2007/ 7/ 9　ERL研究会「コンパクトERLが拓く世界」
KEK, Tsukuba

テラヘルツ時間領域分光法と基礎科学への応用

Department of Physics, Kyoto University

Koichiro Tanaka

SCOPE project
2003-2007

ATTOC

Advanced Terahertz Technologies adapted for Optical Communication

KYOTO UNIVERSITY
KYOTO JAPAN
FOUNDED 1897

JSPS project
2006-2010

Exploitation of Organic Electronic Materials of Potential Dynamic Switches for Non-equilibrium Condensed Matter Sciences

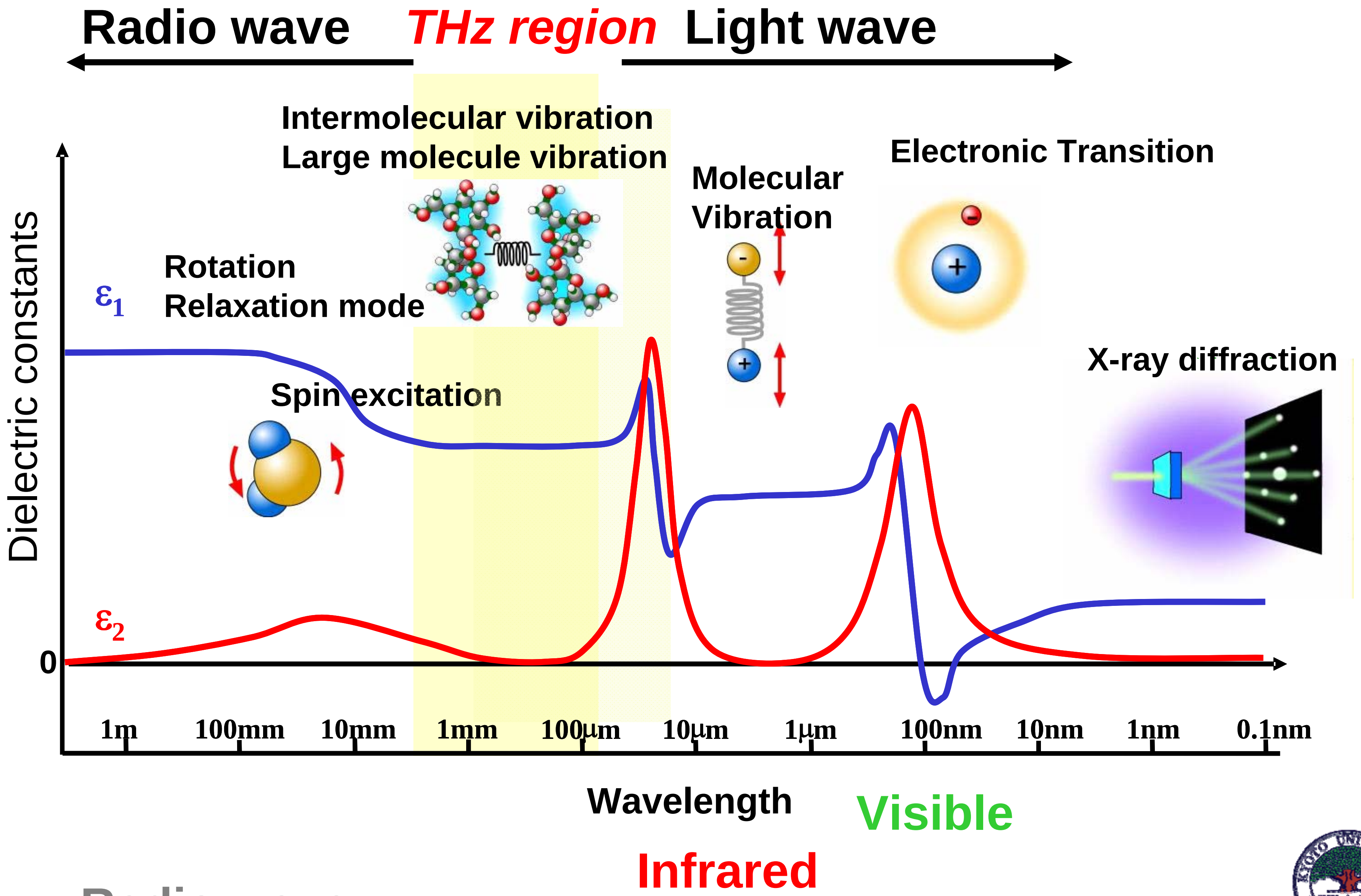
講演内容

- テラヘルツテクノロジーとは何か？
