# 高強度テラヘルツパルスで誘起する非線形光学現象

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*物質－細胞統合システム拠点*

# Contents

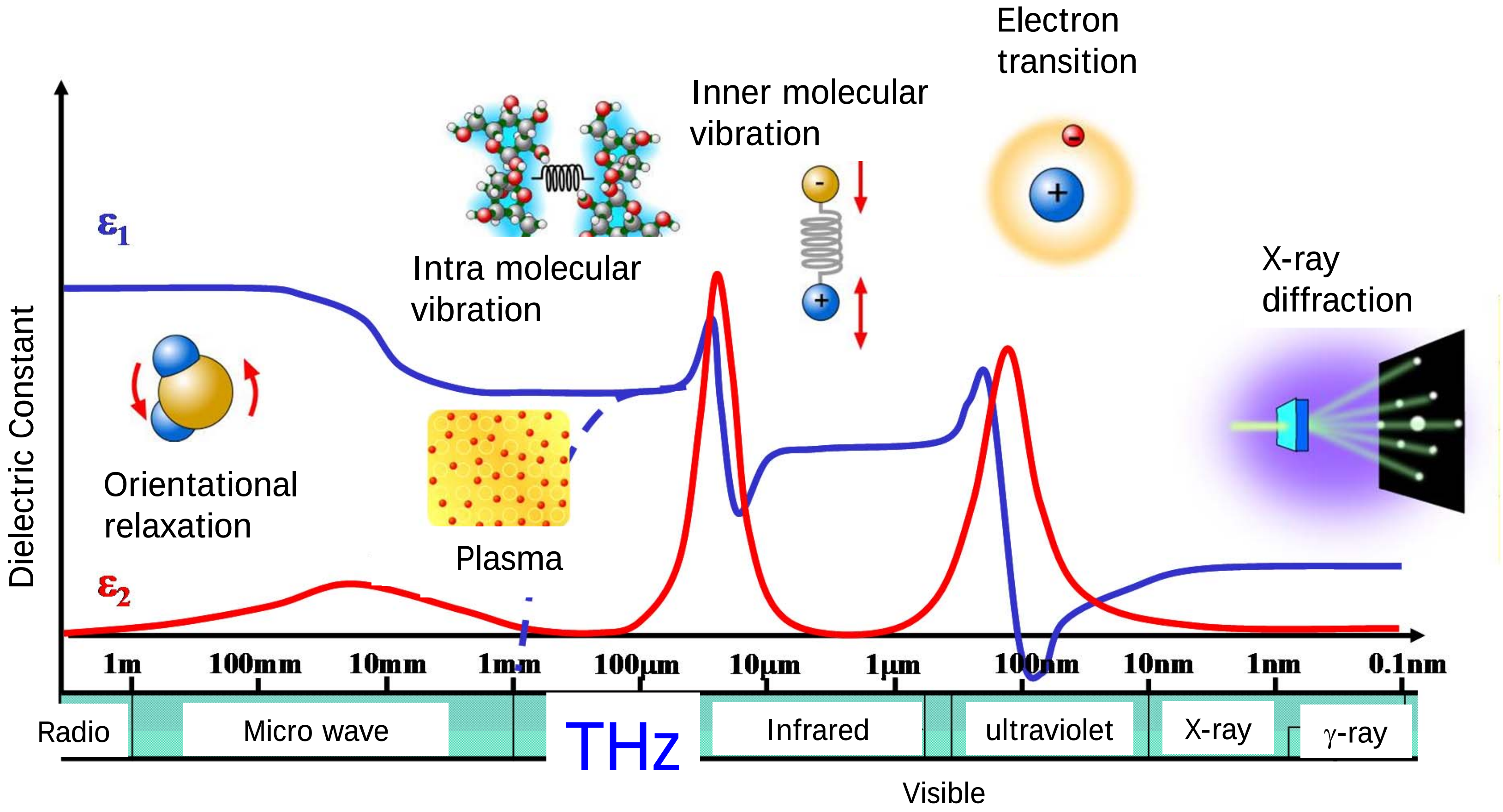
- THz time-domain spectroscopy

- Intense THz pulse generation
    - Tilted–pulse front scheme with a $LiNbO_3$ crystal

      H. Hirori, et al., Appl. Phys. Lett. 98, 091106 (2011)
- Nonlinear THz phenomena
    - Carrier multiplication (GaAs/AlGaAs multiple quantum wells)

      H. Hirori, et al., Nature Comms. 2, 594(2011)

# THz region (0.1-10 THz)
# (1THz ≙ 300μm ≙ 33cm$^{-1}$ ≙ 4.1meV)

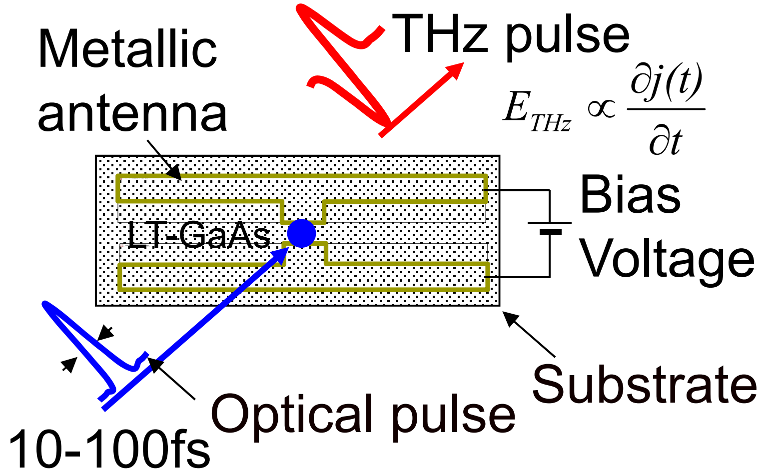


Superconducting gap, Soft phonon modes in ferroelectrics, Excitonic resonance, subband transition, Intra molecular vibration of bio-molecules, Rotational mode of gases, etc.

# THz generation and detection

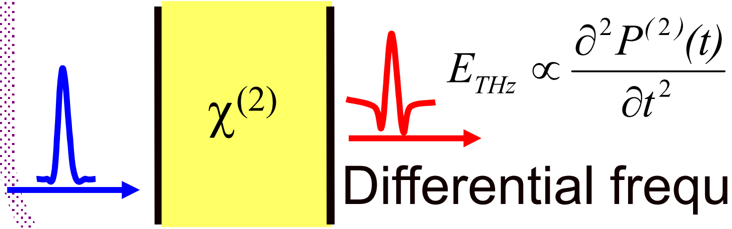
## Generation

### Photoconductive antenna



$$E_{THz} \propto \frac{\partial j(t)}{\partial t}$$

### Nonlinear optical process

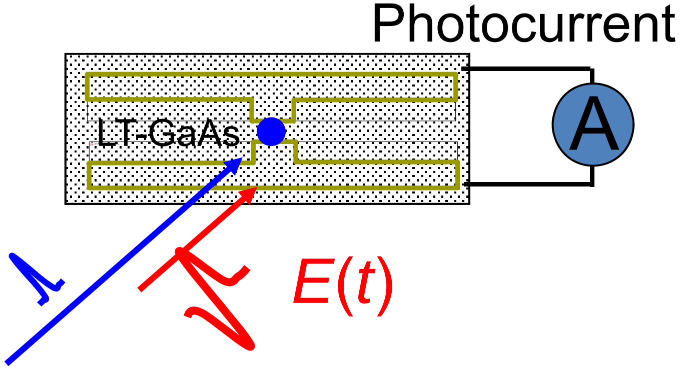


$$E_{THz} \propto \frac{\partial^2 P^{(2)}(t)}{\partial t^2}$$

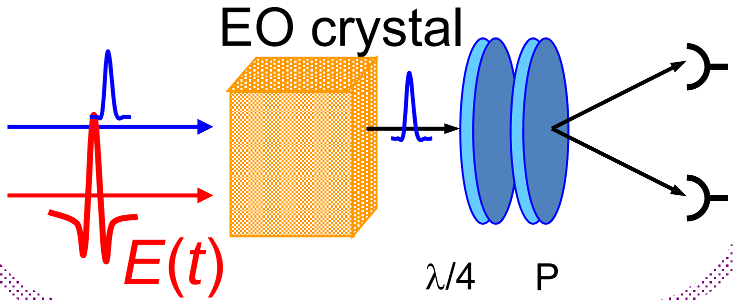
Differential frequency generation

## Detection

### Photoconductive antenna



### Electro optic sampling

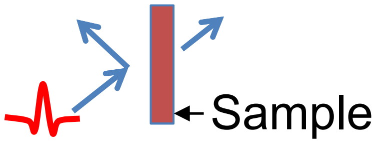


✓0.1-100 THz (3 mm-3 μm, 0.4-400 meV) is available.

