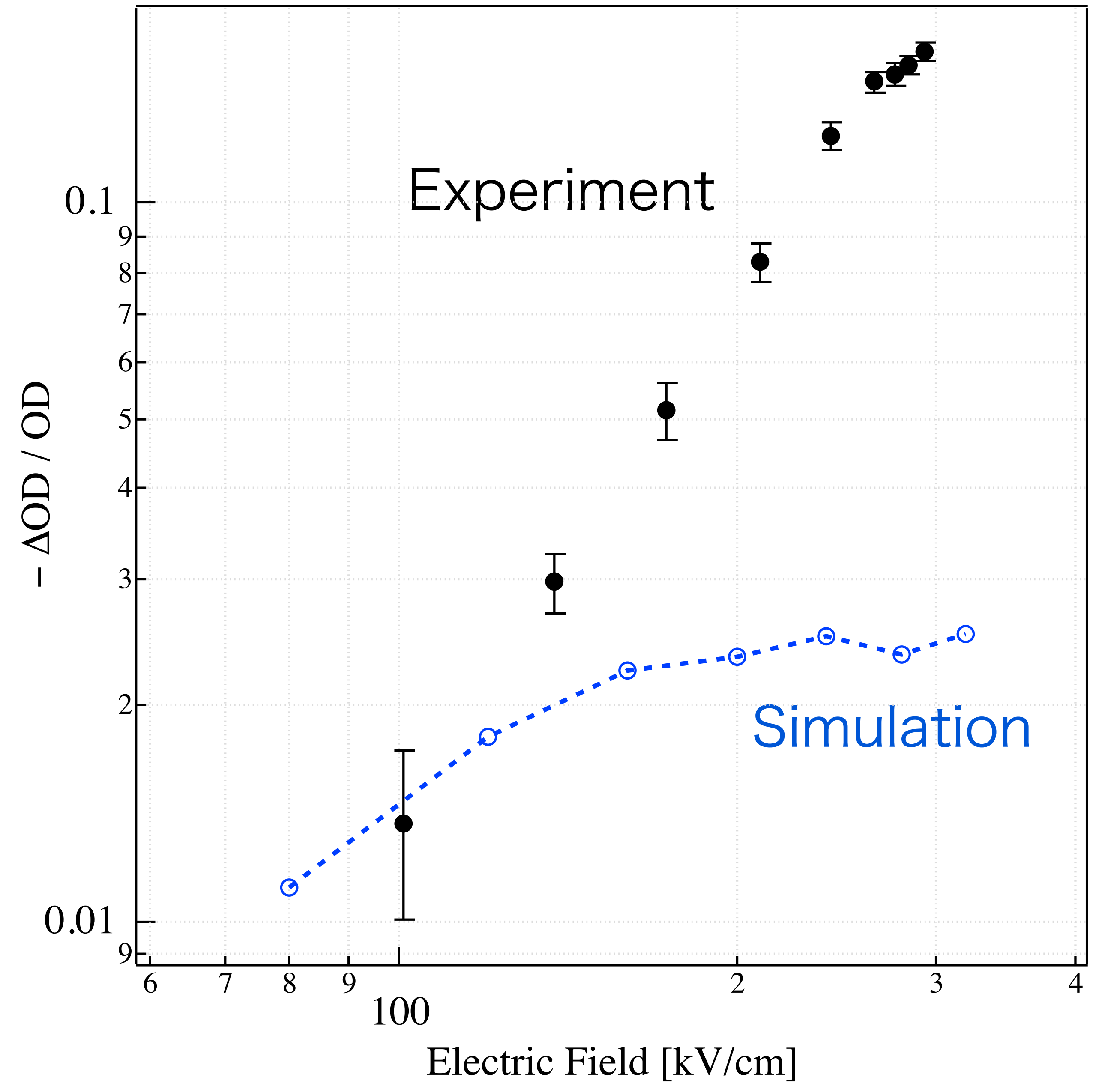


Parameters are chosen to reproduce experimental results.

Simulation Results



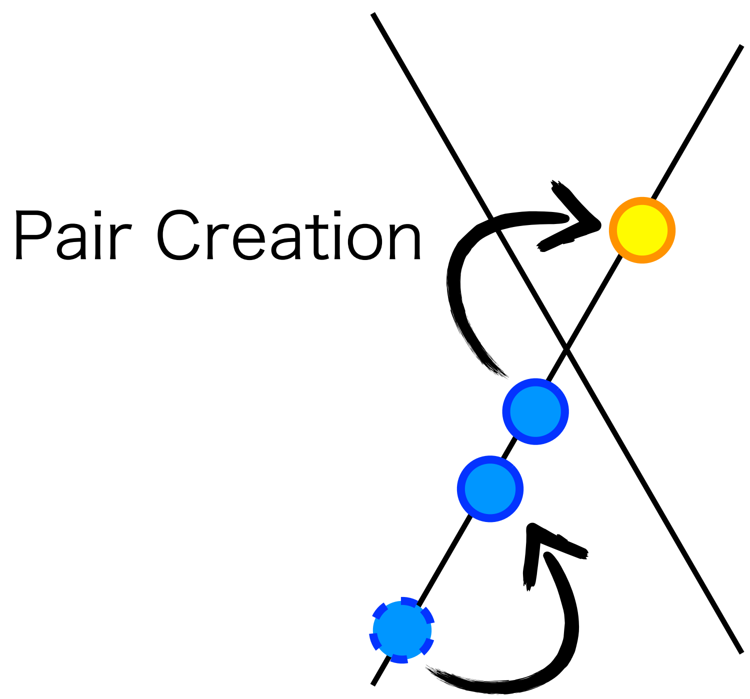




Carriers may not be enough.

Interband Carrier Scattering

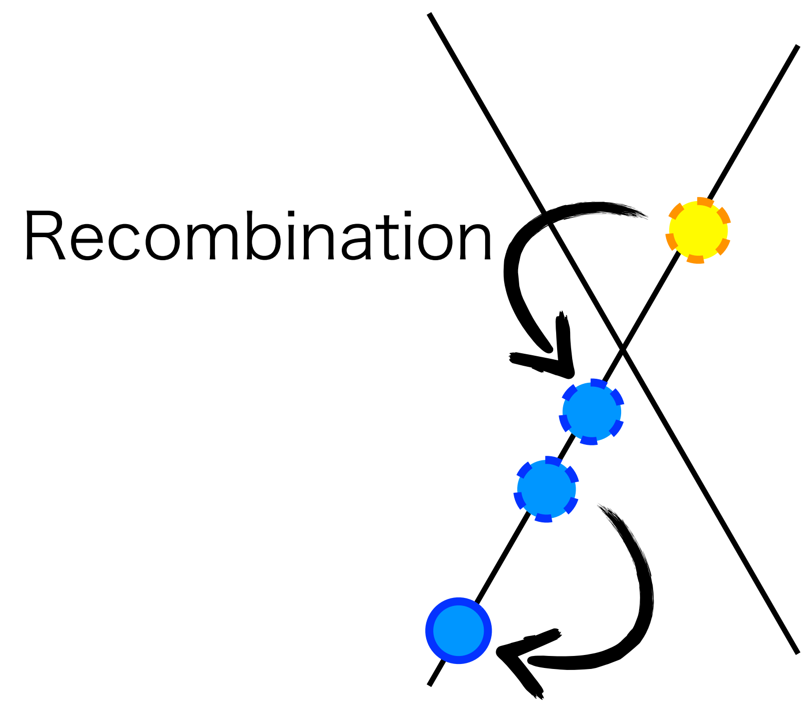
Impact Ionization



$$h \rightarrow h + h + e$$
$$e \rightarrow e + e + h$$

$\Rightarrow$ Carrier multiplication

Auger Recomibination

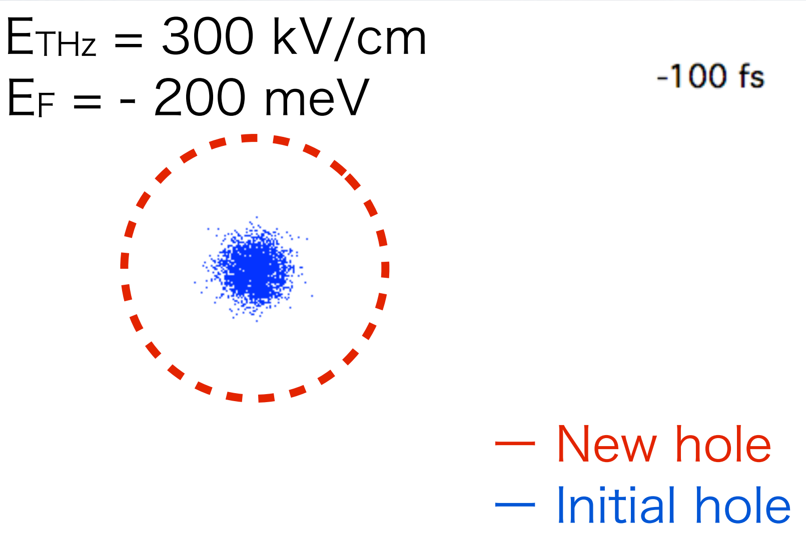


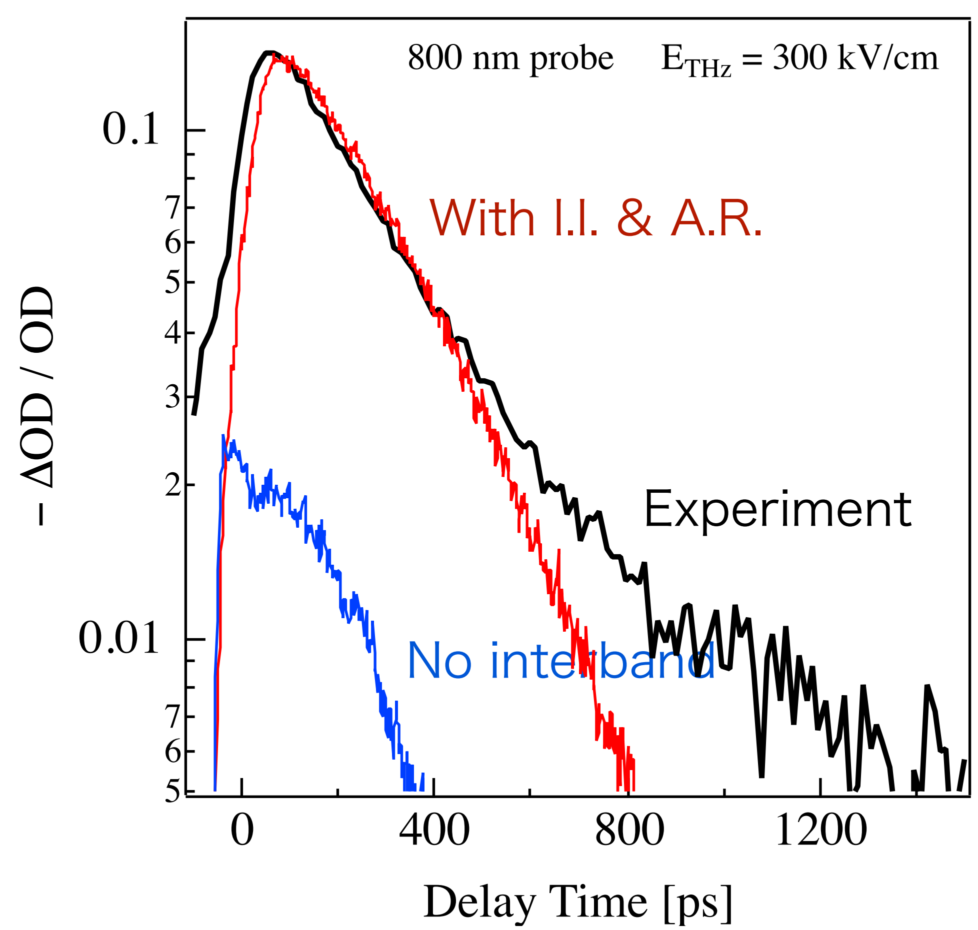
$$h + h + e \rightarrow h$$
$$e + e + h \rightarrow e$$

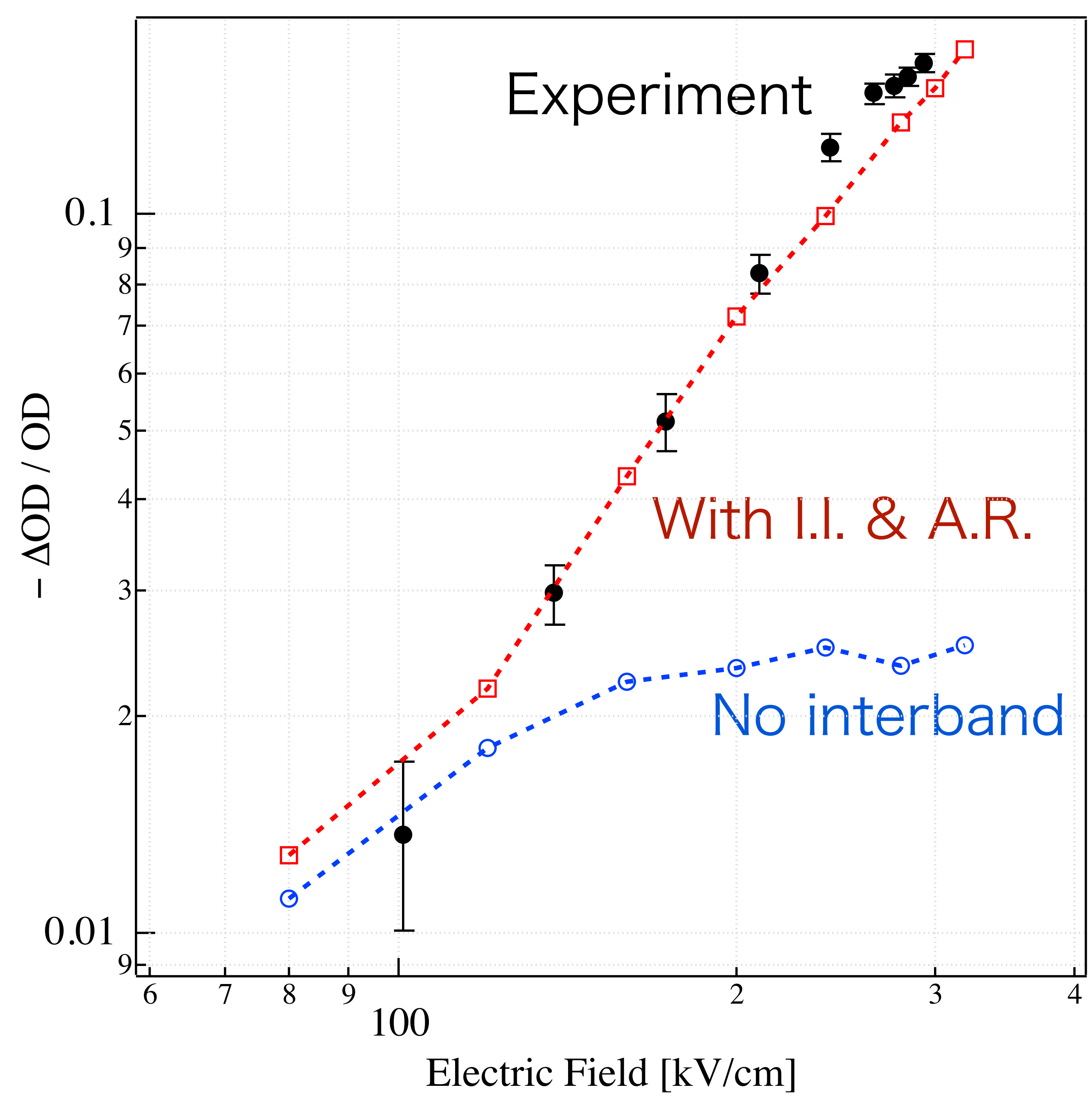
$\Rightarrow$ Carrier annihilation

F.Rana, Phys. Rev. B 76, 155431 (2007)