- 時間領域テラヘルツ分光法の基礎
- 時間領域テラヘルツ分光法による物性測定

  有機材料，強誘電体、半導体、超伝導体

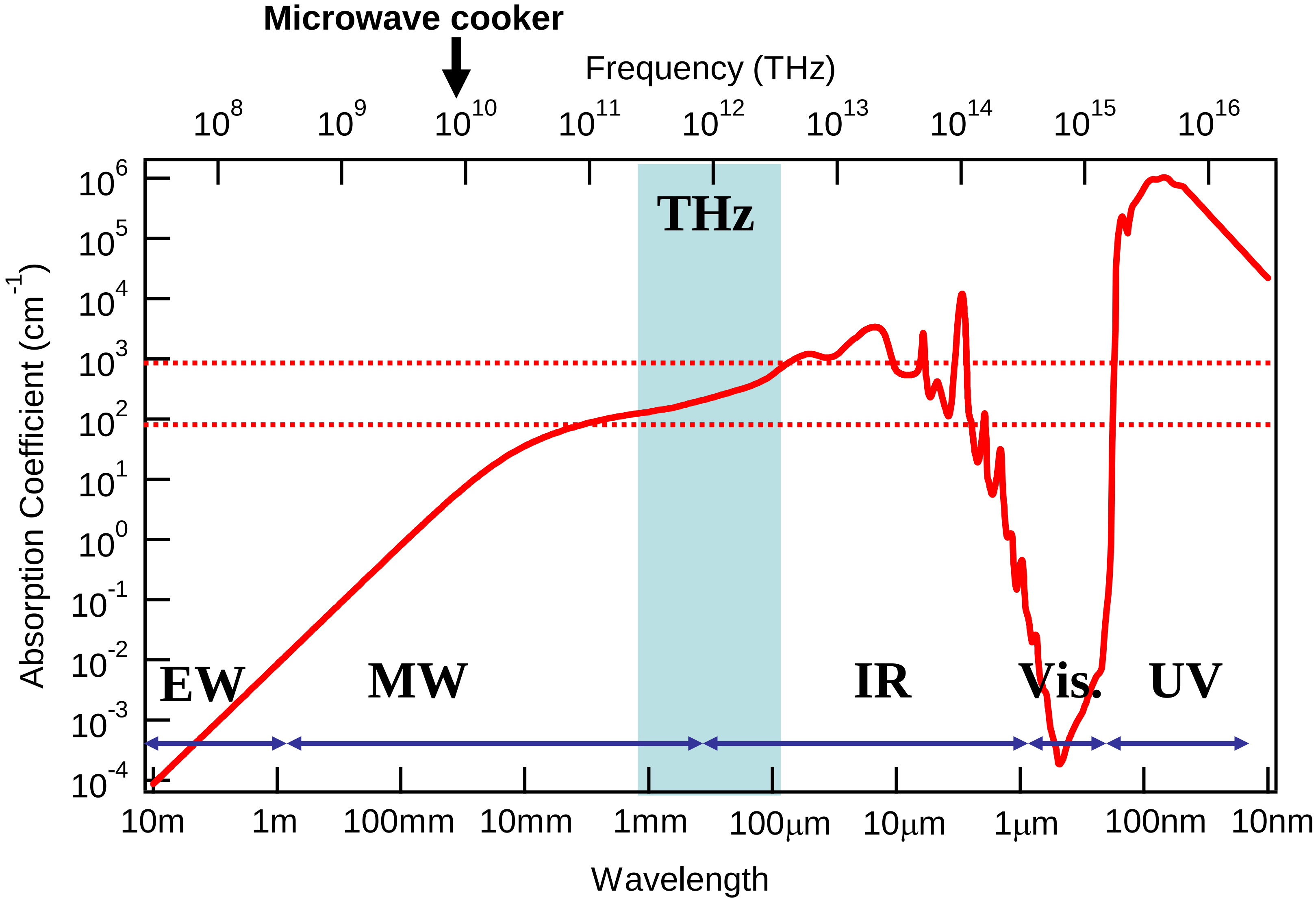
  水、水溶液
- 非線型テラヘルツ分光の必要性と戦略

Importance of terahertz frequency region



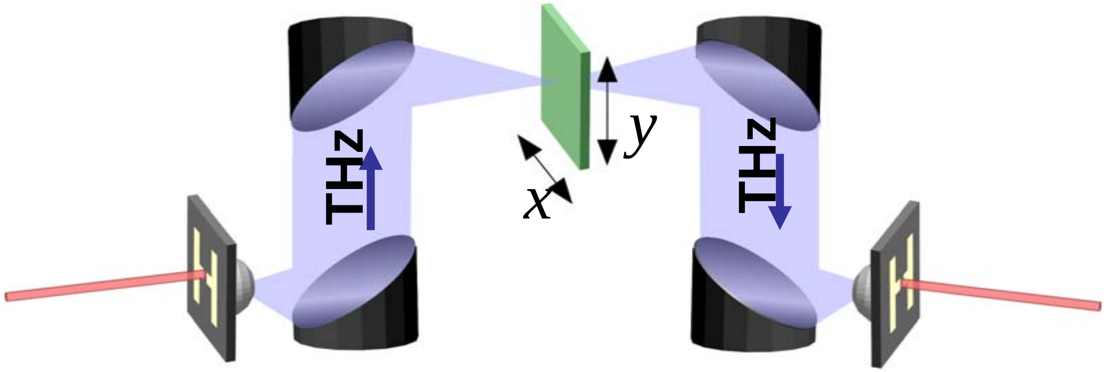
Radio wave

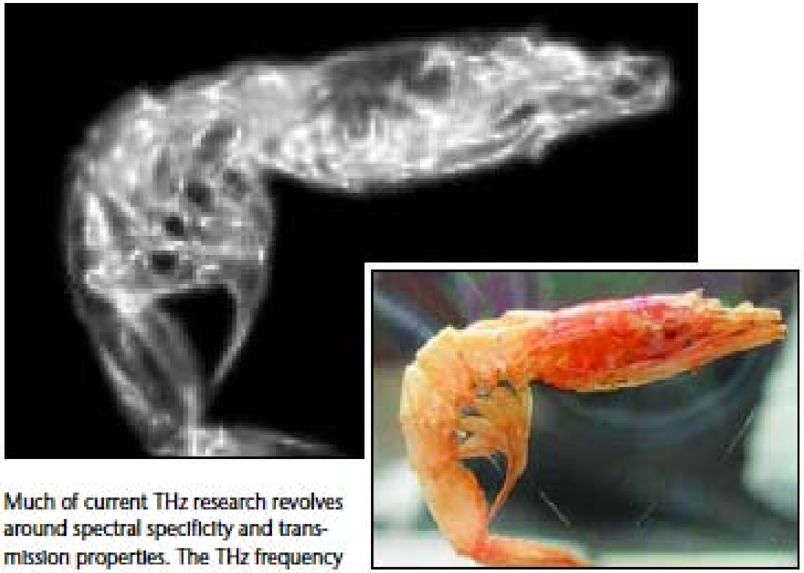
Absorption of Liquid Water



Jackson, Classical Electrodynamics

THz-Imaging





Much of current THz research revolves around spectral specificity and transmission properties. The THz frequency