# THz Time-domain spectroscopy

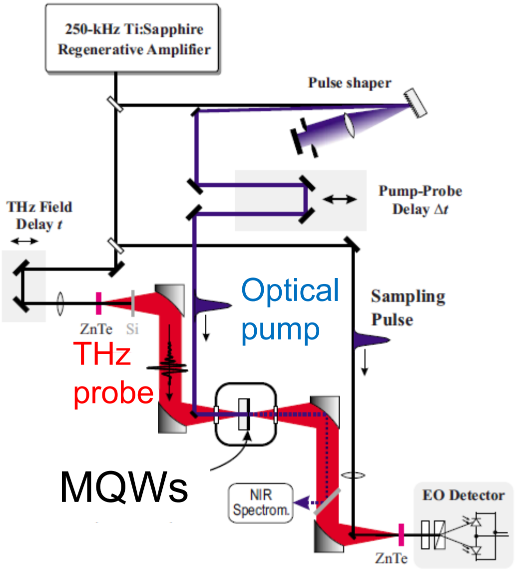
✓Electric field measurement ⇒ Complex dielectric constants.

$E(\omega)$, $\phi(\omega)$

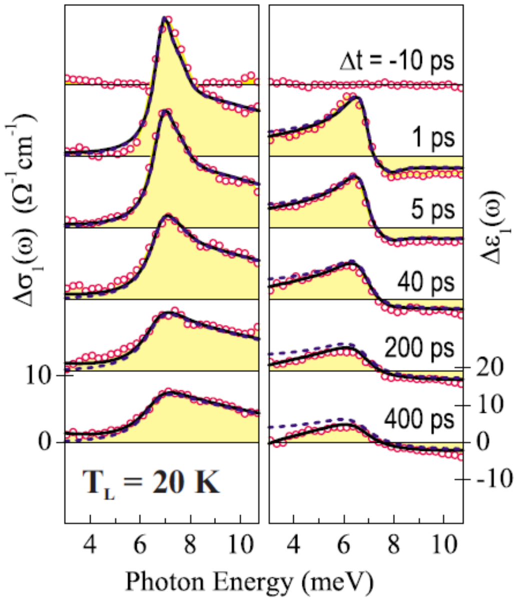


✓Allowing for ultrafast time-resolved measurement.
Ultrafast phenomena in semiconductors, phase-transition and superconducting materials.

R. Huber et al., Nature (2001). C. Kaindl et al., Nature (2003).
T. Suzuki and R. Shimano, PRL(2009).



C. Kaindl et al., PRB(2009).



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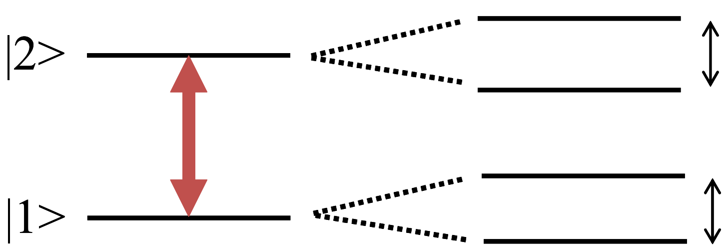
# Nonlinear phenomena in solids with intense THz pulse

- **Resonantly THz-Driven Systems**
  Rabi oscillations, dressed states, the AC (or optical) Stark effect, the Autler-Townes effect, electromagnetically-induced transparency, gain without inversion, …, etc.

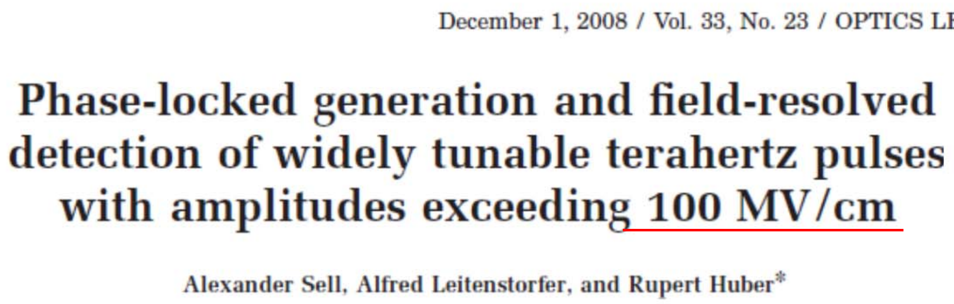


$$\boxed{\hbar\Omega = \left|\hat{\mu}_{12}\right| E}$$

- **Nonlinear transport phenomena**
  Bloch oscillation, Inter-valley scattering (Gunn’s effect), Impact ionizations (carrier multiplication)

- **Available for inducing phase transition phenomena**

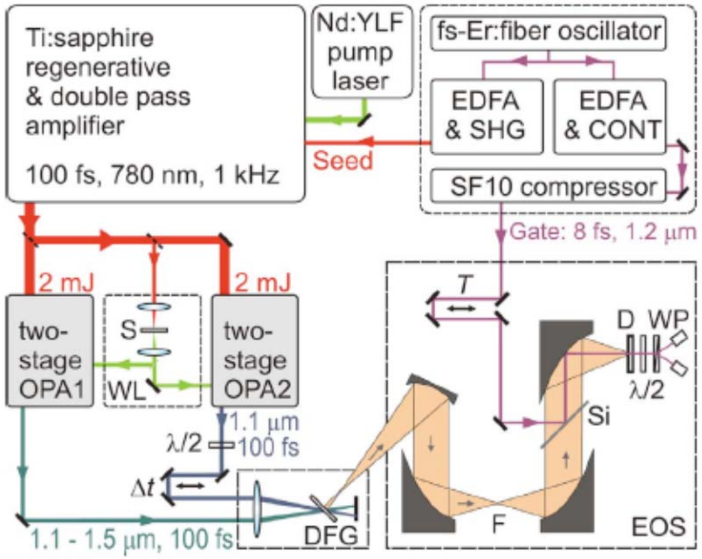
# Intense THz pulse generation (10-80 THz)



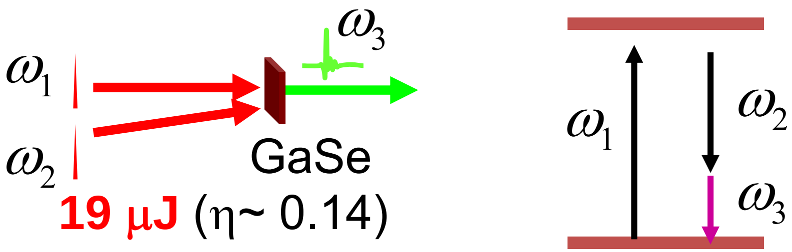
December 1, 2008 / Vol. 33, No. 23 / OPTICS LETTERS 2767