Simulation With Interband Carrier Scattering

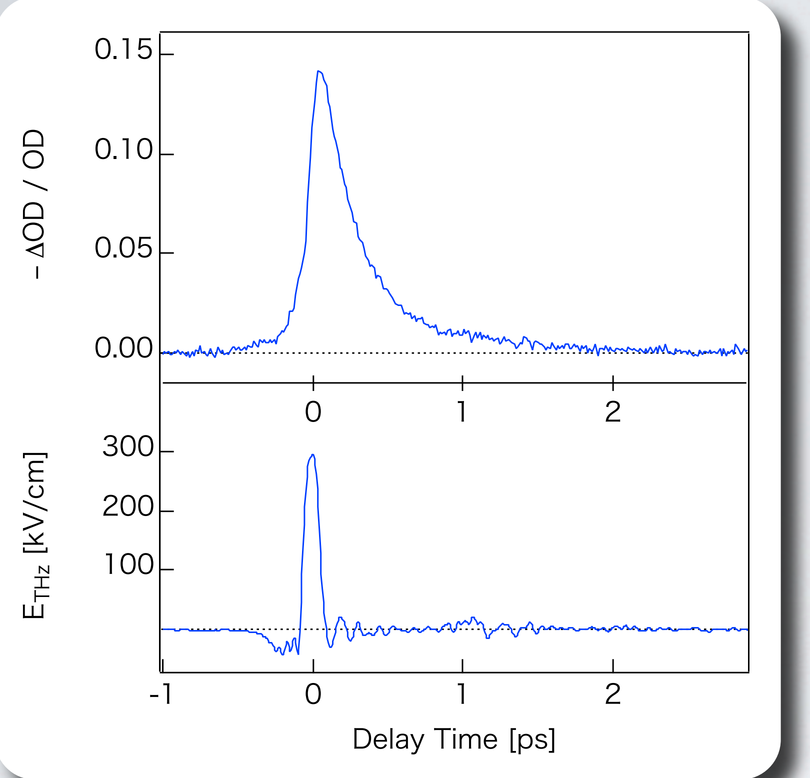






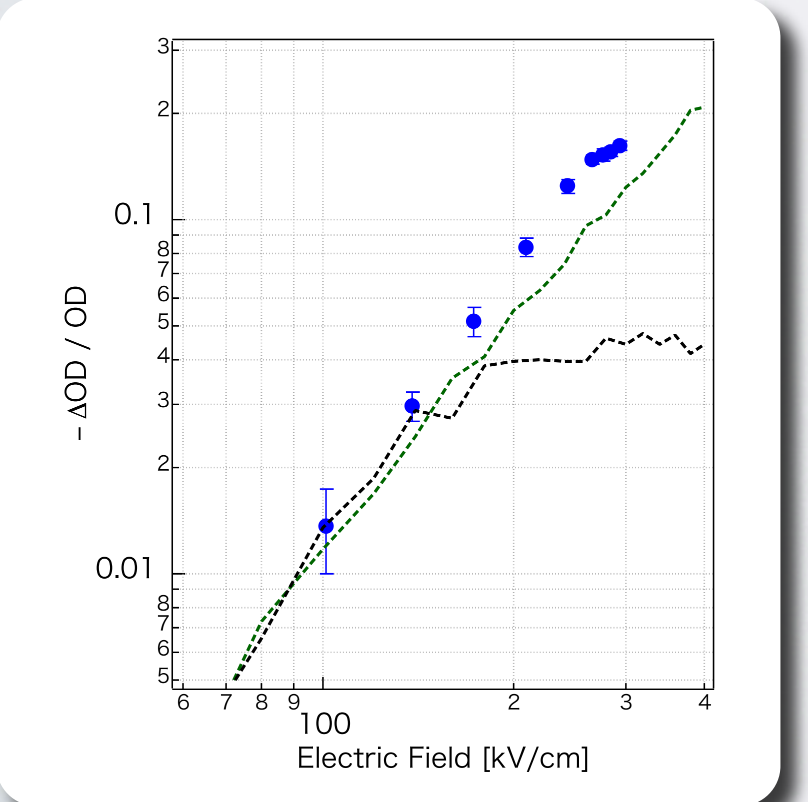
Carrier multiplication is important in extreme nonlinear regime.

Summary : Thank you for your attention



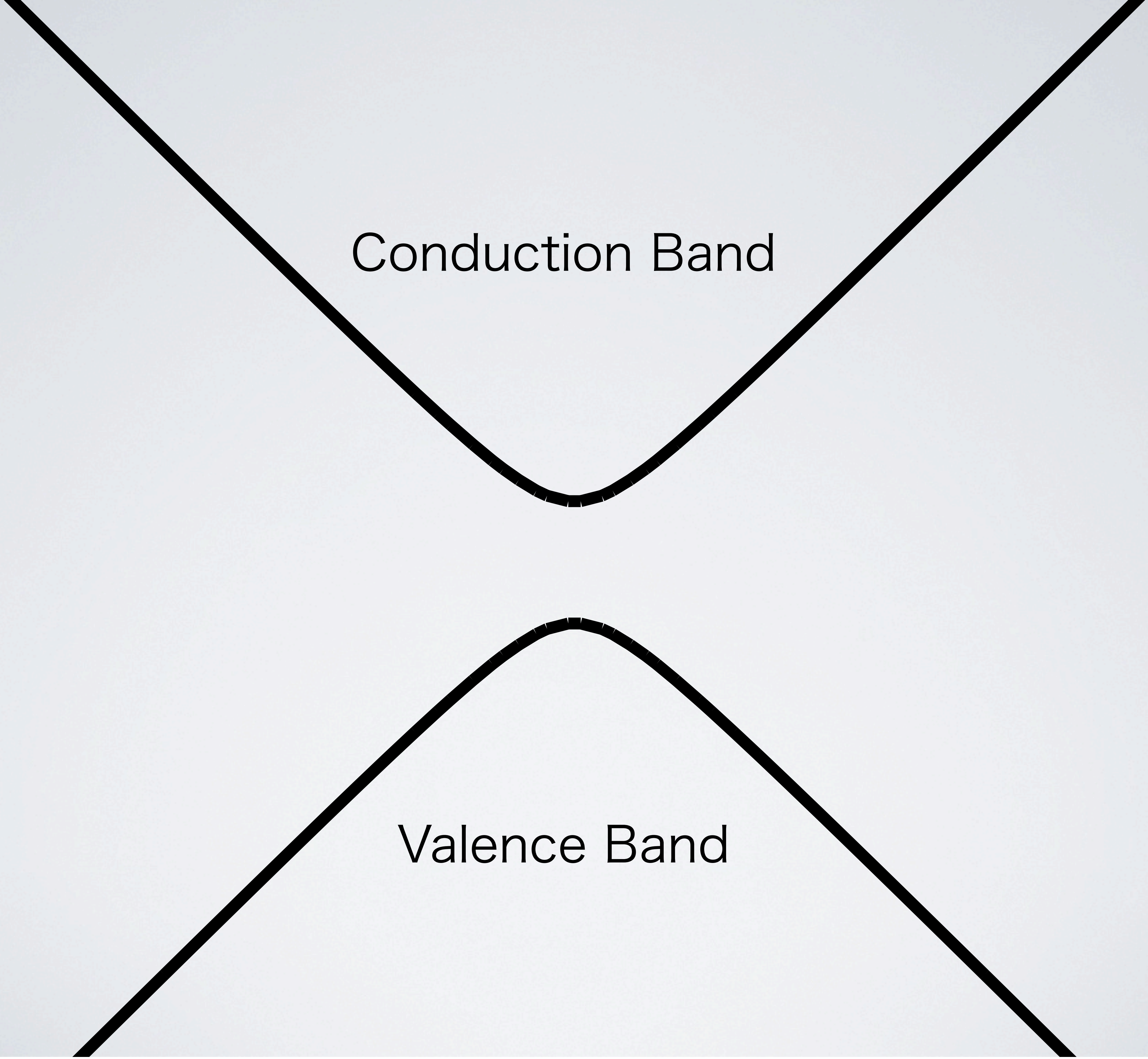
THz-field driven carrier dynamics is investigated with transient absorption measurement in NIR region.

Huge THz induced transparency over 14% is observed at 800 nm.



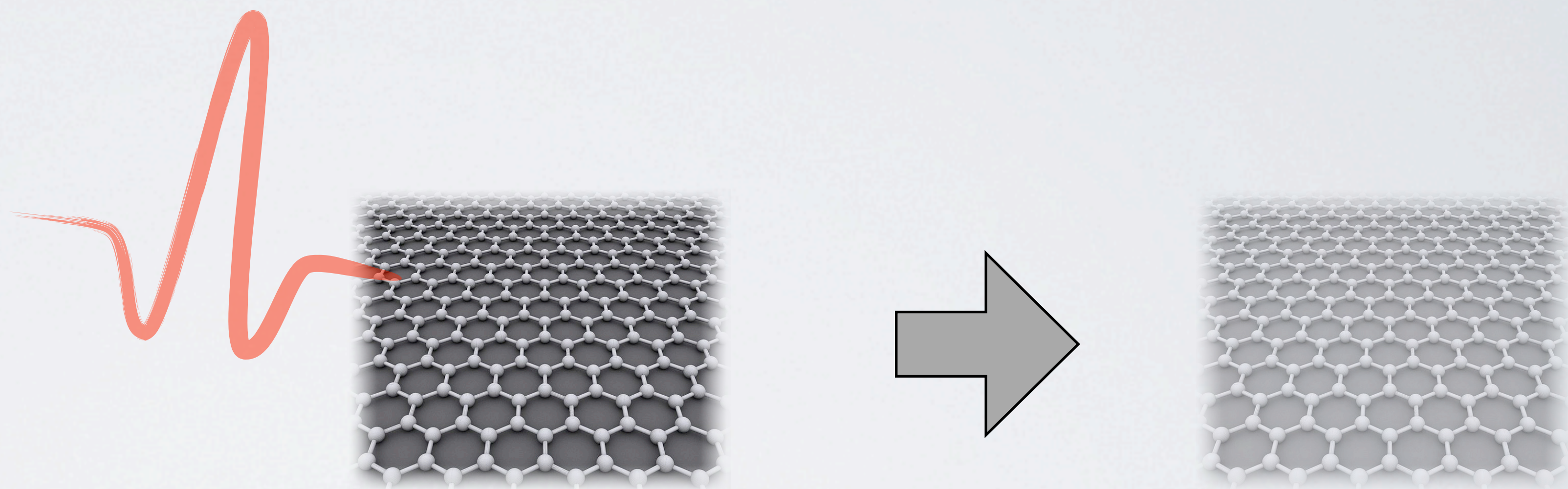
Carrier multiplication with interband carrier scattering is essential in high field dynamics.

Backup Slides


Conduction Band
Valence Band

Experiment

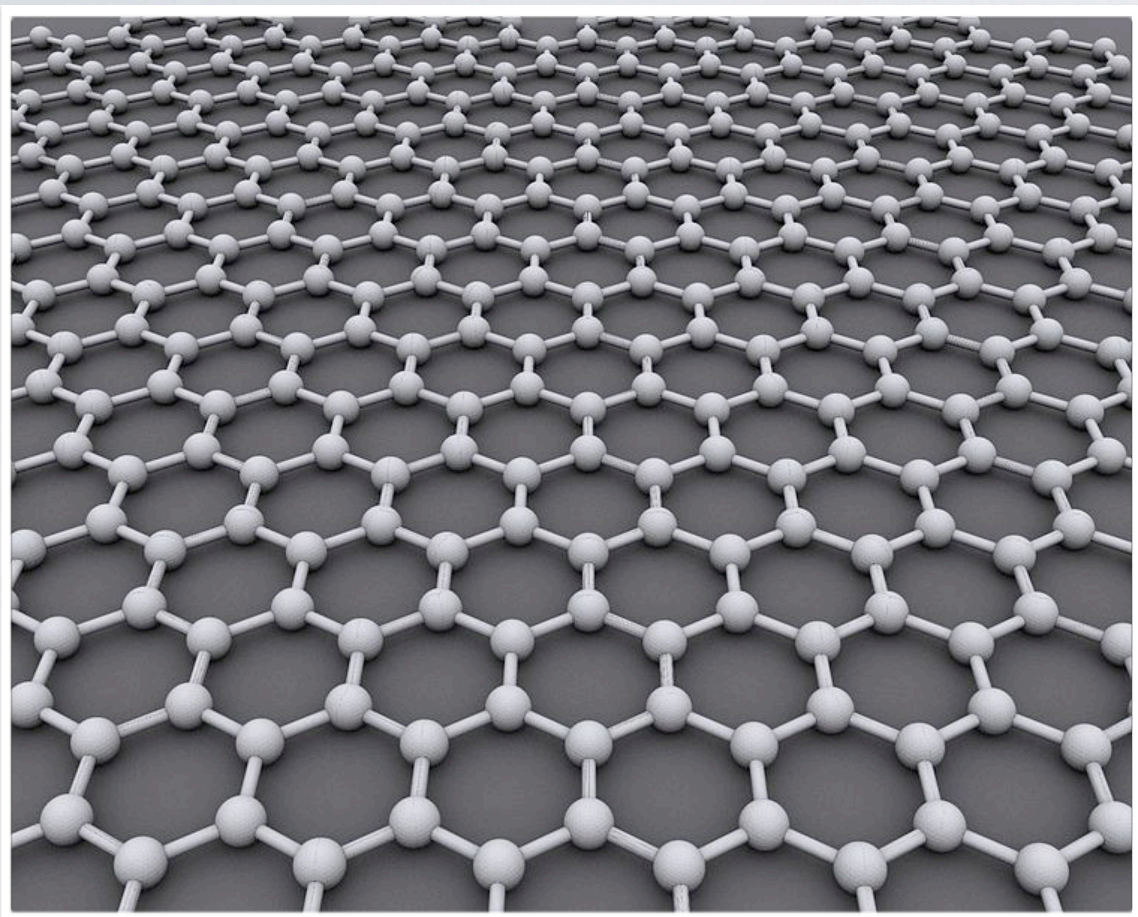
- THz pump - NIR transient absorption measurement.
- Induced transparency over **14 %** at 800 nm by THz wave.



Simulation

- Monte Carlo simulation including carrier-carrier and optical phonon scattering.

Graphene



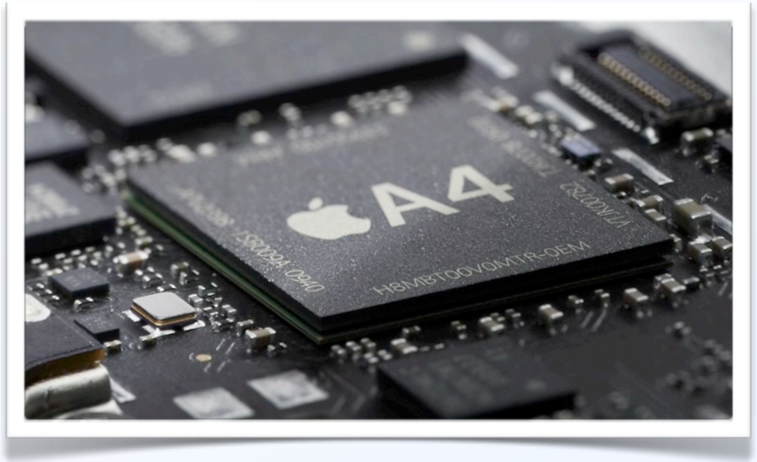
- Monoatomic layer carbon material
- Characteristic band structure

  ➡ Massless electrons
- High conductivity at T = 300 K

  ➡ Promising material for ultra high-speed nano devices

  Graphene transistor, graphene LSI, etc...