B. B. Hu and M. C. Nuss, Opt. Lett. 20, 1716 (1995)

Kawase (Nagoya/Riken), Nikon

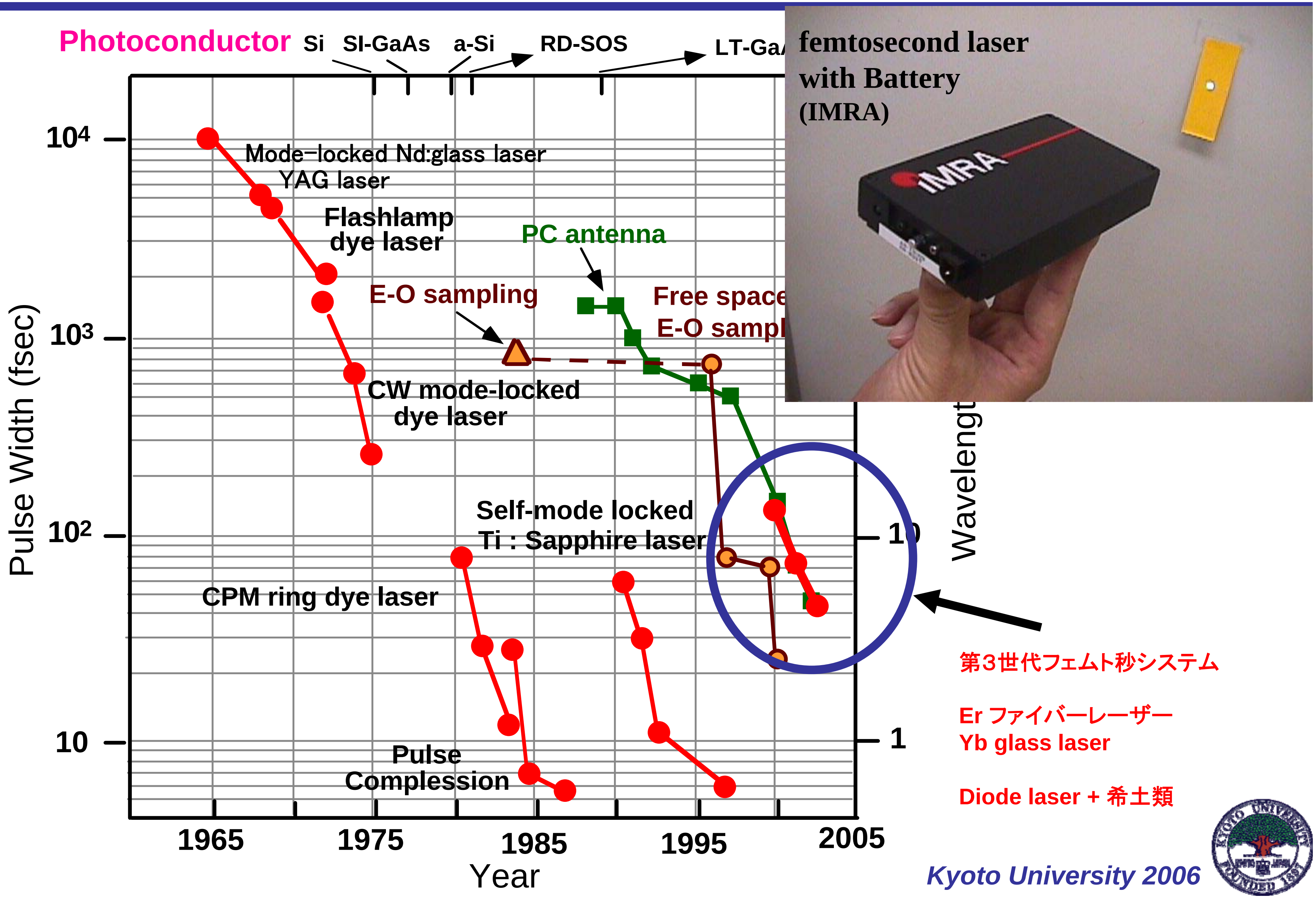
講演内容

- テラヘルツテクノロジーとは何か？
- 時間領域テラヘルツ分光法の基礎
- 時間領域テラヘルツ分光法による物性測定

  有機材料，強誘電体、半導体、超伝導体

  水、水溶液
- 非線型テラヘルツ分光の必要性と戦略

THz-TDSの進展 - フェムト秒レーザーの進歩



femtosecond laser
with Battery
(IMRA)