**Phase-locked generation and field-resolved detection of widely tunable terahertz pulses with amplitudes exceeding 100 MV/cm**

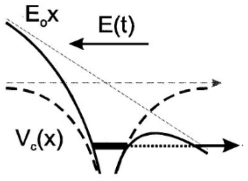
Alexander Sell, Alfred Leitenstorfer, and Rupert Huber*



Differential frequency generation



**19 µJ** (η~ 0.14)



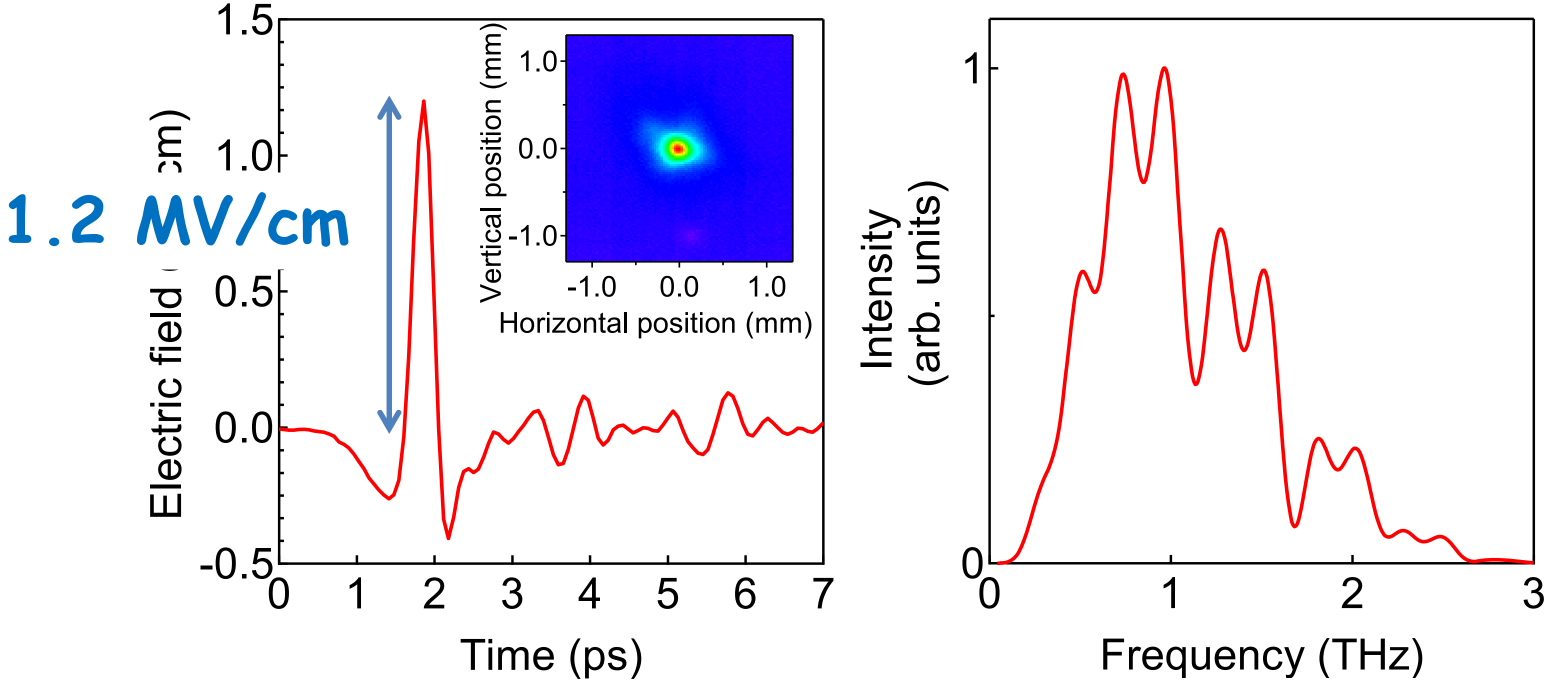
- Field strengths that outer-shell electrons in atoms experiences.
- Higher frequency region (10-80 THz) has a GOOD source! (19 µJ, $\eta \sim 0.14$).
- Lower frequency ???

$e/4\pi\varepsilon_0 a_B^2$ ~ 100 M/cm-1GV/cm

# Tilted-pulse front scheme with $LiNbO_3$ crystal (below 3 THz)

## Temporal profile and spectra

**H. Hirori, et al., APL (2011)**



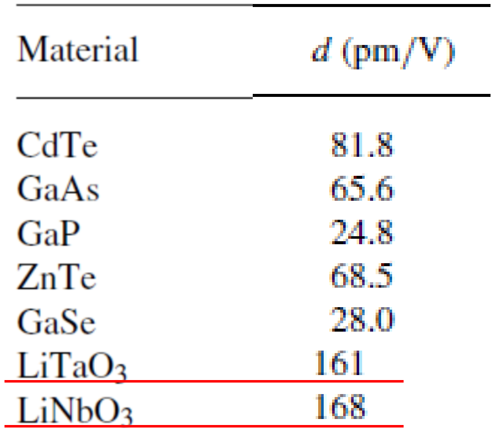
- Nearly half-cycle sub-picosecond pulse with a maximum peak field of 1.2 MV/cm (3 μJ, $\eta \sim 10^{-3}$, spot diameter ~ 300 μm).
- Around 1 THz of center frequency.
- Optical rectification of femtosecond lasers.

# Characteristic of $LiNbO_3$ for THz generation

## Good point

- High second order nonlinear susceptibility
- Large band-gap energy (3.7 eV) (less multi-photon absorption)

Appl. Phys. B 78, 593 (2004)



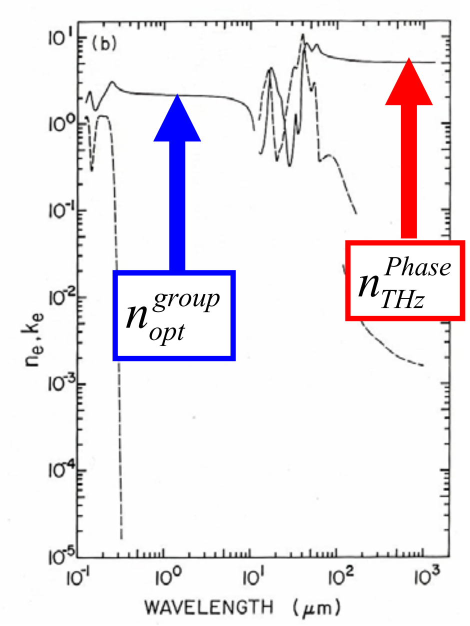
| Material | $d$ (pm/V) |
|---|---|
| CdTe | 81.8 |
| GaAs | 65.6 |
| GaP | 24.8 |
| ZnTe | 68.5 |
| GaSe | 28.0 |
| $LiTaO_3$ | 161 |
| $LiNbO_3$ | 168 |