High Field Carrier Dynamics in Graphene



Typical Nano Device

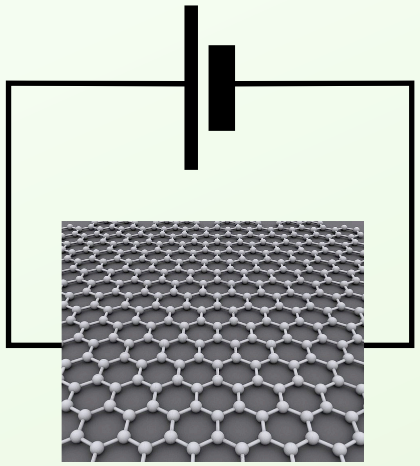
Voltage ~ 1 V

Length ~ 10 nm

→ Electric Field ~ 0.1 V/nm = 1 MV/cm

High electric field ⇒ Extremely non-equilibrium state

For future applications such as high-speed devices, ultrafast carrier dynamics under high field is important.



Under high DC field

J. Phys. Cond. Mat. 21 (2009) 344201

- Drift velocity saturates by phonon scattering.

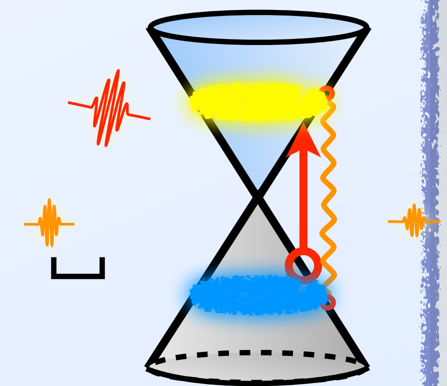
→ Averaged dynamics

→ Difficult to understand ultrafast response

High Field Carrier Dynamics in Graphene

According to optical-pump optical-probe measurement

- Carrier-carrier scattering : 2 ~ 10 fs
- Optical phonon emission : 100 ~ 400 fs



Requirement

To investigate ultrafast carrier dynamics under high field, time resolution better than 100 fs is required.

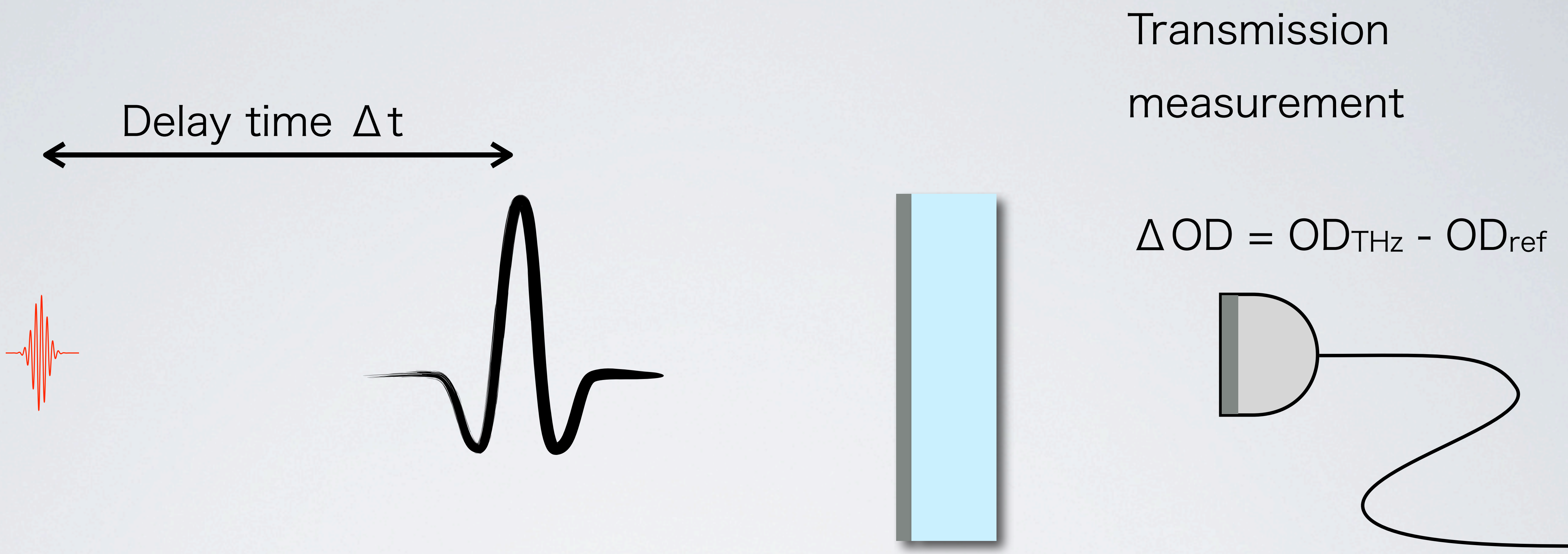
Solution

Near-infrared transient absorption measurement with single cycle THz pulse excitation

→ Apply high electric field in very short time duration

→ High time resolution ~ 50 fs

Experimental Condition



Probe pulse

- 800 nm
- Pulse width : 50 fs

THz pulse

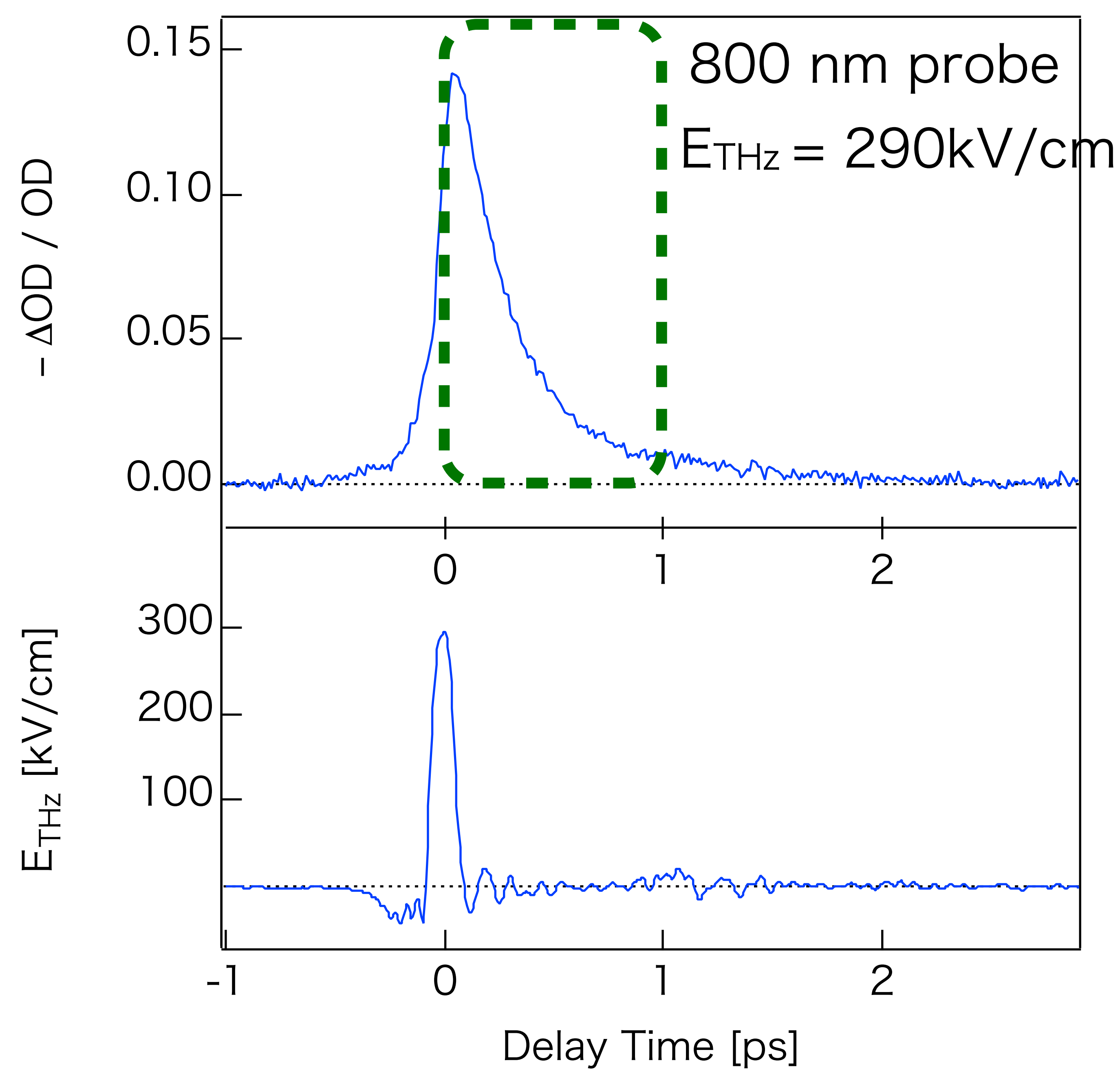
- Air plasma method
- Field : ~300 kV/cm
- Pulse width : 100 fs

Sample

- CVD growth graphene
- On SiO_2 substrate
- Transmission 97 % at 800 nm
- T = 294 K

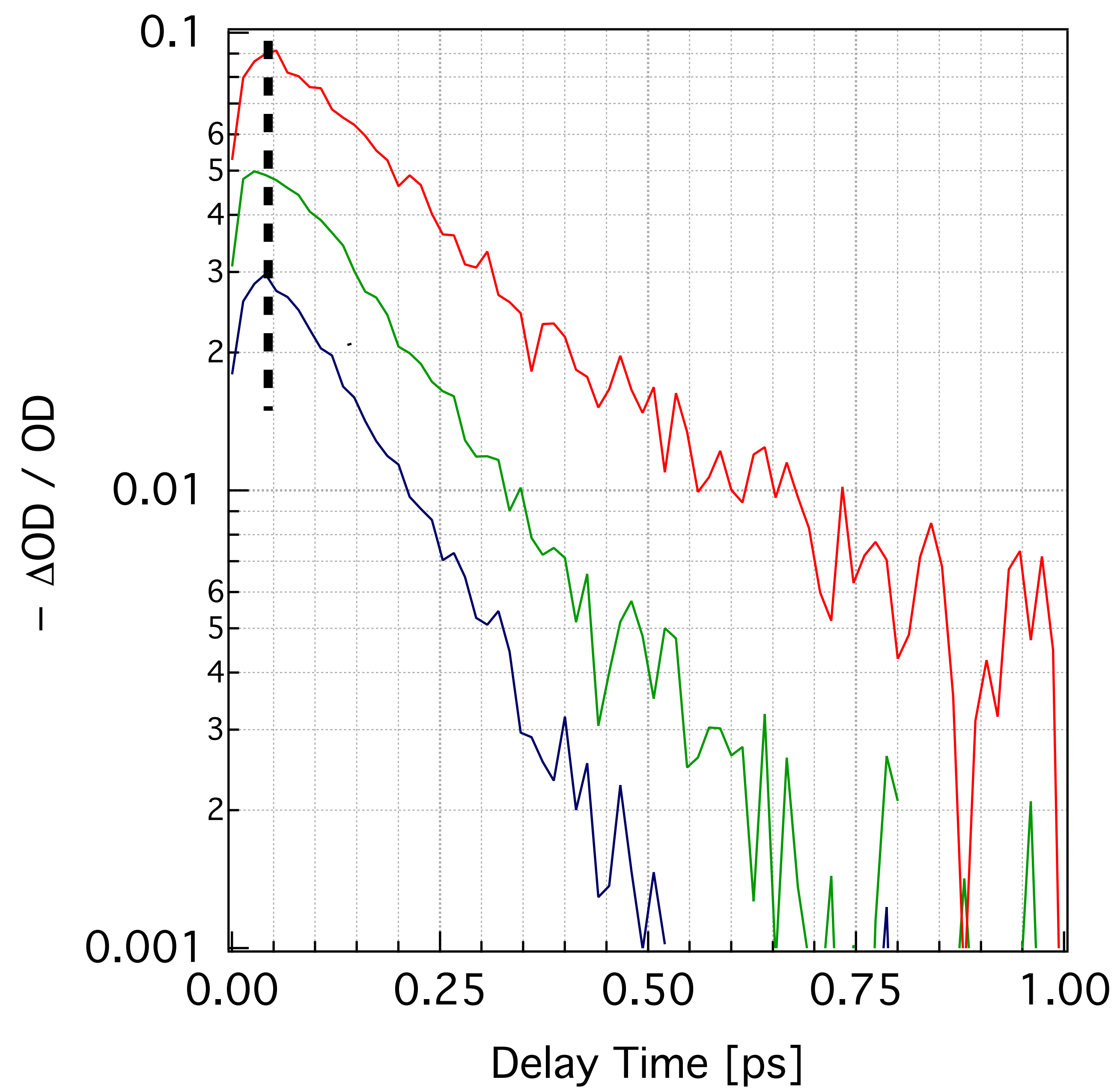
J.Dai, et.al., Phys. Rev. Lett. 103, 023001 (2009)

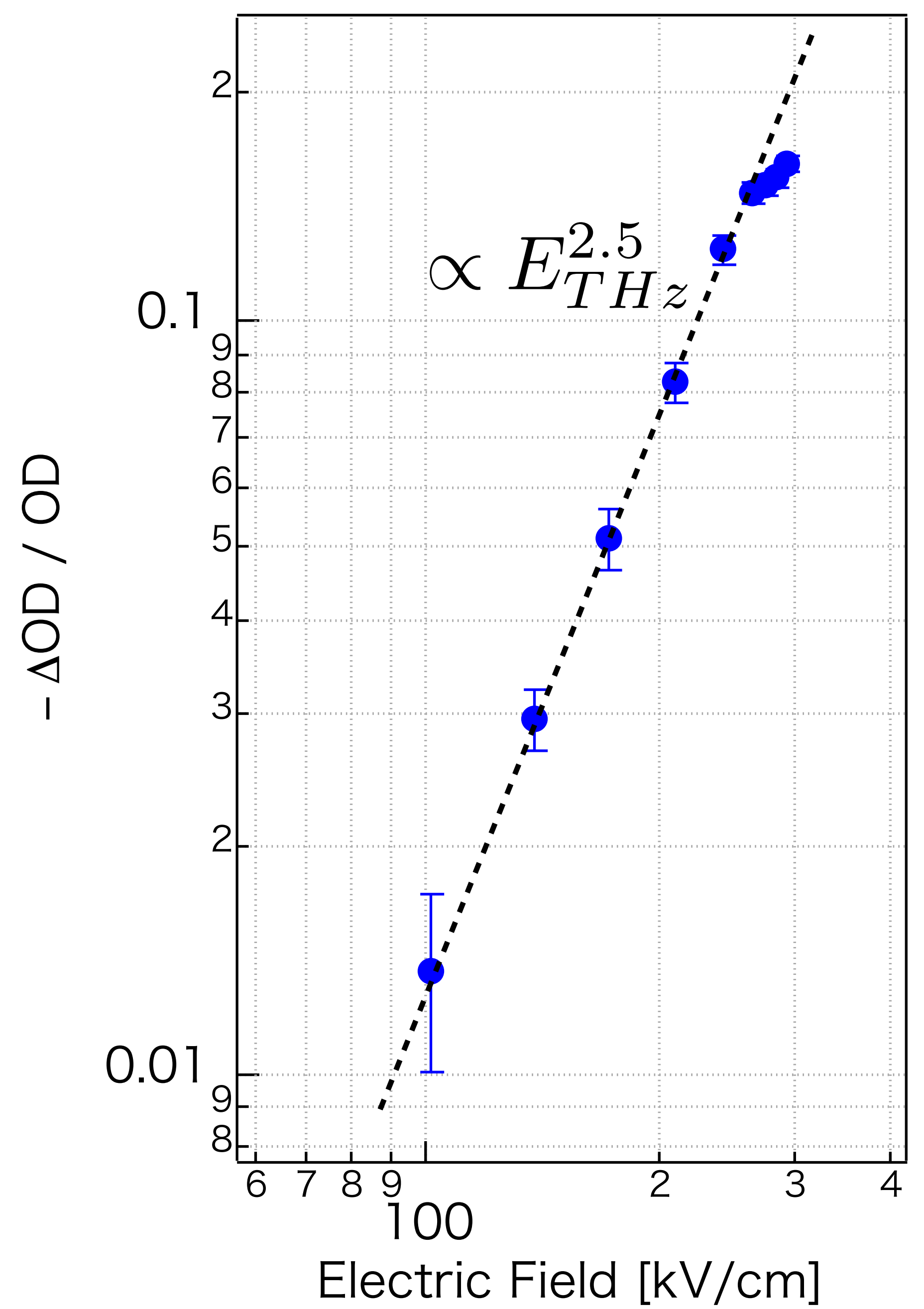
Results : Typical THz-induced Transparency in Near-infrared Region