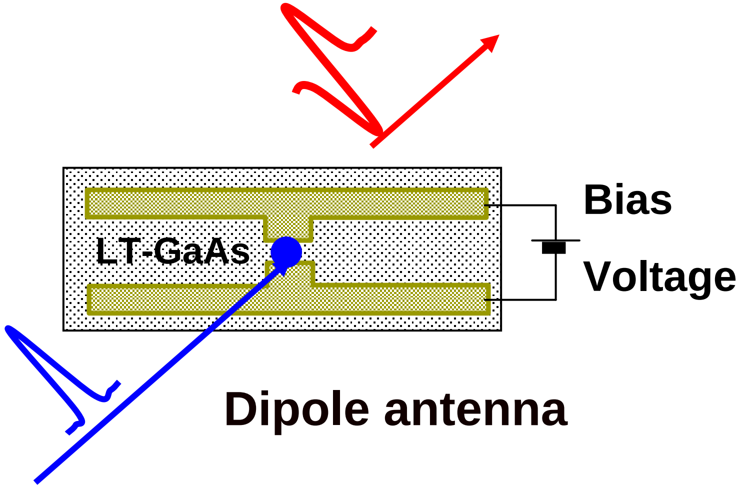
第3世代フェムト秒システム

Er ファイバーレーザー
Yb glass laser

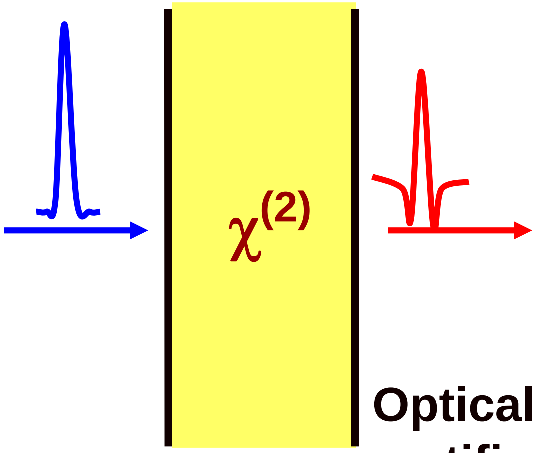
Diode laser + 希土類

THz-detection

Generation



Dipole antenna



Optical rectification

Detection

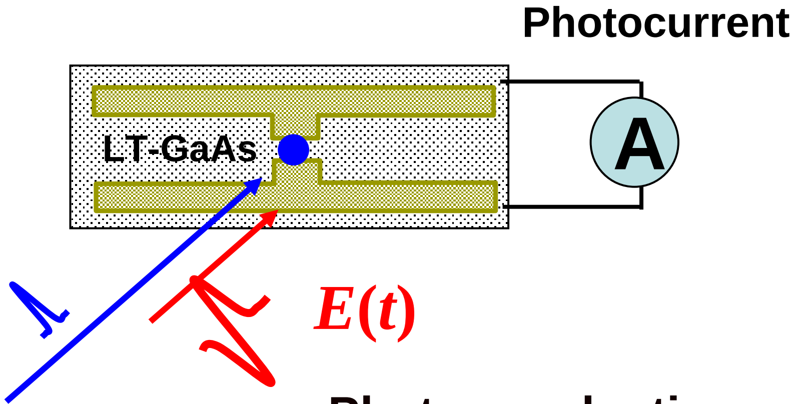
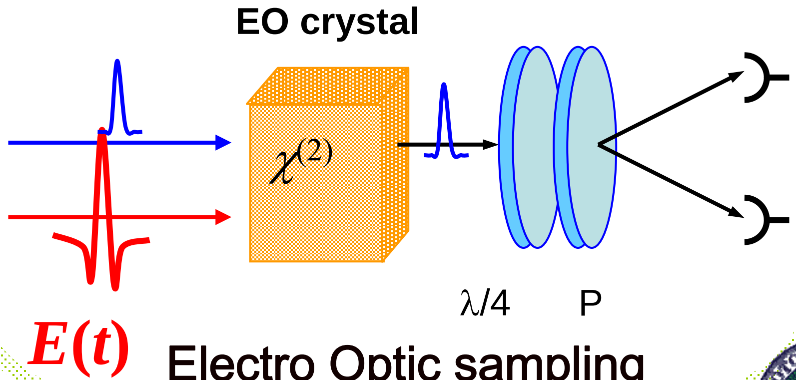
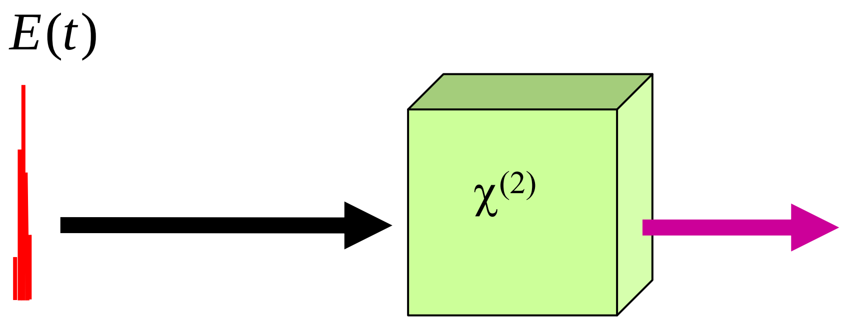


Photo-conducting antenna



Electro Optic sampling

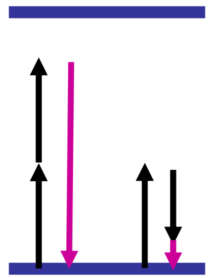
THz-wave generation with second-order non-linear optical process



Pulse width: Δτ

$\Delta\tau \cdot \Delta\omega = 1$

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E_1(t) E_2(t)$$