## Bad point

- Large difference between $n_{THz}^{phase}(=5)$ and $n_{opt}^{group}(=2.2)$

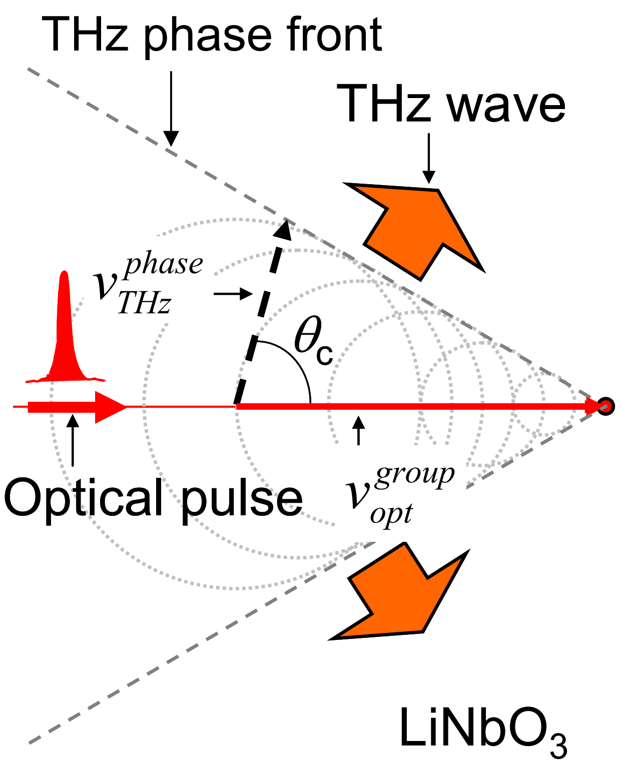
➡ $v_{THz}^{phase} << v_{opt}^{group}$

Velocity (phase) mismatching



D. Palik, Academic Press (1985).

# THz Cherenkov wave



D. H. Auston et al.,
PRL **53**, 1555 (1984).

$$v_{THz}^{phase} << v_{opt}^{group}$$

➡ The condition necessary for the shock (or Cherenkov) wave radiation of a supersonic aircraft or bullet.

## Direction of THz wave

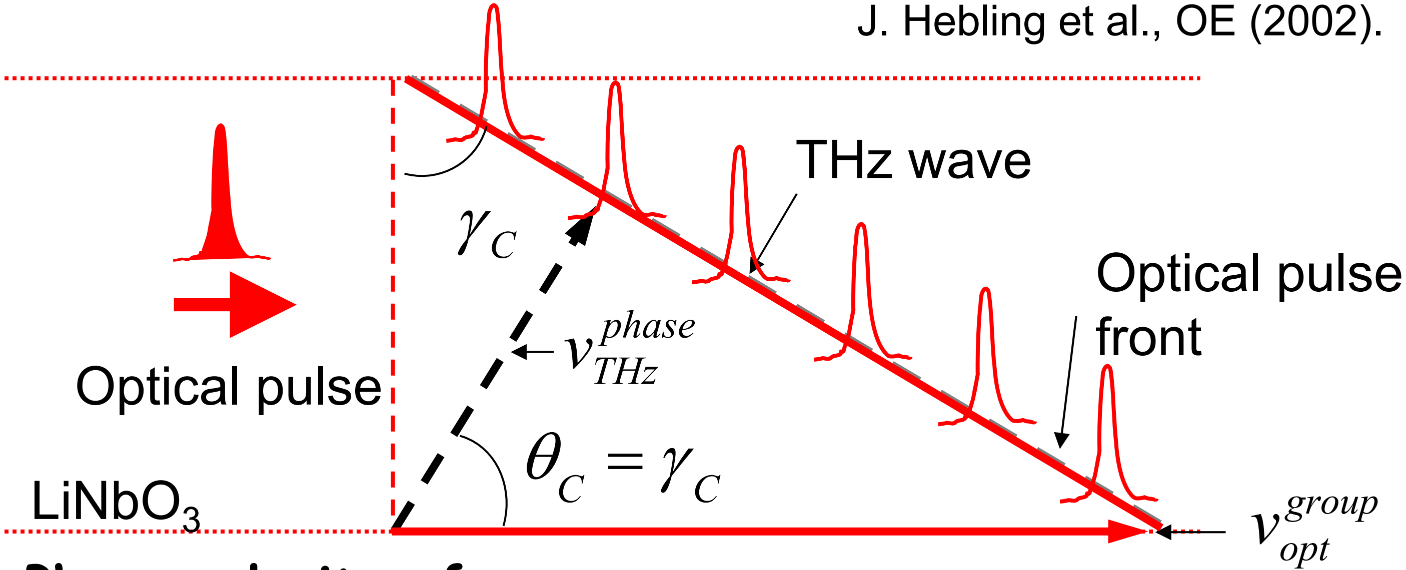
$$v_{THz}^{phase} = v_{opt}^{group} \cos\theta_C$$

$$\cos\theta_C = n_{opt}^{group} \Big/ n_{THz}^{phase}$$

➡ $\theta_C = 62 \text{ deg}$

# Velocity matching by the tilted-pulse-front scheme

J. Hebling et al., OE (2002).



**Phase velocity of THz wave**

$$v_{THz}^{phase} = v_{opt}^{group} \cos\theta_C$$

**Noncollinear velocity of tilted optical pulse**

$$v_{opt}^{\gamma_C} = v_{opt}^{group} \cos\gamma_C$$

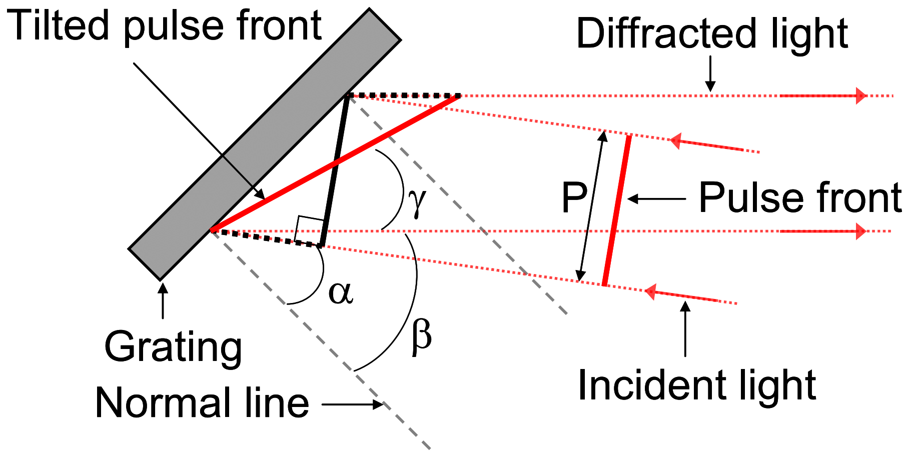
**When** $\gamma_C = \theta_C$,

$$\Rightarrow v_{THz}^{phase} = v_{opt}^{\gamma_C}$$

Phase matching condition can be satisfied.

# How to tilt the pump pulse front?

## Grating

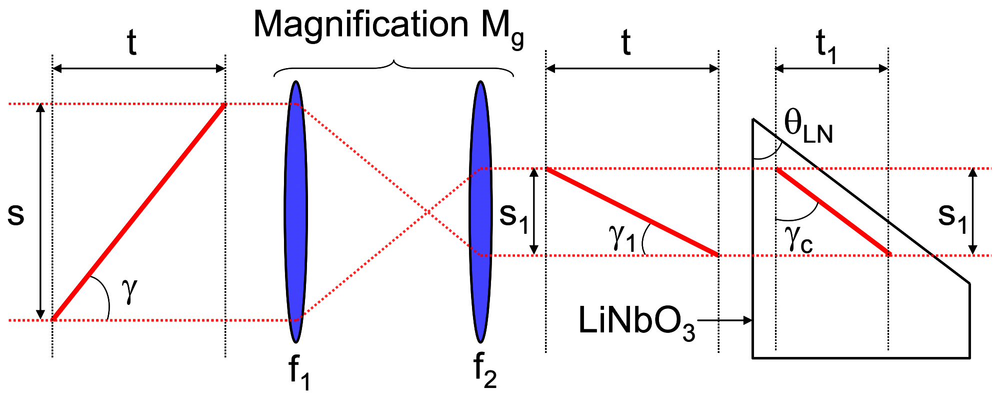


$$\tan\gamma = \frac{\cos\beta}{\sin\alpha + \sin\beta}.$$

$$\sin\alpha + \sin\beta = mp\lambda_0,$$

$m$: diffraction order
$p$: groove density of a grating
$\lambda_0$: the central wavelength