THz induced transparency over 14% at 800 nm

Results : Field Dependence





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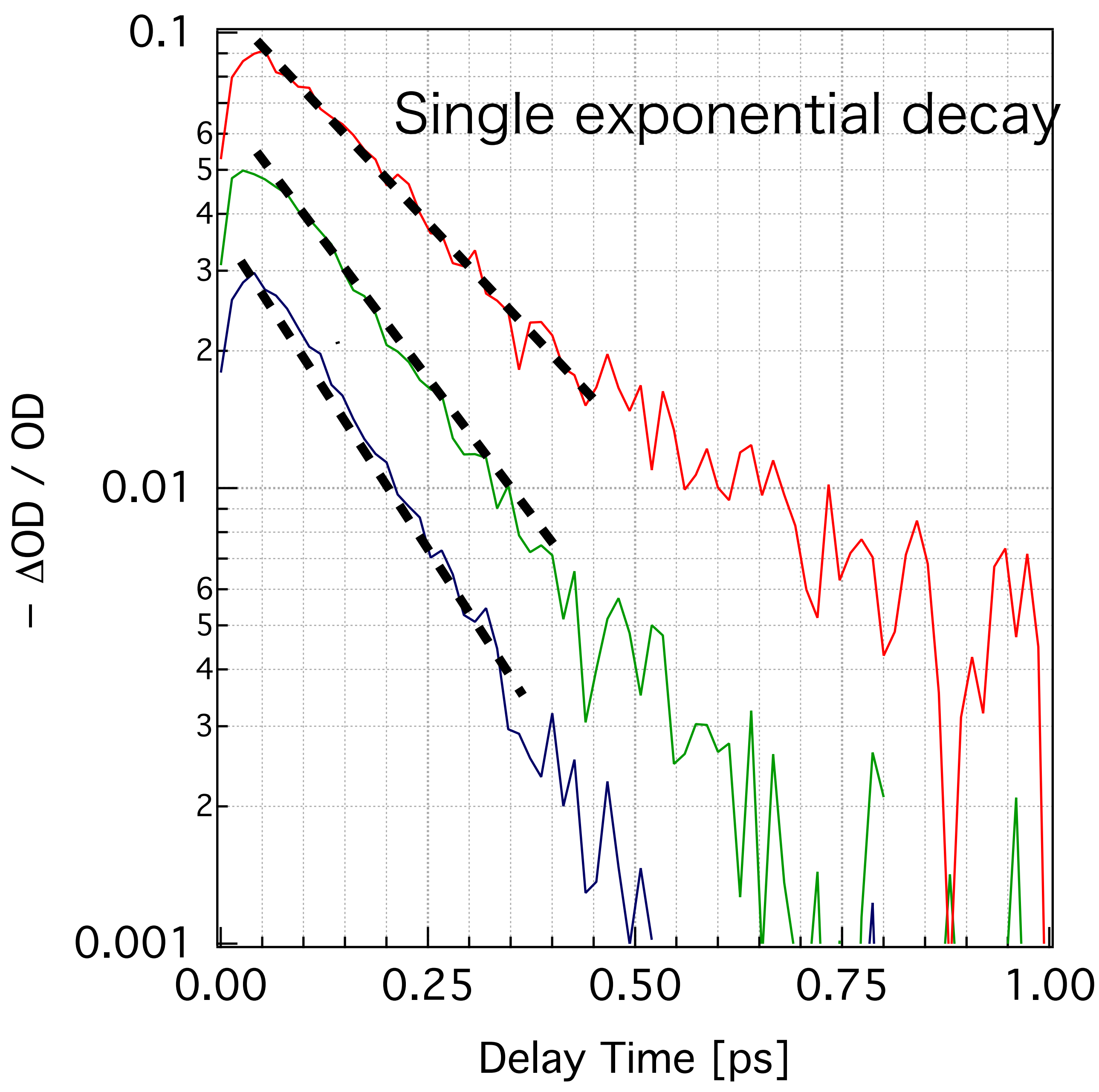
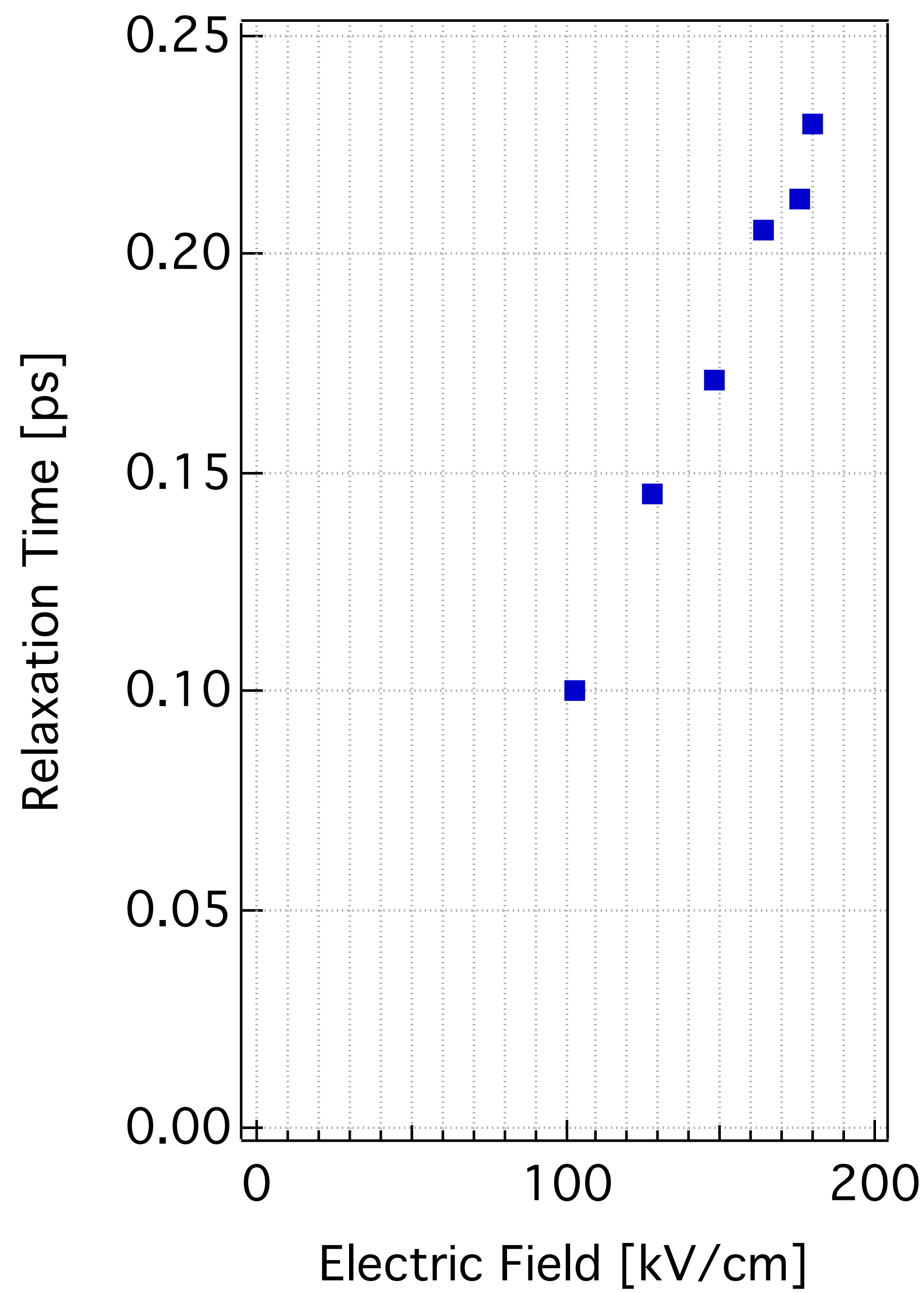
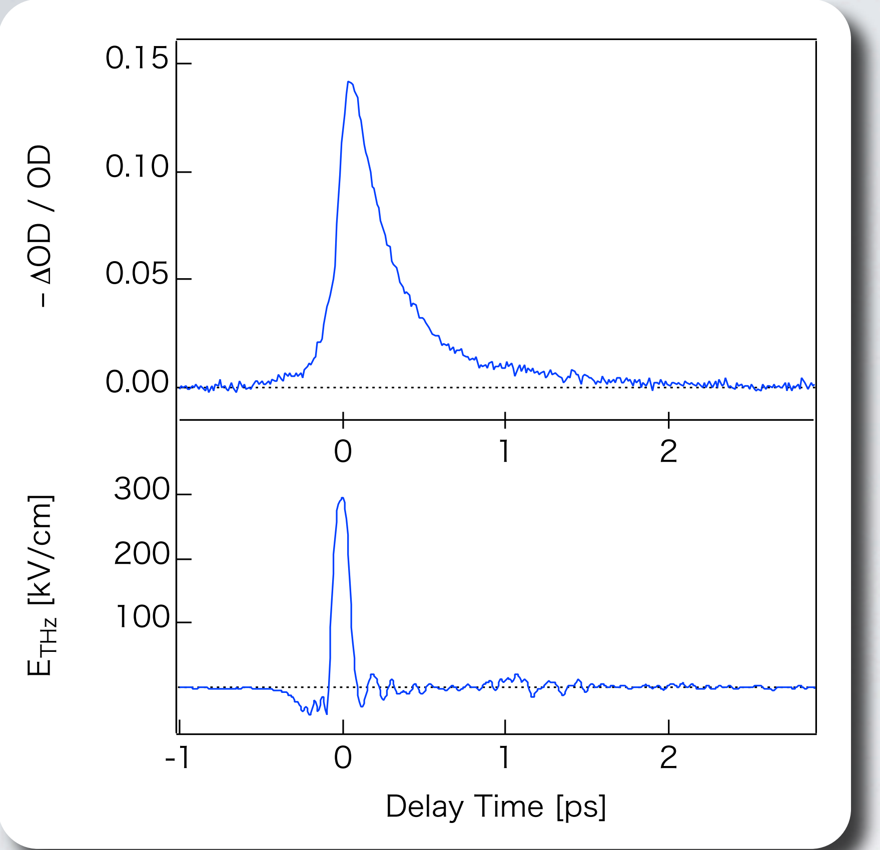
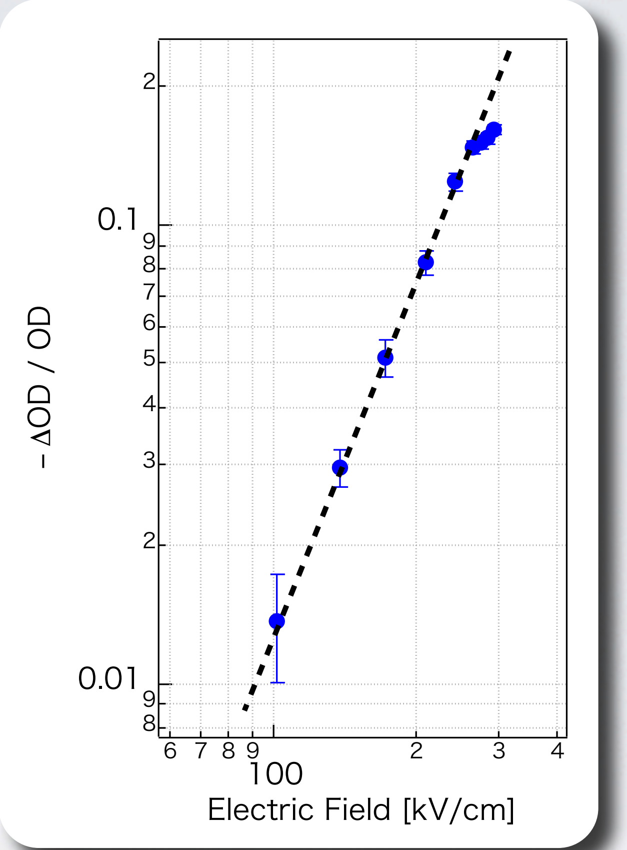
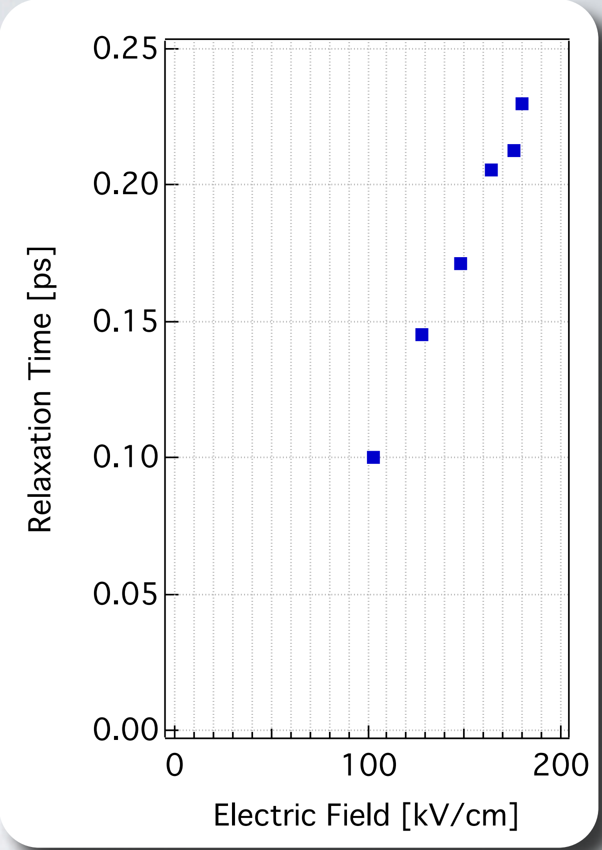