Sum frequency **Differential frequency**

$$E_1(t) = E_2(t) = A(e^{i\omega t} + e^{-i\omega t})$$

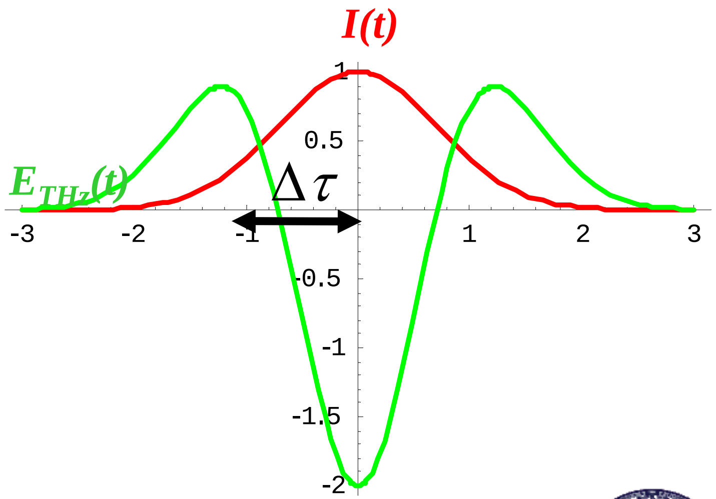
$$P^{(2)}(t) = A^2 (\underbrace{\chi^{(2)}(2\omega,\omega,\omega)e^{i2\omega t}}_{\text{SHG}} + \underbrace{\chi^{(2)}(0,\omega,-\omega)}_{\text{Optical rectification}})$$

SHG **Optical rectification**

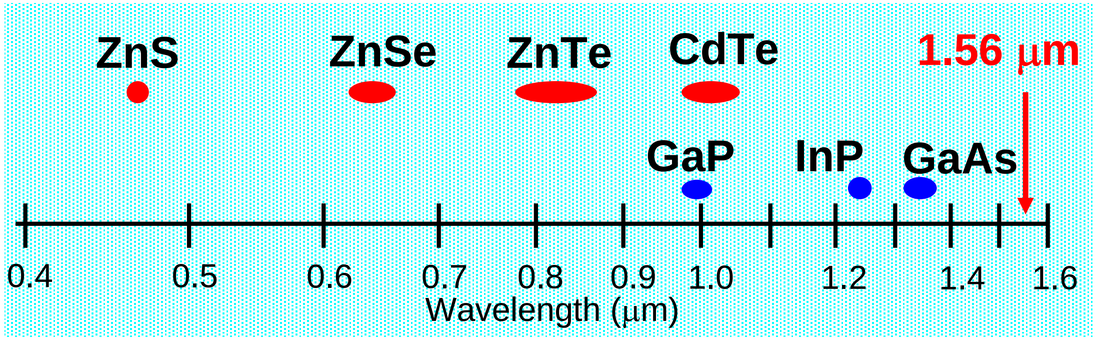
$P^{(2)}(2\omega)$ $P^{(2)}(0)$

$\Delta\tau \cdot \Delta\omega = 1$ $\Omega < \Delta\omega$

$$P^{(2)}(t) = A^2 \chi^{(2)}(\Omega, \omega+\Omega, -\omega))e^{i\Omega t}$$



Phase matching condition



Phase match condition:

$$k(\omega+\Omega) - k(\omega) - k(\Omega) = 0$$

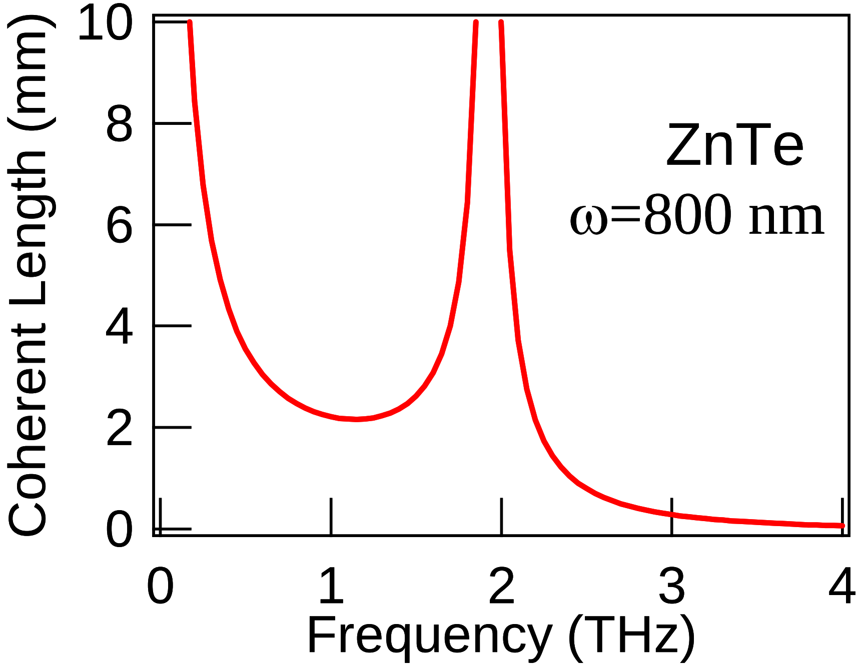
Including dispersion:

refractive index in THz region = **optical Index of group velocity**

$$\boxed{V_{phase}^{THz} = V_{group}^{visible}}$$

Coherent length:

$$l_c = \frac{c}{2f\left|n_{THz} - n_g\right|}$$



Nahata et al., APL **69**, 2321 (1996).

THz-wave detection

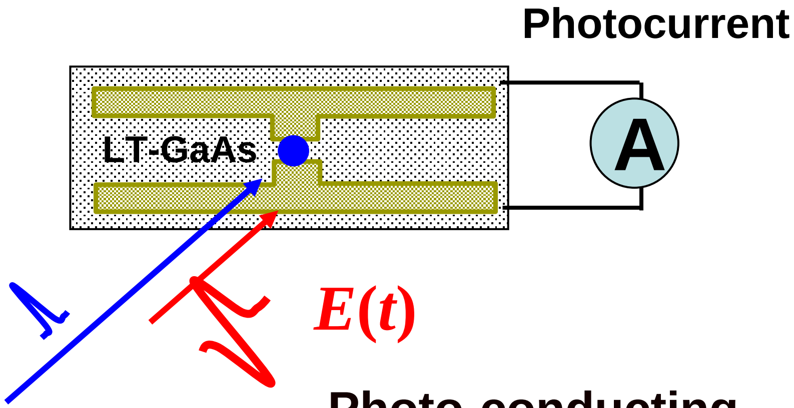
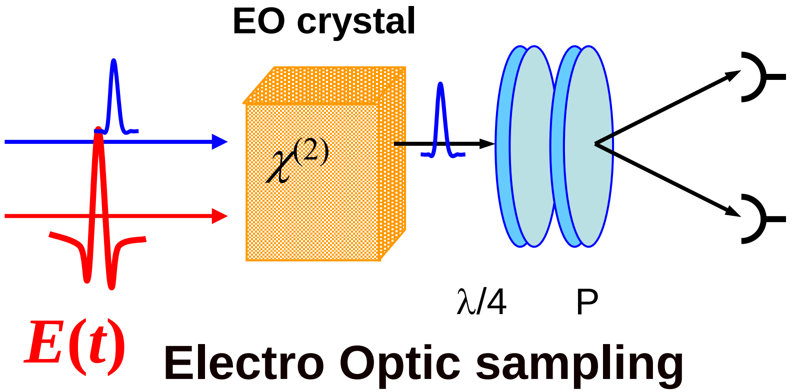
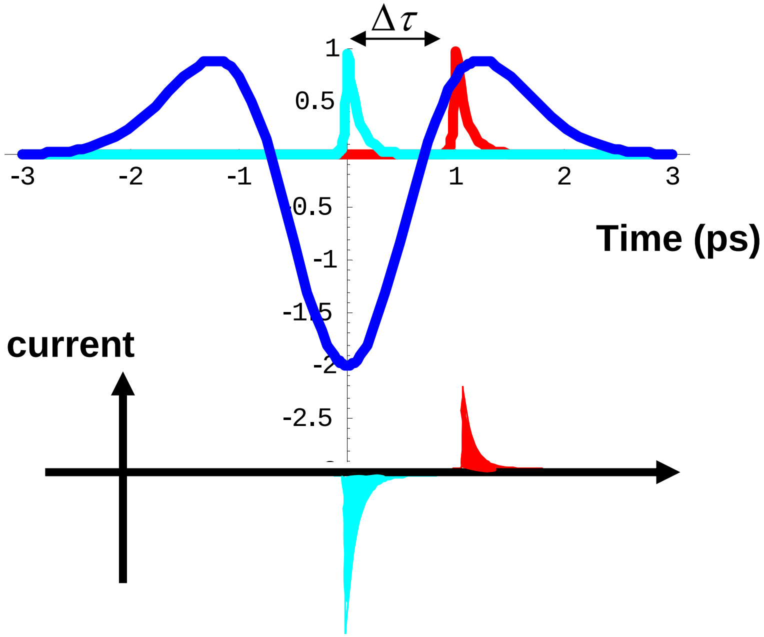