## Lens pair

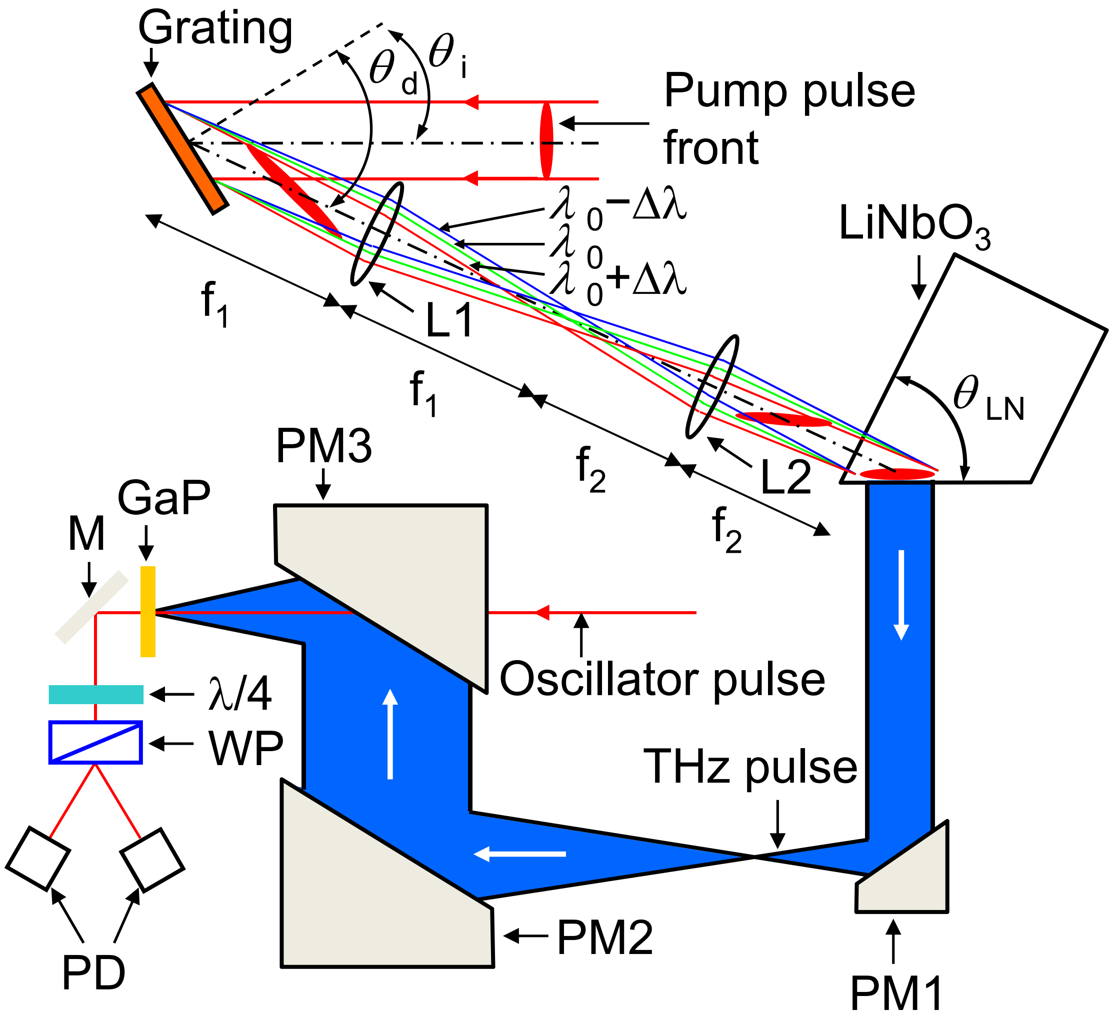


$$\tan\gamma_c = \frac{m\lambda_0 p}{n_g M_g \cos\beta}.$$

$n_g$: group refractive index of LN crystal

$$\gamma_C = \theta_C = 62°$$

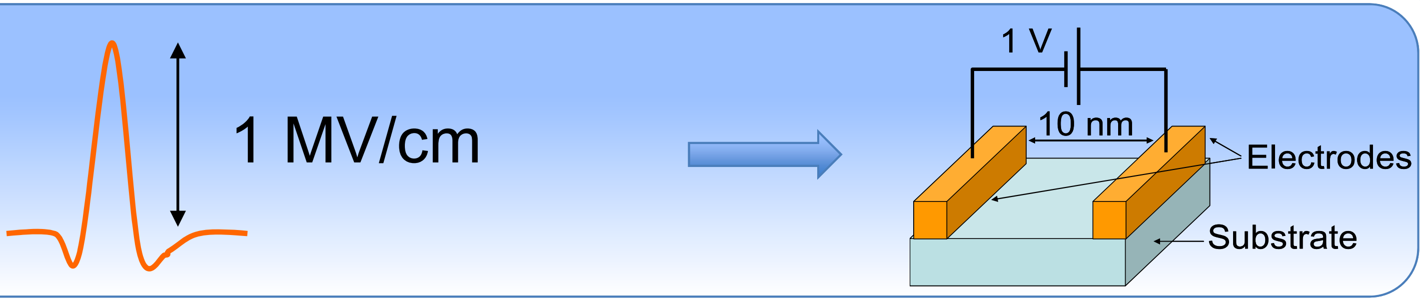
# THz pulse generation setup



Laser source
- Ti: sapphire
- 4 mJ/pulse
- 1 kHz
- 780 nm
- 85 fs (FWHM)

Key factors
- Good phase matching condition
- High nonlinear susceptibility
- High pumping power
- Tight focusing the collimated THz beam

# Strong picosecond DC electric field



- Achieved in actual electric devices
- Useful for characterization of electric devices

Nonlinear transport phenomena

- Bloch oscillations
- Inter-valley scattering
- Impact ionization
- Zener tunneling

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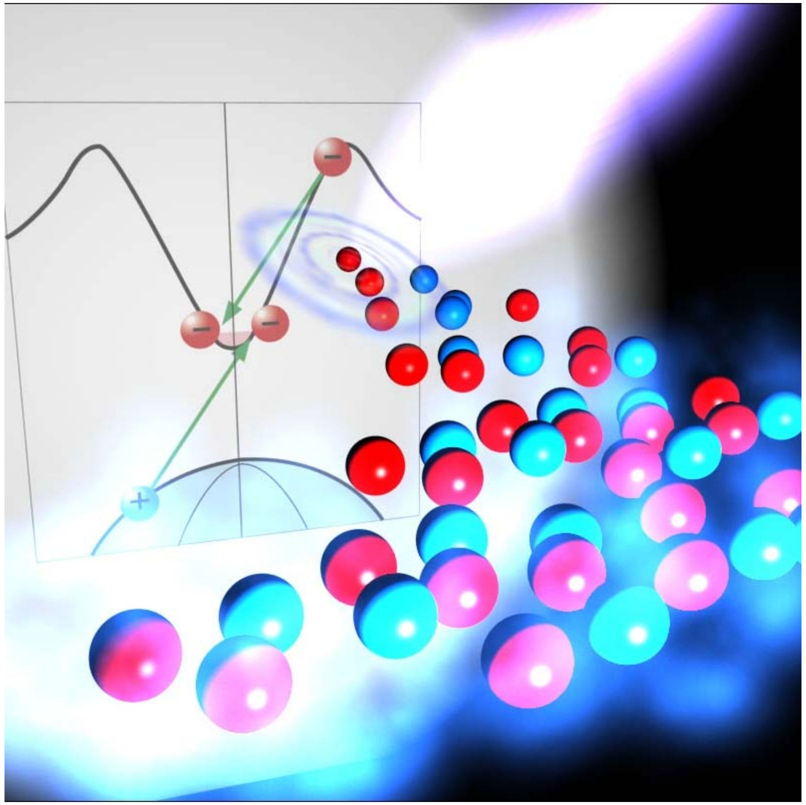
    H. Hirori, et al., Nature Comms. 2, 594(2011)