Summary : Experimental Results







THz induced transparency over 14% at 800 nm

$$-\Delta OD/OD \propto E_{THz}^{2.5}$$

High field

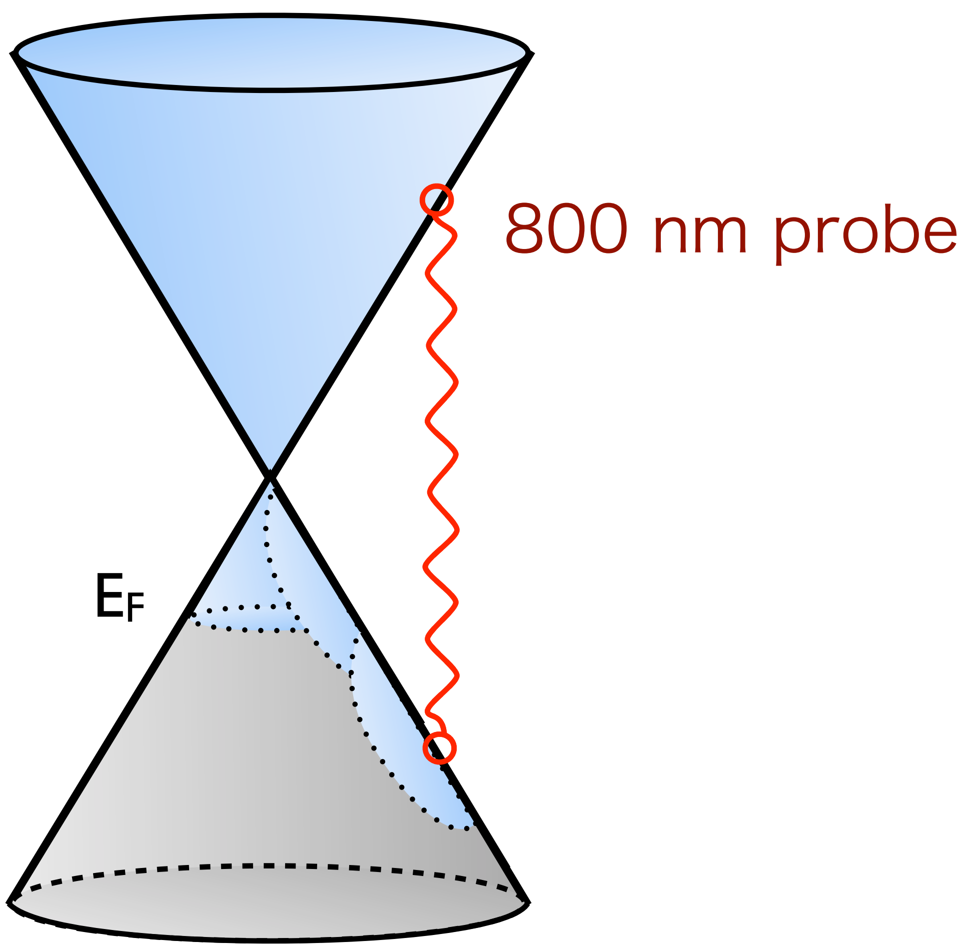
↓

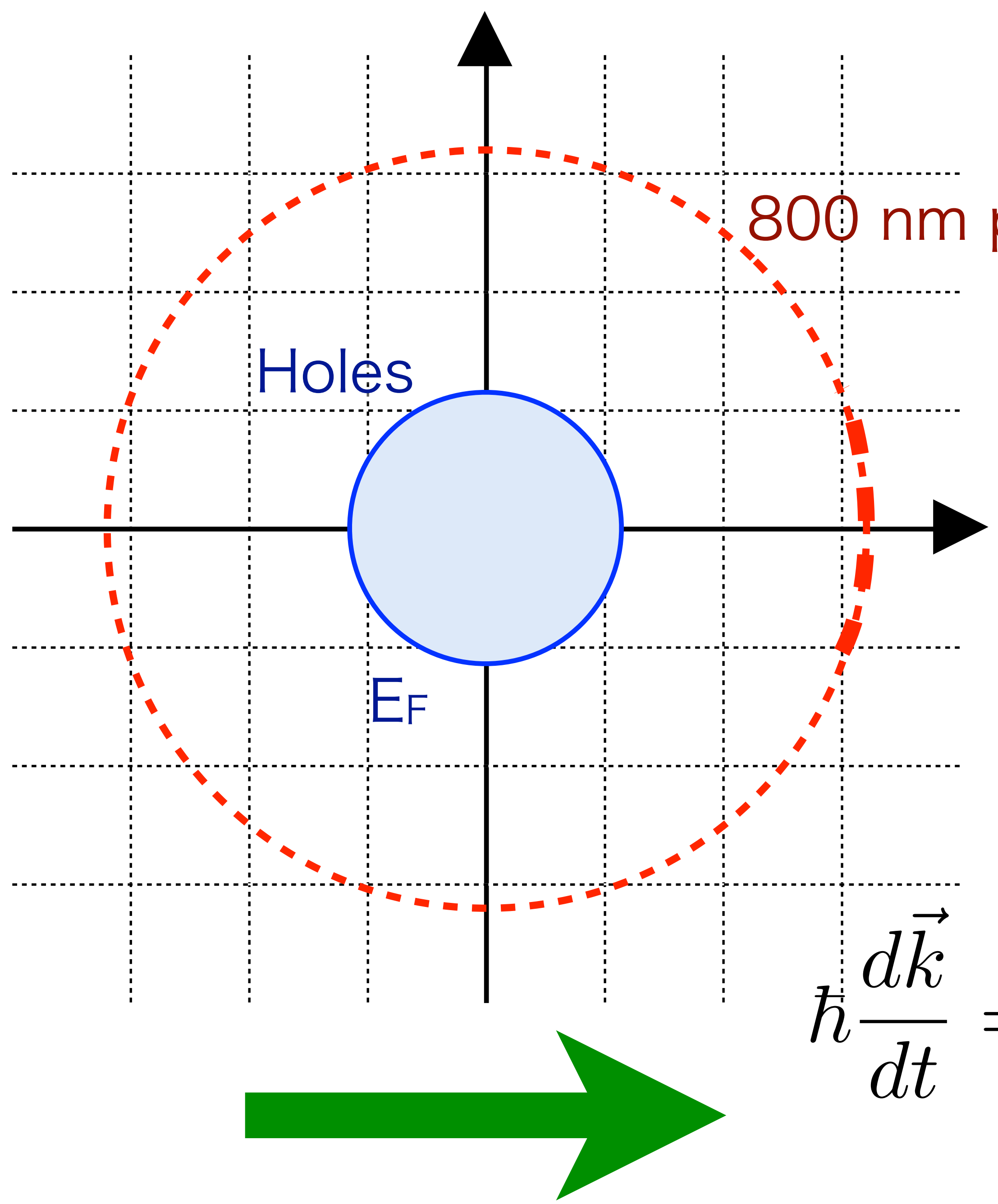
Longer relaxation time

→ Monte Carlo simulation

Origin of THz Induced Transparency

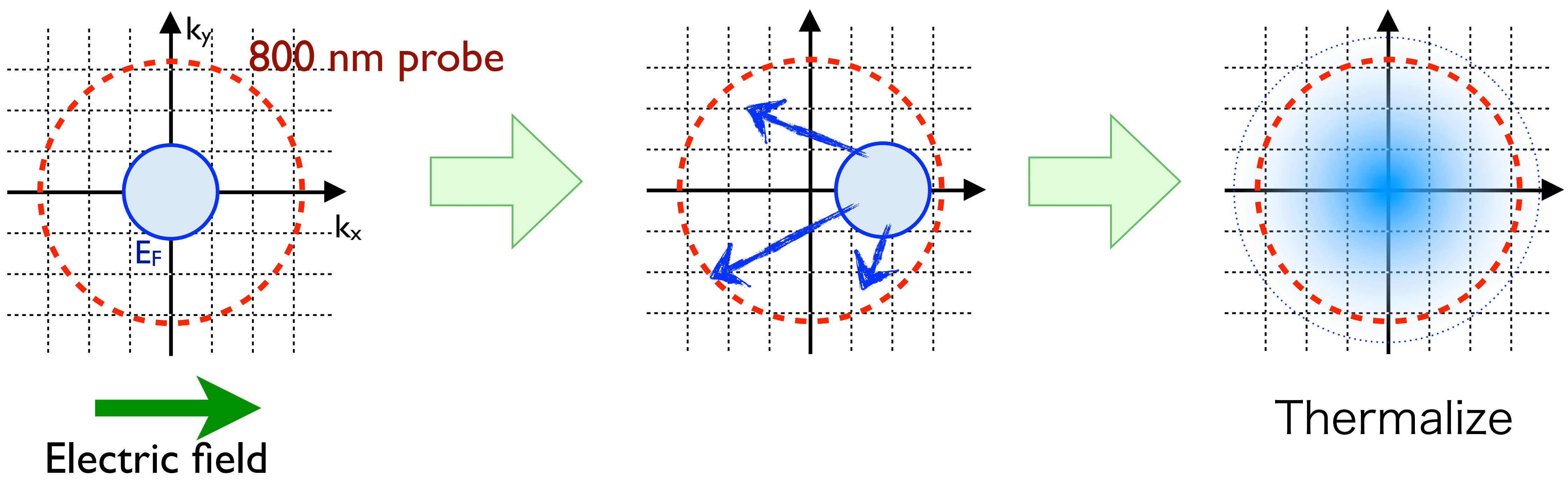
Hole distribution in the **momentum space**





$$\hbar \frac{d\vec{k}}{dt} = e\vec{E}$$

Carrier Scattering Process



Boltzmann equation for $f(\vec{k},t)$

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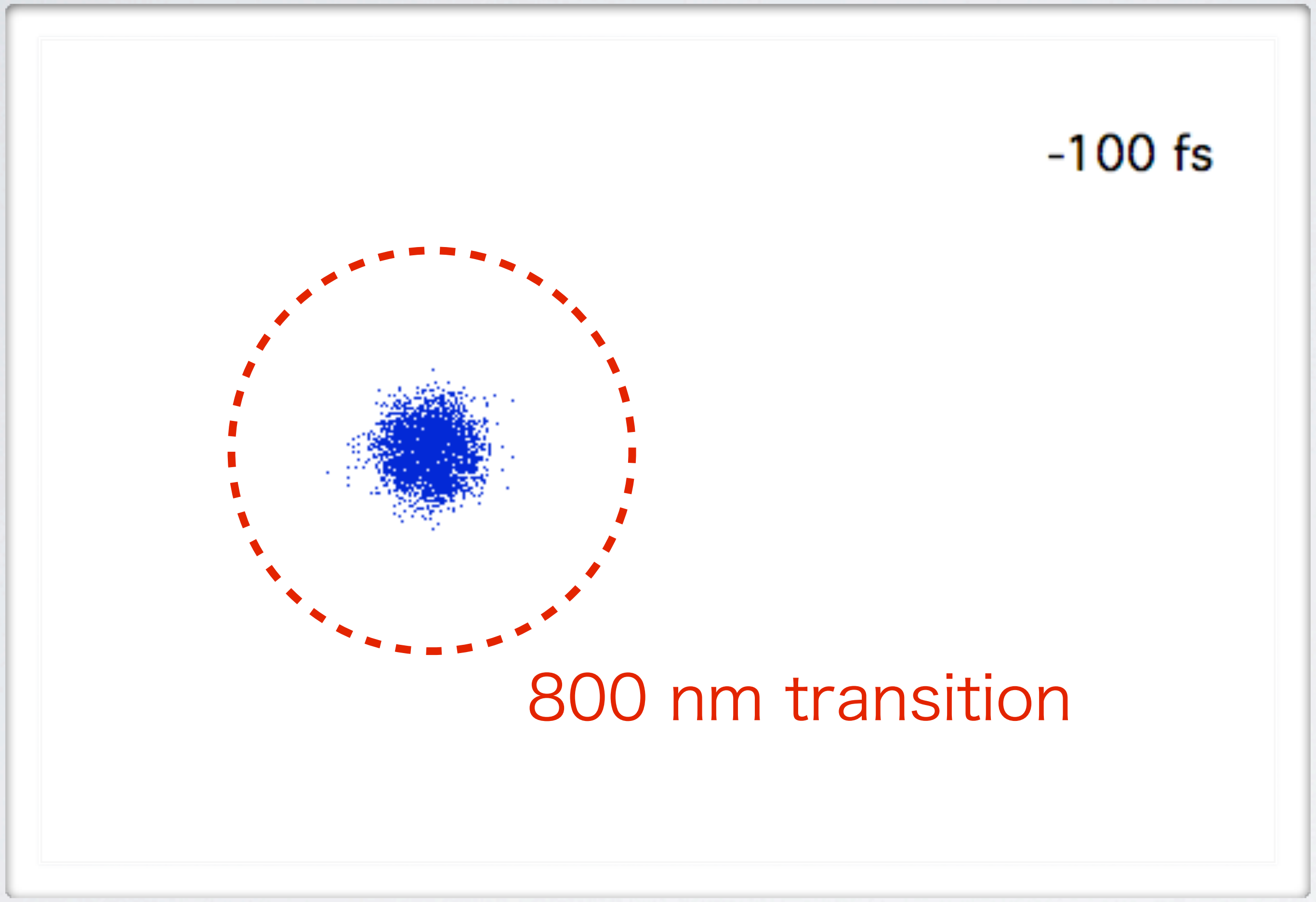
$$= \left.\frac{\partial f}{\partial t}\right|_{e-e} + \left.\frac{\partial f}{\partial t}\right|_{h-h} + \left.\frac{\partial f}{\partial t}\right|_{OPE}$$

- No screening
- $\omega_{TO} = 160$ meV

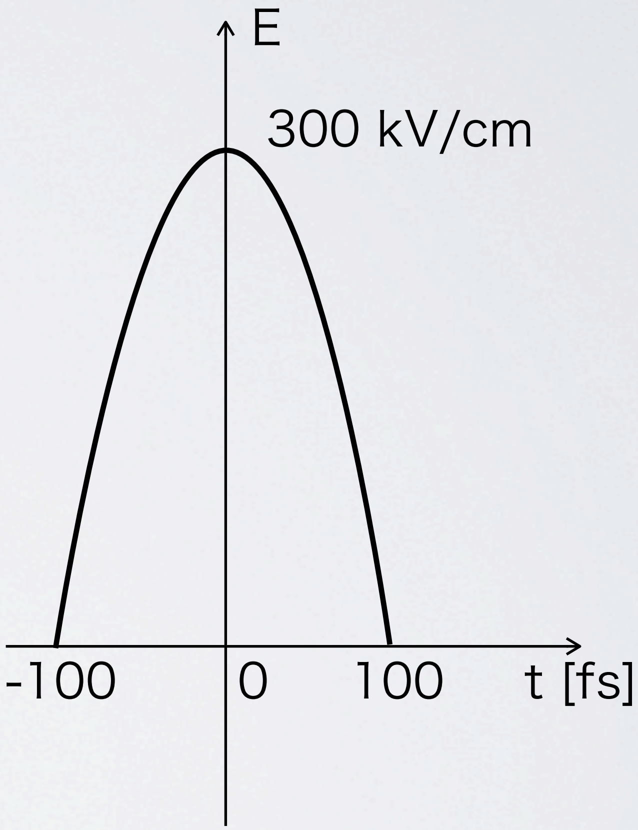
Time Development of Hole Distribution

Hole distribution in the momentum space



E_{THz} = 300 kV/cm

E_F = - 200 meV



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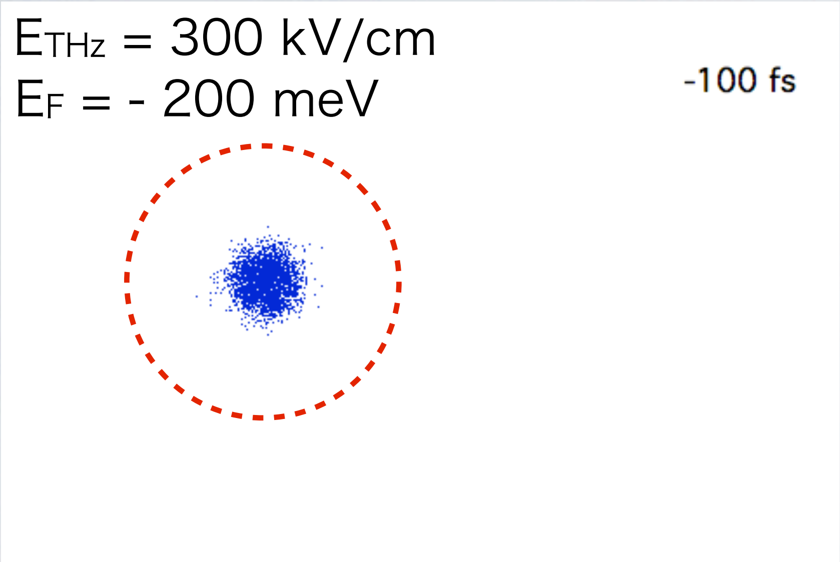