Photo-conducting antenna

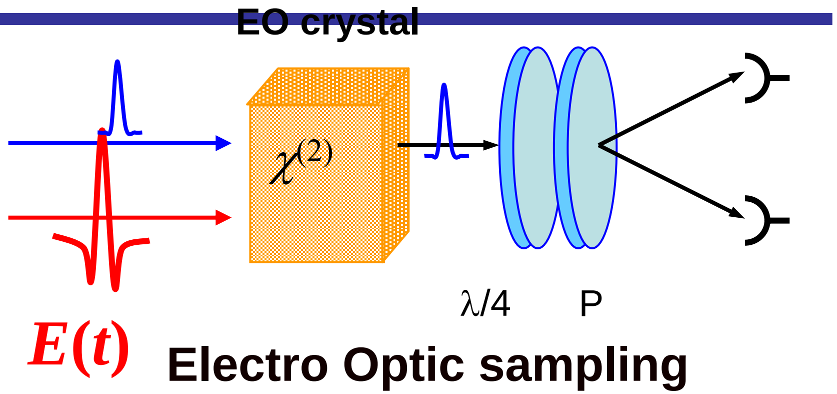


Electro Optic sampling



$$J(\Delta\tau) = e\mu \int_{-\infty}^{\infty} dt E^{THz}(t) N(t-\Delta\tau)$$

Non-linear THz spectroscopy



Electro Optic sampling

State of the art in our laboratory

(Kumiko YAMASHITA, 2005) :

- **Estimation was made by EO sampling technique.**

$$\frac{\Delta I}{I} = -\Gamma = -2\pi \frac{l}{\lambda} n^3 r_{41} \left| E_{THz} \right| = 0.2$$

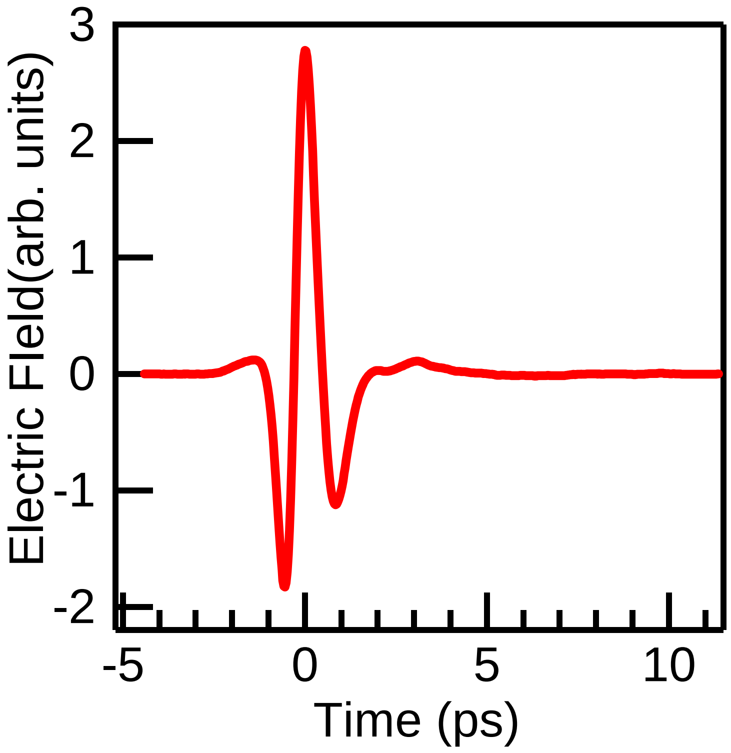
Optical pump: λ =800nm 485 mW (485 μJ /pulse)