# Extraordinary carrier multiplication in GaAs MQWs gated by intense terahertz pulse



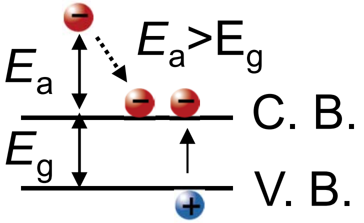
H. Hirori, et al., Nature Commun. 2, 594(2011)

# Nonlinear Transport Phenomena in Semiconductors

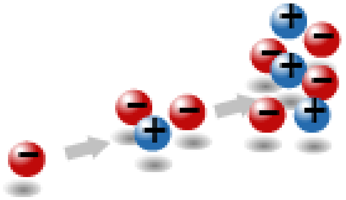
## Carrier multiplication in high electric field

- Electron accelerated by electric field can gain a kinetic energy and excite other electrons.

### Impact ionization

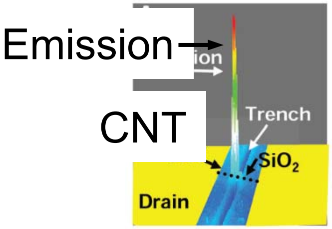


### Carrier multiplication



## Various applications

- Avalanche photodiodes
- Electroluminescent and photovoltaic nano-devices



J. Chen et al., Science (2005).

## Fundamental

- Important for nonlinear transport phenomena.

➡ The elementary scattering process has been unclear.

# Purpose

Clarifying carrier multiplication process of GaAs under high electric field

- THZ pulse with an amplitude exceeding 1 MV/cm

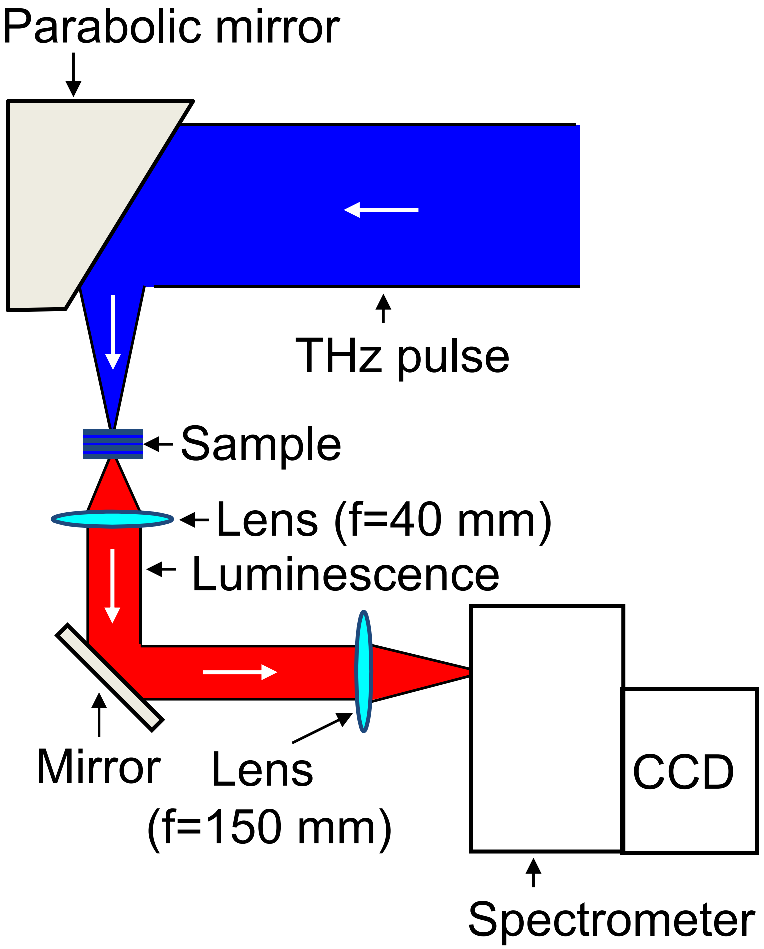
➡ Sufficient carrier multiplication.

- Luminescence measurement

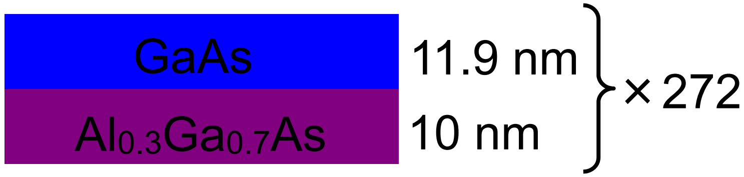
➡ Direct evidence of carrier generation.