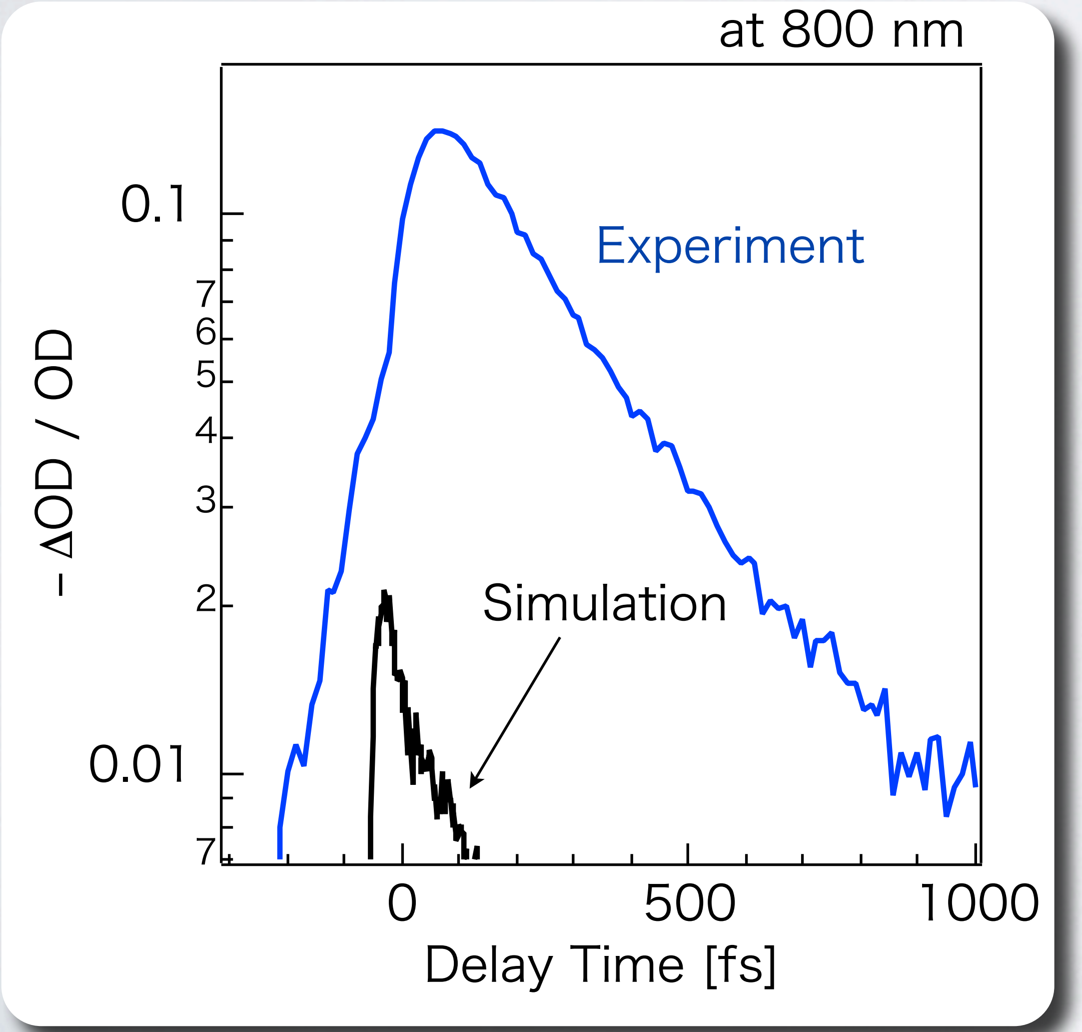

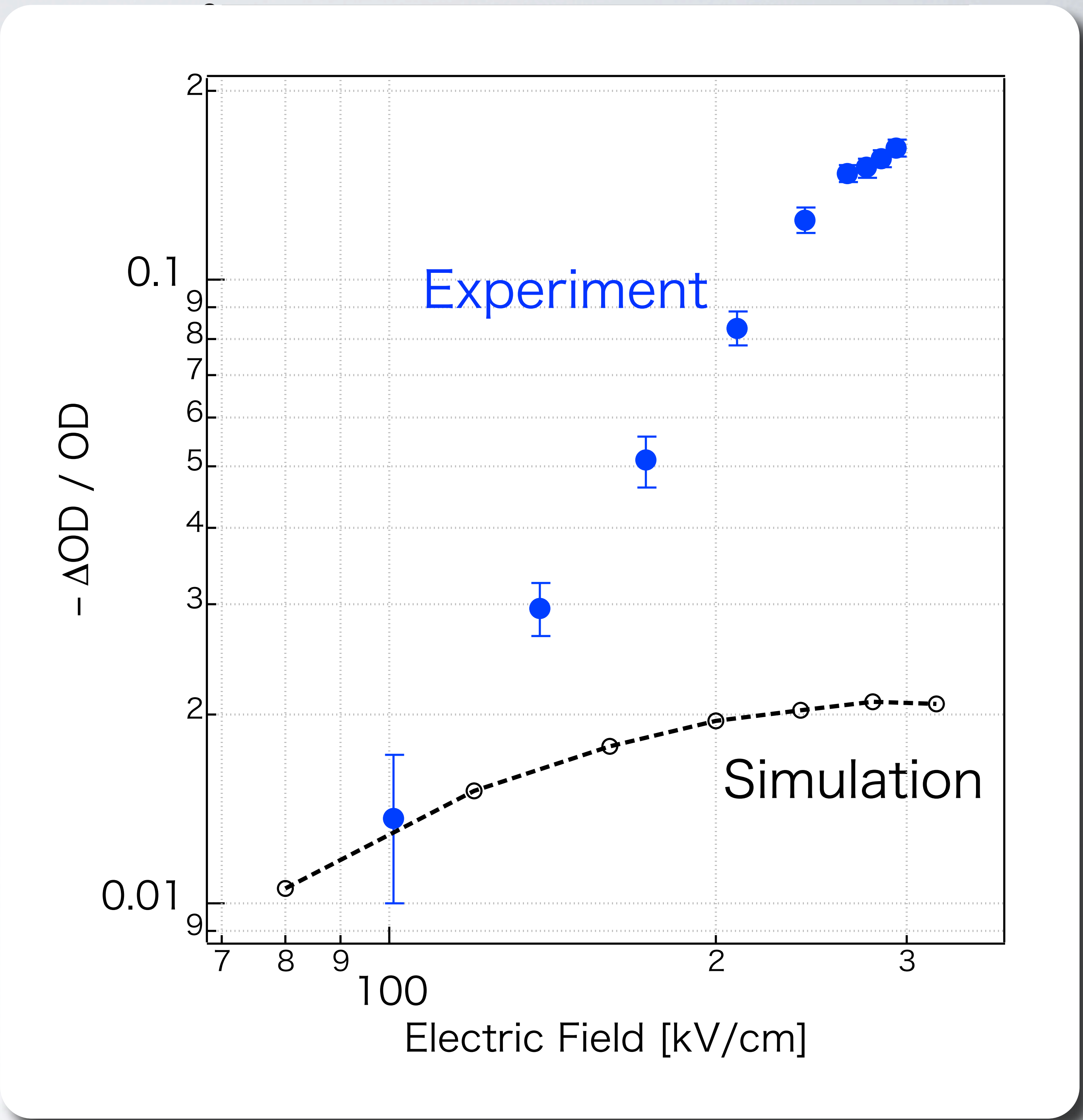

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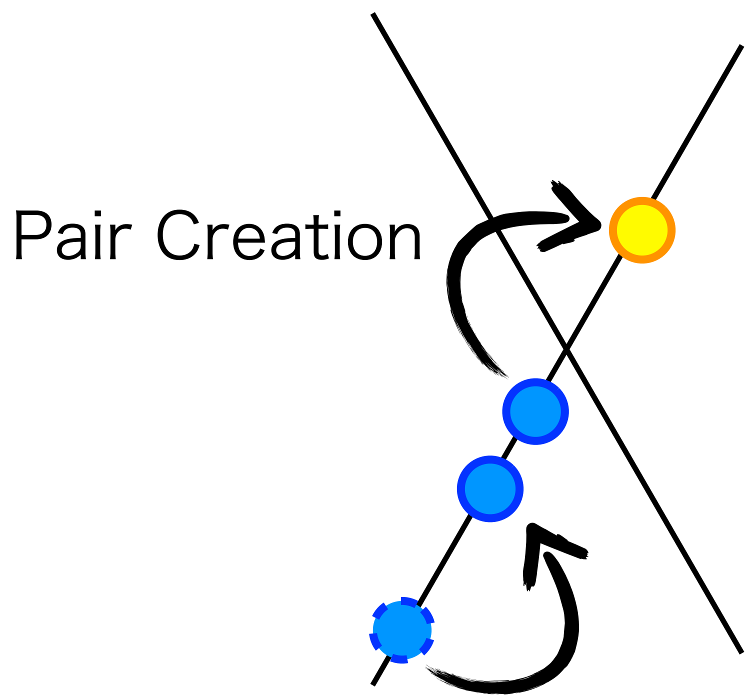
Interband Carrier Scattering