# Experimental setup for THz induced luminescence

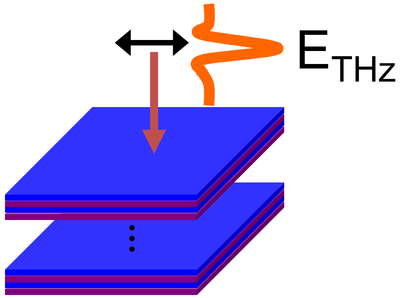
## Setup



## GaAs quantum wells

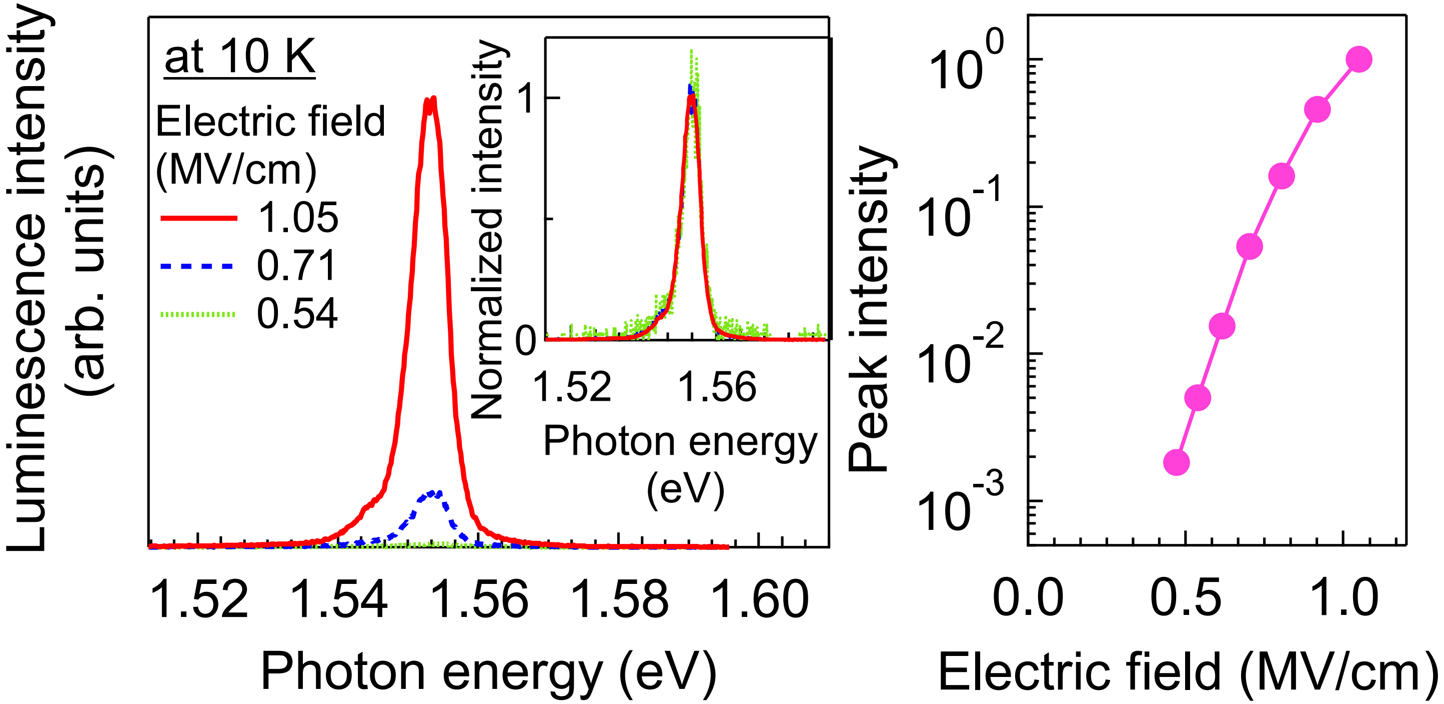


- Nominally non-doped (Residual dominant impurity donors are Sulfur.)
- Low temperature measurement (10 -150 K)
- The polarization of electric field is in plane and along the (100) direction of the sample



# THz induced luminescence without photoexcitation

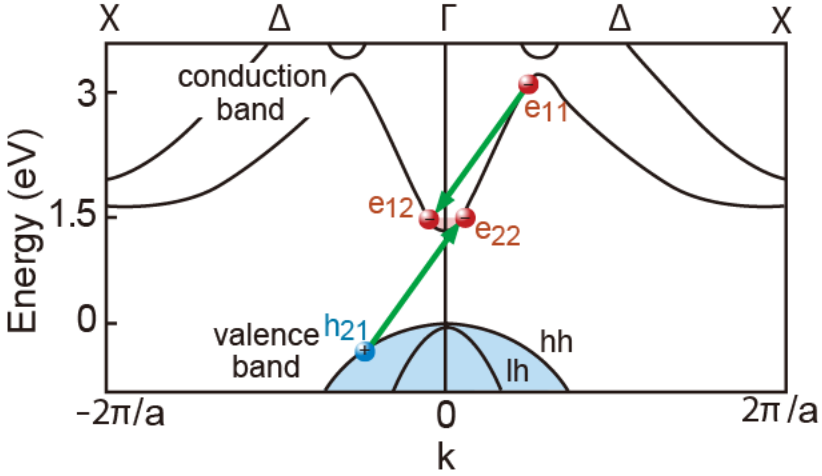
## Electric field dependence



- Luminescence centered around 1.55 eV.
- Electric field dependence shows extremely nonlinear ($\propto \varepsilon^{8}$).
- The number of carriers increases by about three orders of magnitude.

# Carrier multiplication process in GaAs

## Single impact ionization



- Electrons seeded from residual impurity donors by the field ionization
- Doubling the number of electrons.

$$e_{11} \to e_{12} + e_{22} + h_{21}$$

$e_{ij}$, $h_{ij}$ : electron and hole

## Carrier multiplication

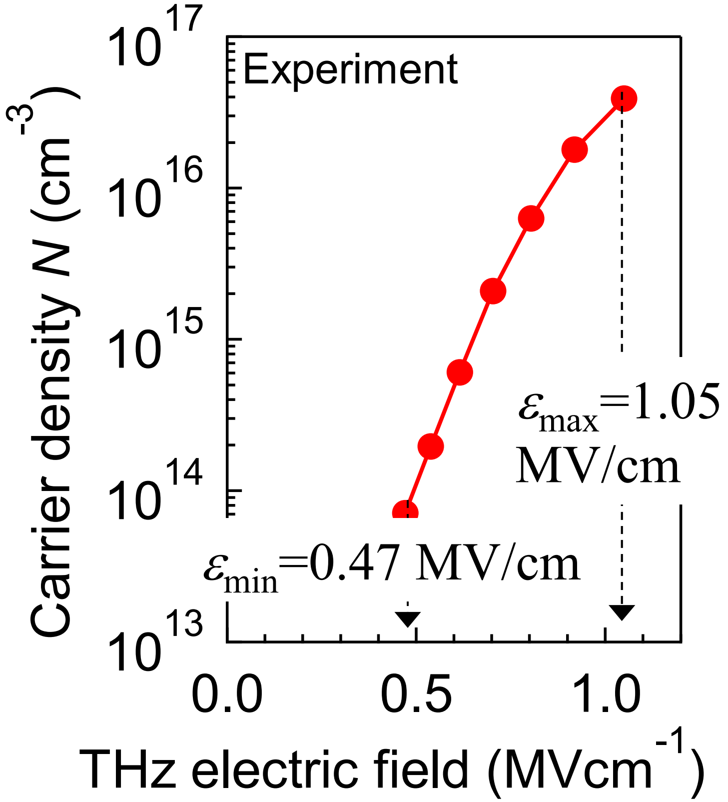
Carrier density $N(\varepsilon)$ after $< n_{\mathrm{I}} >$ times impact ionization events:

$$N(\varepsilon) = N_0 \times 2^{\langle n_I \rangle}$$

$N_0$ : initial electron density
$< n_{\mathrm{I}} >$ : number of impact ionization events

# Impact ionization number derived from experimental result

## Carrier density

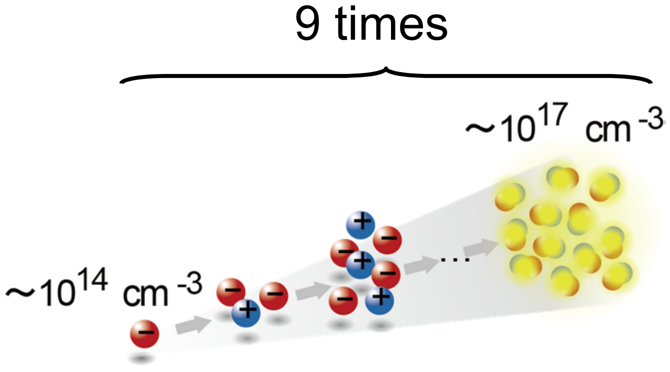


## Increment of impact ionization number $< \Delta n_I >$

$$N(\varepsilon) = N_0 \times 2^{\langle n_I \rangle}$$

$$< \Delta n_I > = \log_2(N(\varepsilon_{max})/N(\varepsilon_{min})) \sim 9$$

- 9 impact ionizations induce numerous *e*-*h* pairs.



# How many times do impact ionization occur?

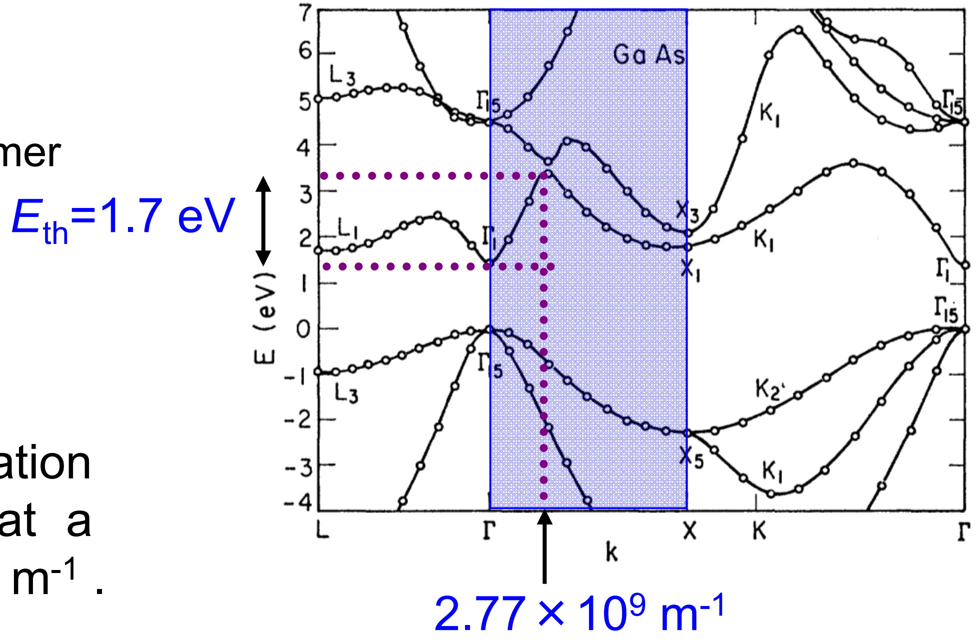
## Changing $k$ with electric field $\varepsilon$ ($t$)

$$\hbar \frac{dk(t)}{dt} = -e\varepsilon(t)$$

$k(t)$ : average electron wavenumer
$\varepsilon(t)$ : electric field
$e$ : elementary charge
$\hbar$ : Planck's constant

The GaAs dispersion relation yields the $E_{th}$ of 1.7 eV at a wavenumber $k$ of $2.77 \times 10^9$ m$^{-1}$ .

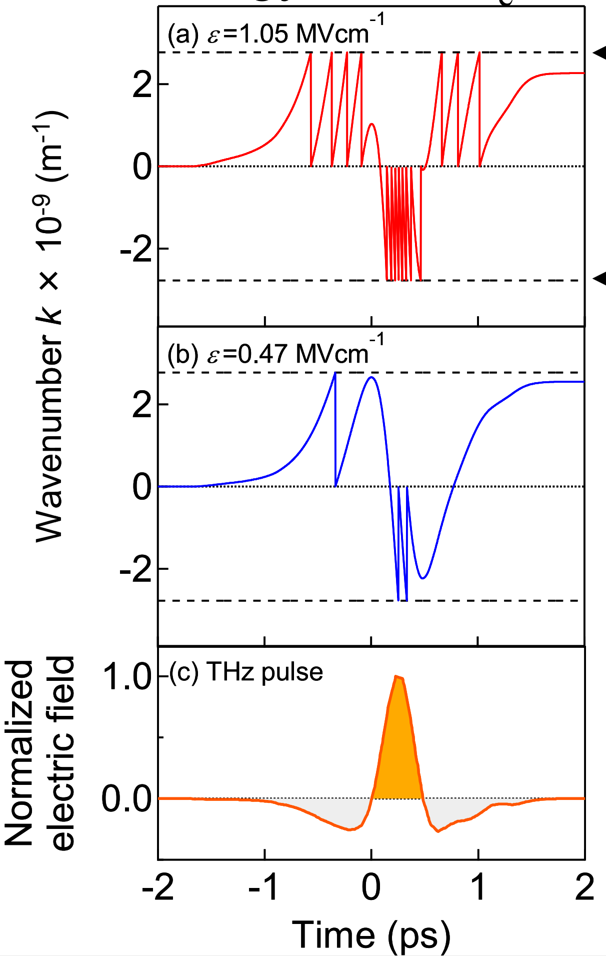
M. L. Cohen et al. *Phys*. *Rev*. **141**, 789 (1966).



The number of times the $k$ exceeds $2.77 \times 10^9$ m$^{-1}$ in electric field of THz pulse should be calculated.

# Calculated number of impact ionization events

- Assumption that the accelerated electrons lose all kinetic energy when $K_e$ achieve the $E_{th}$.



← $k = 2.77 \times 10^{9}$ m$^{-1}$

$< n_I(\varepsilon_{max}=1.05 \text{ MV/cm}) > = 15$

← $k = -2.77 \times 10^{9}$ m$^{-1}$

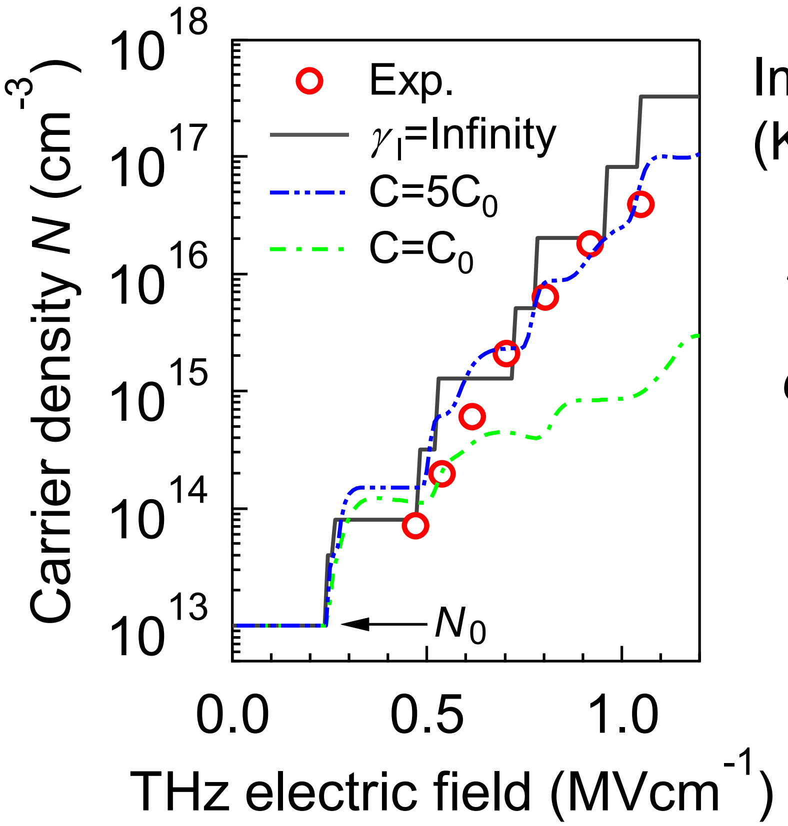
$< n_I(\varepsilon_{min}=0.47 \text{ MV/cm}) > = 3$

Increment number of impact ionization events

$$\Rightarrow < n_I(\varepsilon_{max}) > - < n_I(\varepsilon_{min}) > = 12$$

Good agreement with the experimental value (9 times).

# Experiment v.s. calculation



Impact ionization rate (Keldysh formula):

$$\gamma_I = C(E - E_{th})^2, \quad E > E_{th}.$$

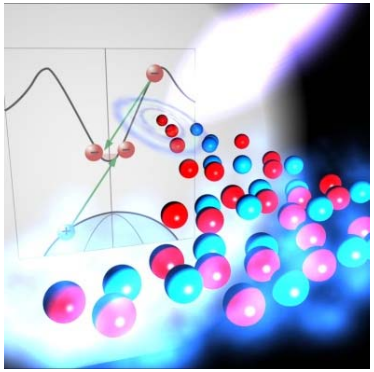
$C = C_0 = 870$ ps$^{-1}$eV$^{-2}$ and $E_{th} = 1.7$ eV.

M. V. Fischetti and S. E. Laux, *Phys. Rev. B* **38**, 9721 (1988).

Good agreement of impact ionization numbers and carrier densities between the experimental and calculated results.

# Summary

## Result



✓ Generating the world’s strongest THz pulse.
(Achieving at electric field of 1 MV/cm.)

✓ Observation of bright luminescence by irradiating 1-MV/cm THz pulse to GaAs MQWs ($E_g \sim 400\ h\nu_{\mathrm{THz}}$).

## Outlook

✓ Further increasing up the THz generation efficiency.

✓ Studying Zener tunneling and Bloch oscillations with higher electric field.

✓ Appling new materials showing phase transitions.