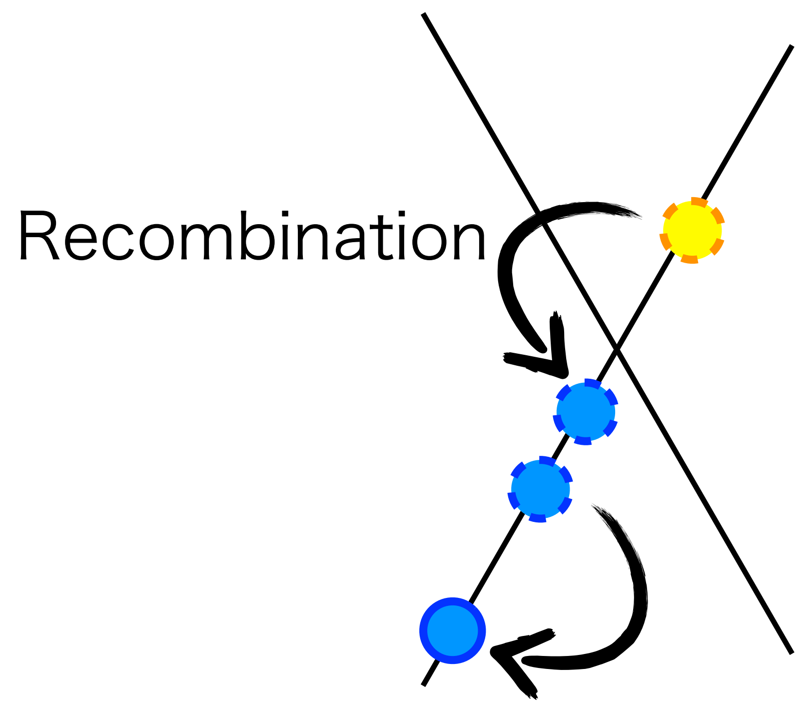
Impact Ionization



h → h + h + e
e → e + e + h

⇒ Carrier multiplication

Auger Recomibination

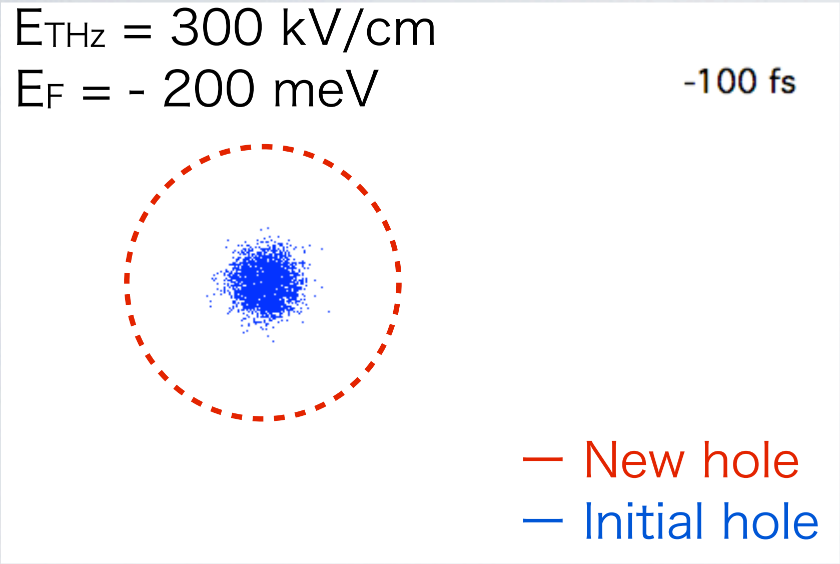


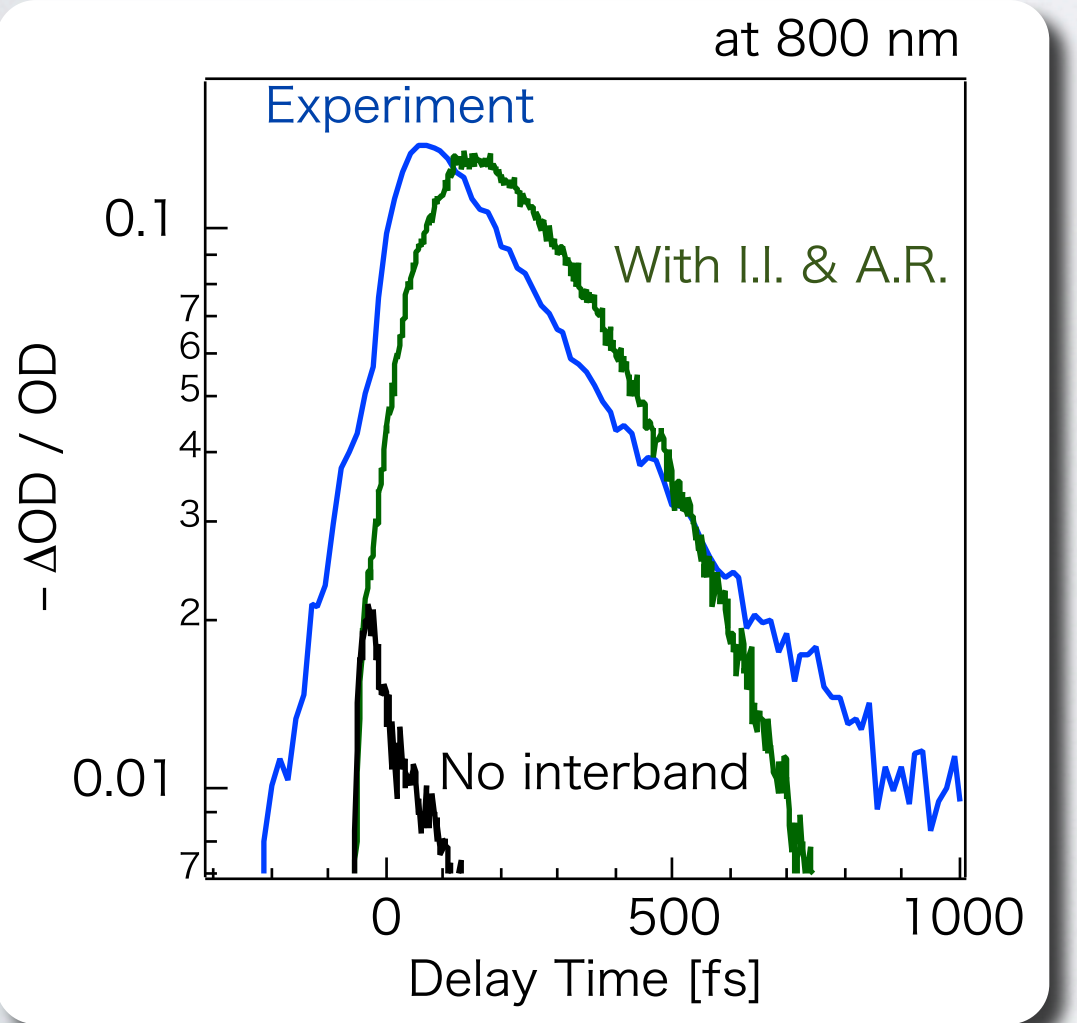
h + h + e → h
e + e + h → e

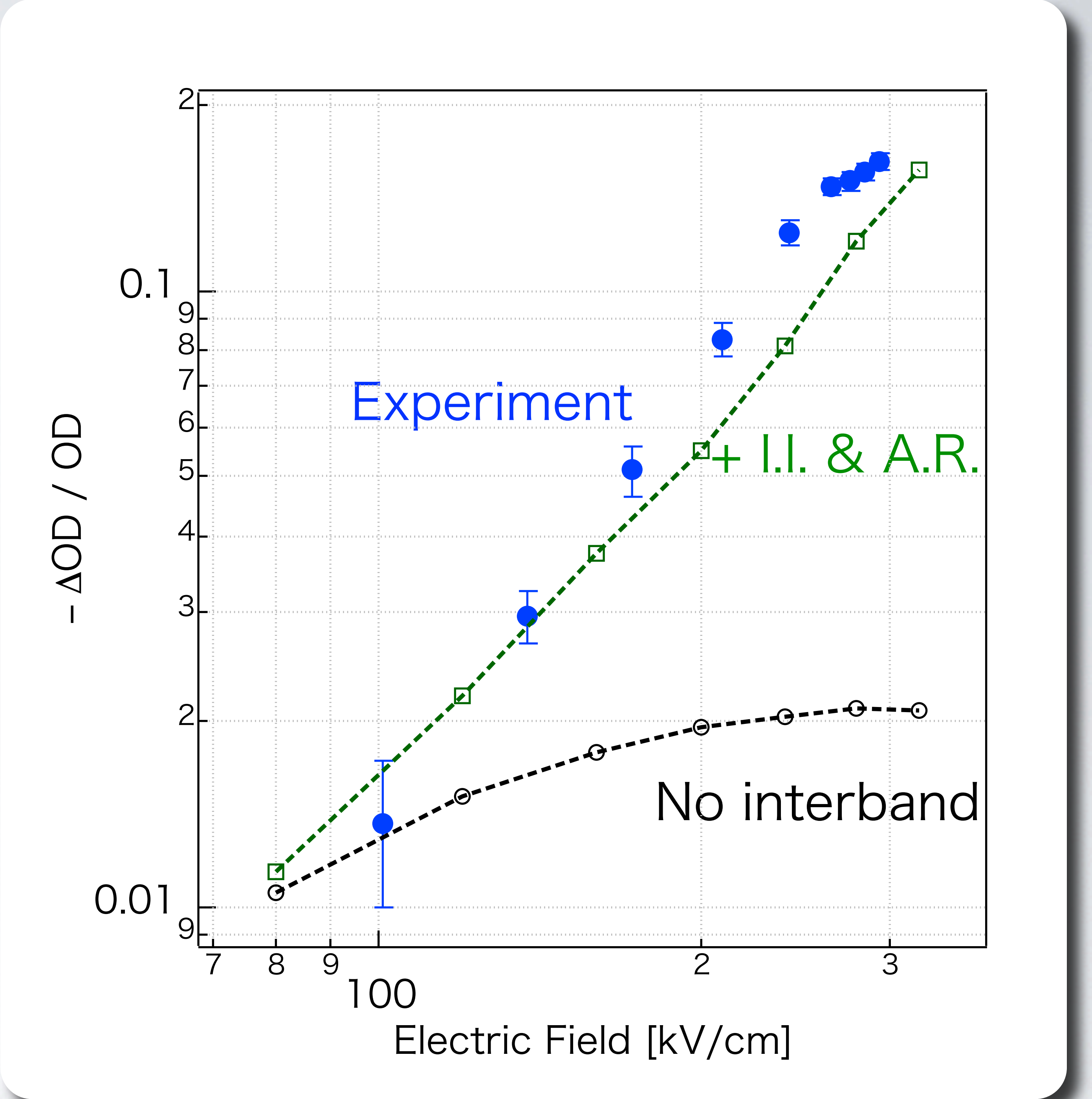
⇒ Carrier annihilation

F.Rana, Phys. Rev. B 76, 155431 (2007)

Simulation With Interband Carrier Scattering

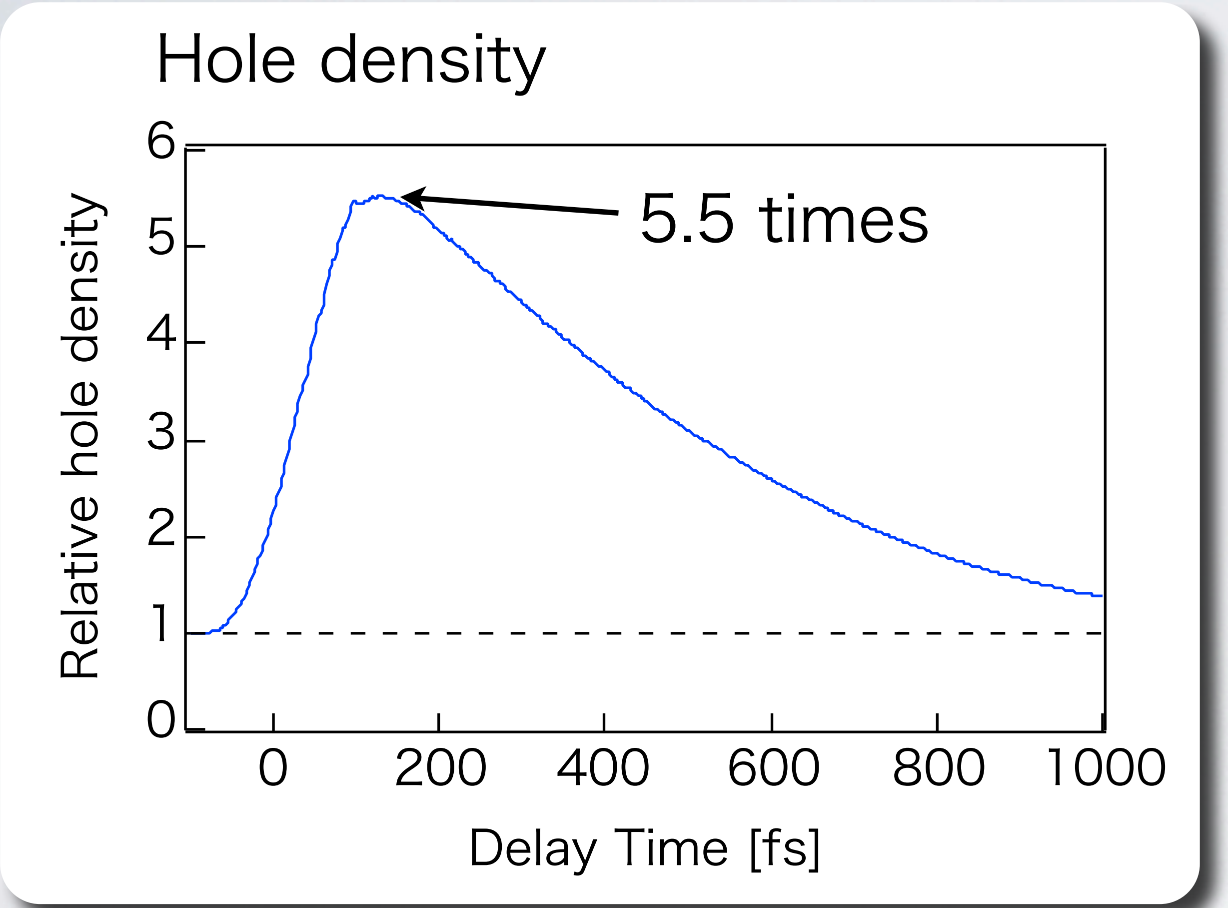


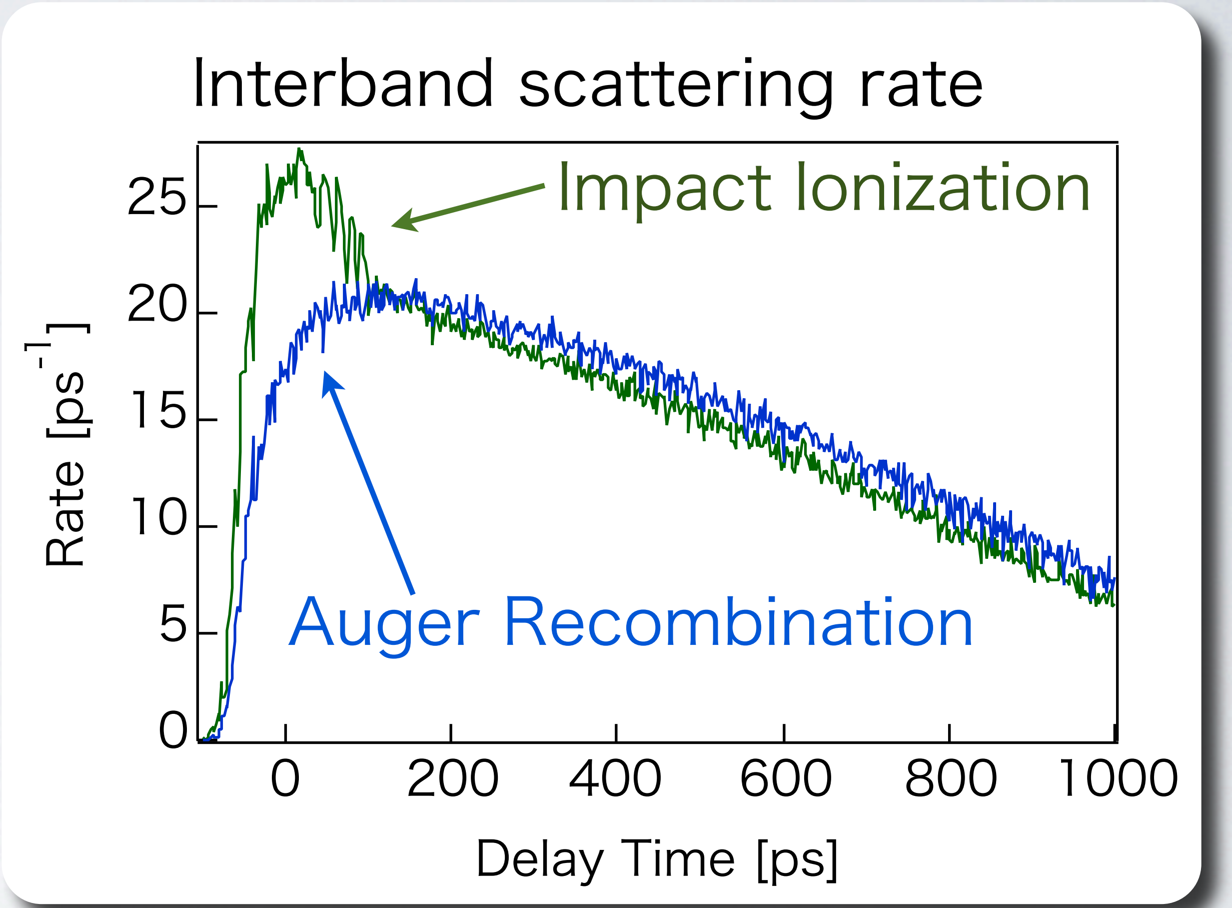




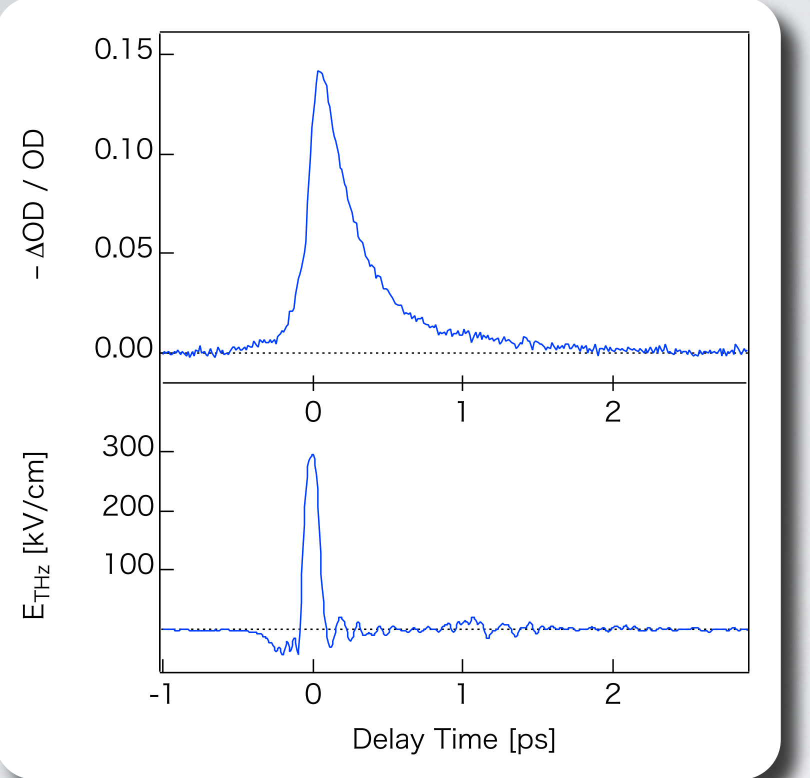
Carrier multiplication is important in high field dynamics.

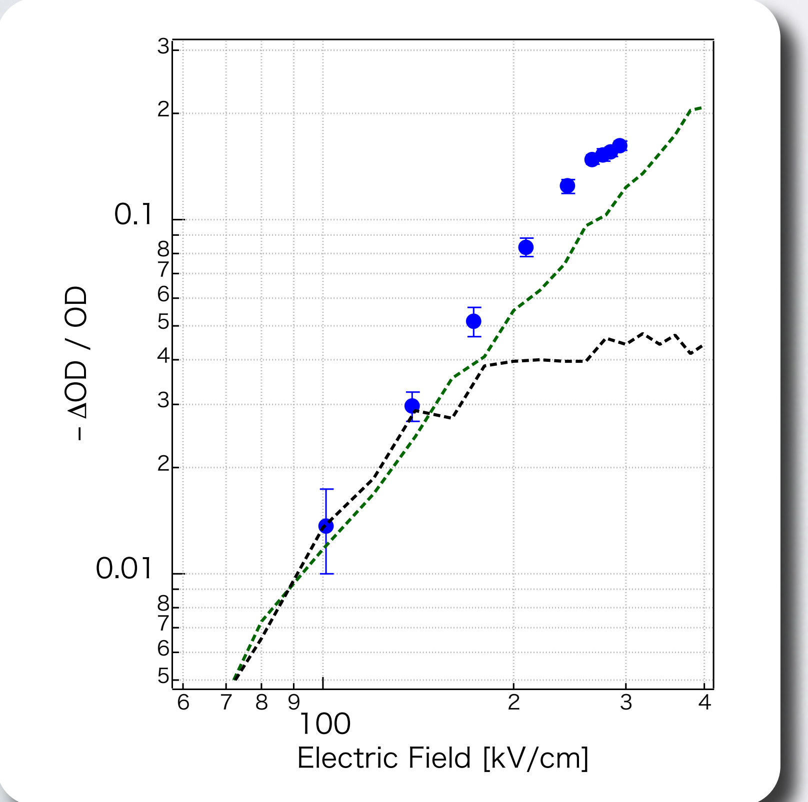
Simulation : Carrier Multiplication





Summary : Thank you for your attention.





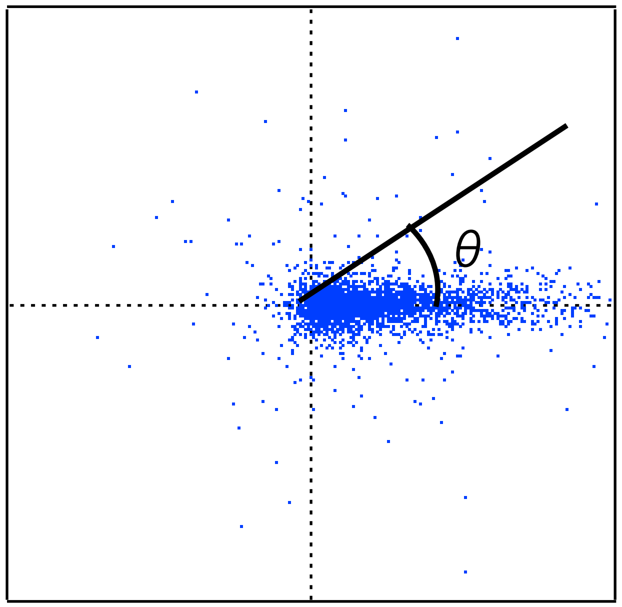
THz-field driven carrier dynamics is investigated with transient absorption measurement in NIR region.

Huge THz induced transparency over 14% is observed at 800 nm.

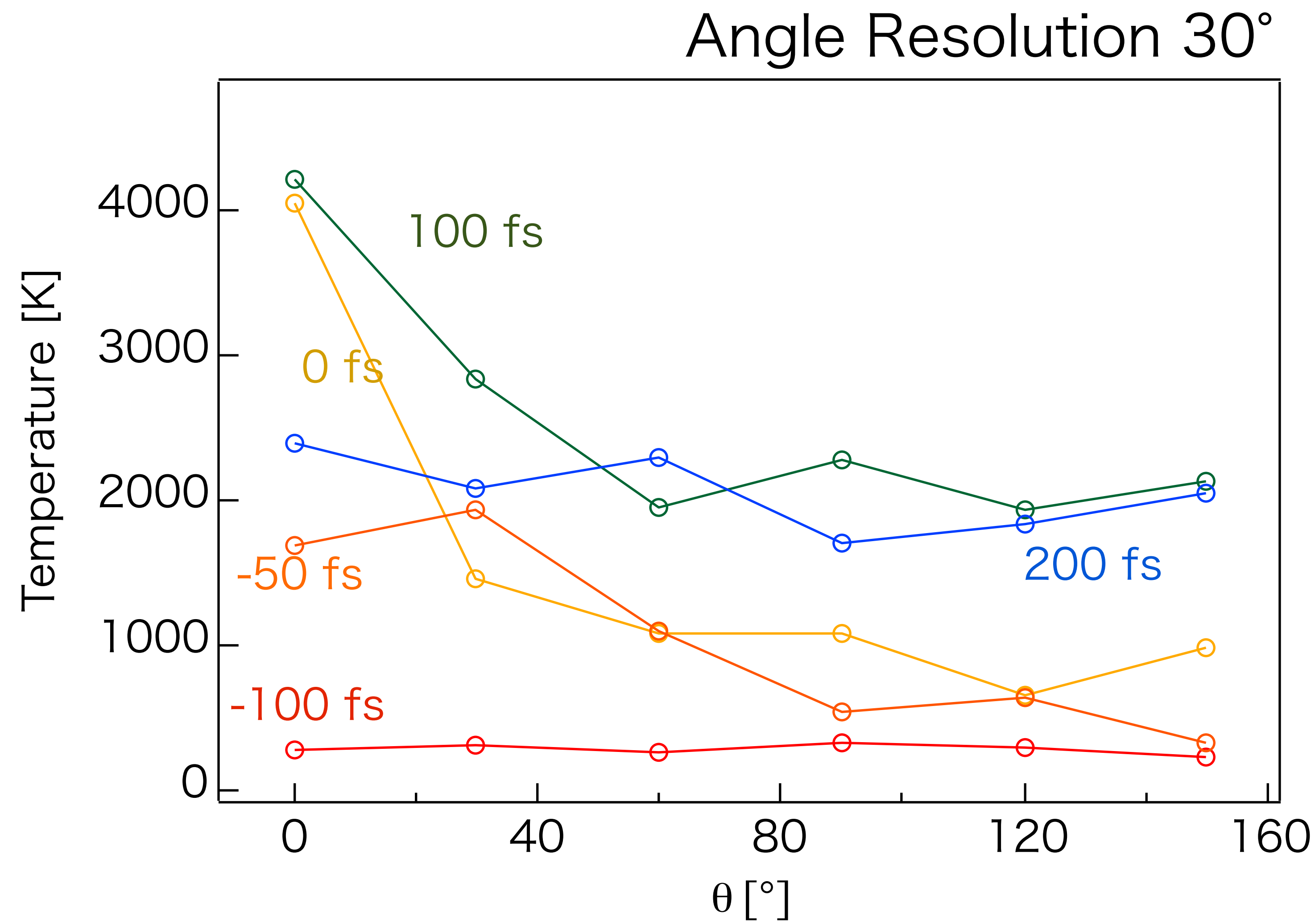
Carrier multiplication with interband carrier scattering is essential in high field dynamics.

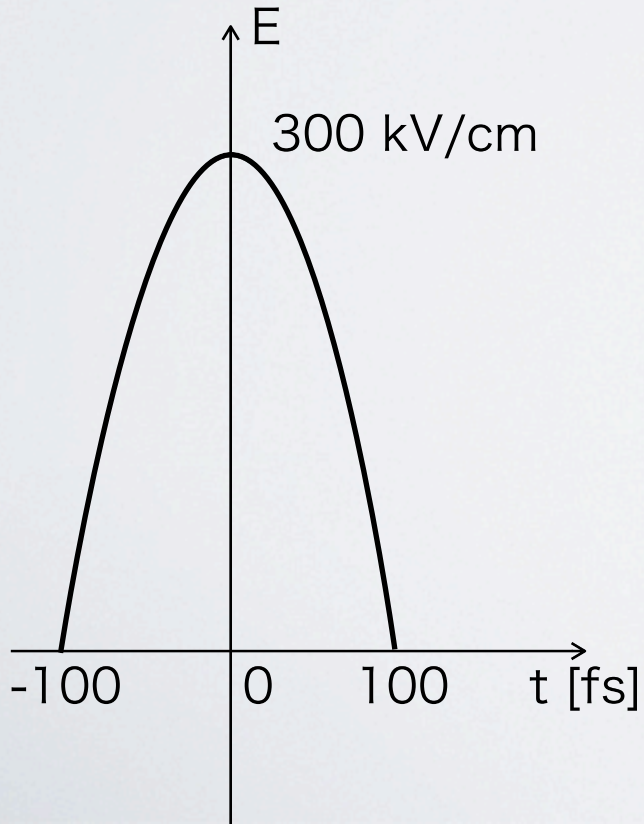
Angle Dependent Temperature

Hole distribution



Angle Dependent Temperature



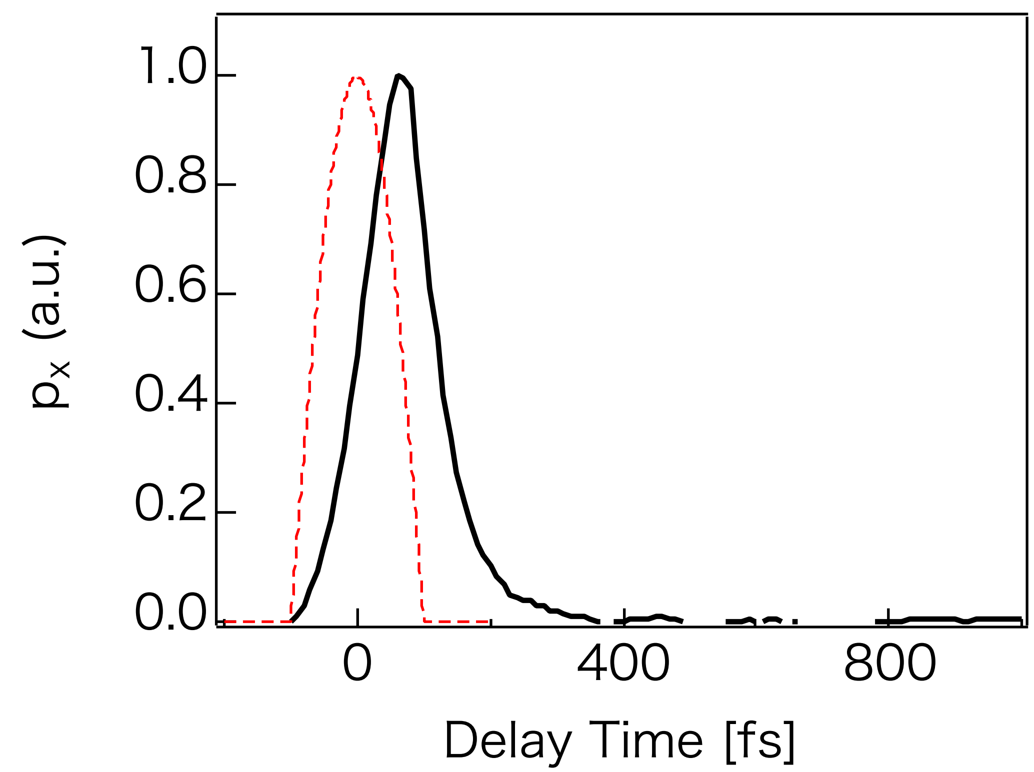


E_{THz} = 300 kV/cm

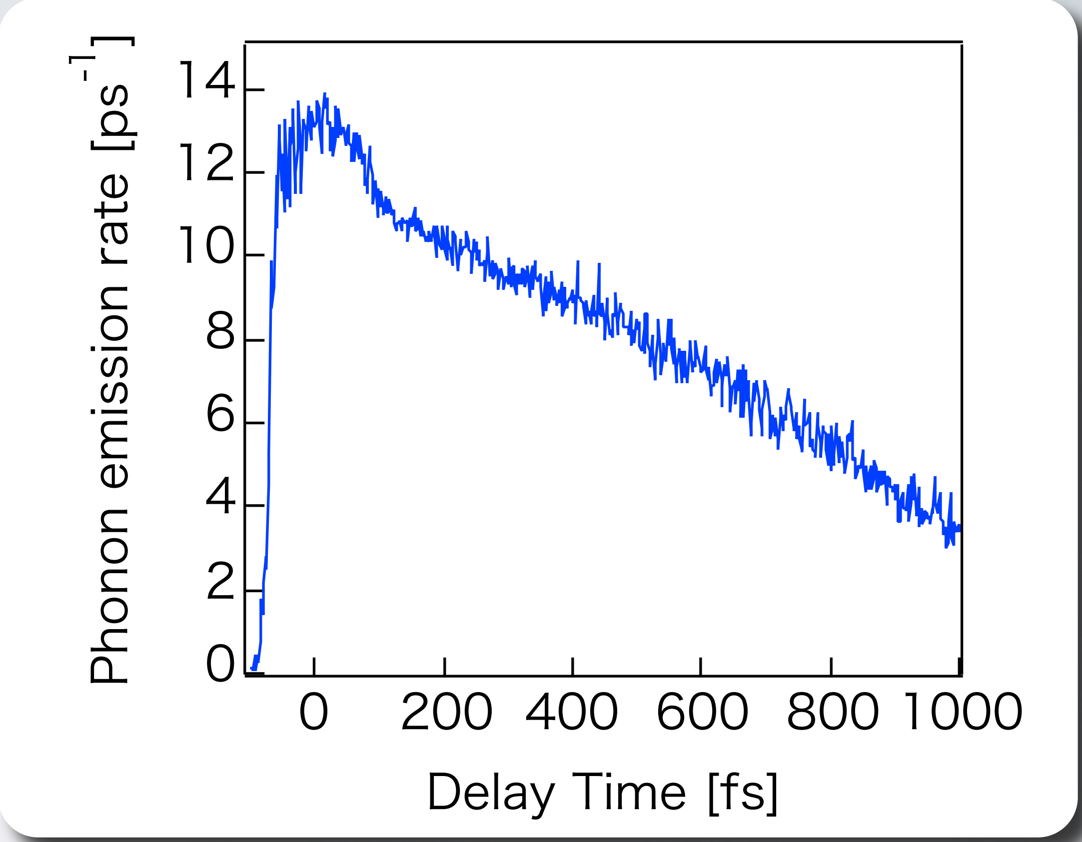
E_F = - 200 meV

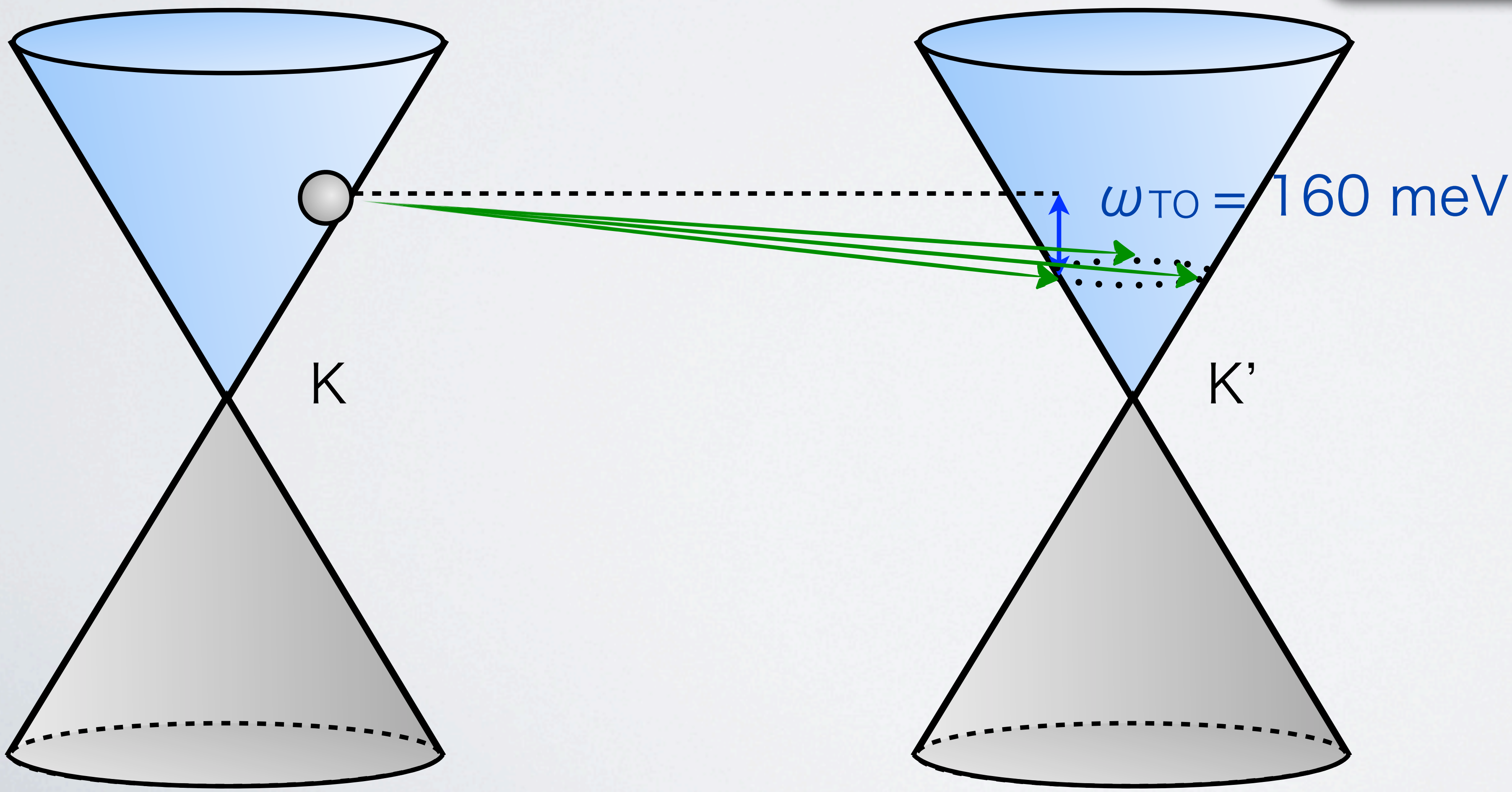
Monte Carlo Simulation: Total Momentum

Hole total momentum in k_x direction



Monte Carlo Simulation : Phonon Scattering





Carrier Scattering