ZnTe(ℓ=1mm、n=3.2、$r_{41}=4\times10^{-12}$m/V)

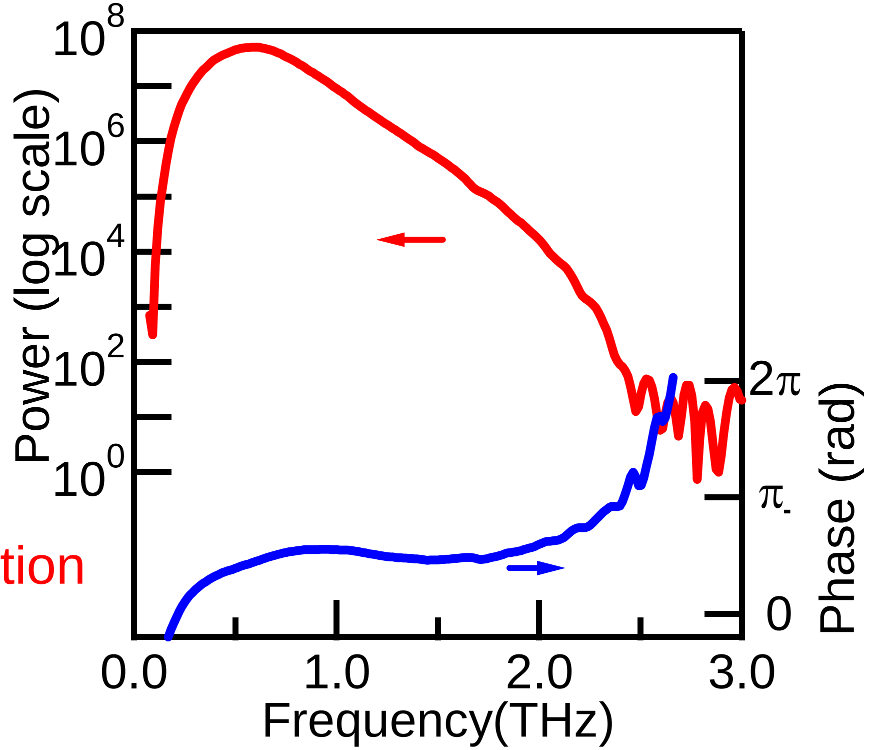
P_{THz}= 100 pJ/pulse, 10 nJ/cm^2, 10 kW/ cm^2, $|E_{THz}|\sim$1kV/cm=100kV/m

Typically, for the non-linear spectroscopy in visible region, we need a MW/cm^2 class laser.

Time-domain spectroscopy (TDS) is a powerful tool in THz region



Fourier transformation



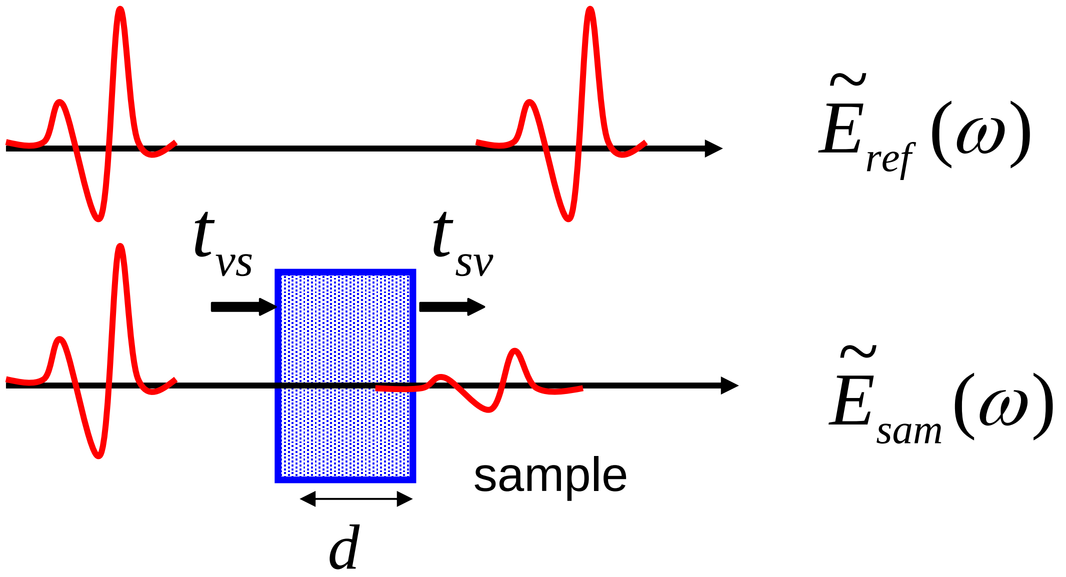
$$\tilde{E}(\omega) = E(\omega)e^{-i\phi(\omega)} = \frac{1}{2\pi}\int E(t)e^{i\omega t}dt$$

✓Pulse measurements ⇒ High sensitivity. cf. FTIR

✓Electric field measurements ⇒ Complex dielectric constants.

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

Terahertz Time-Domain Spectroscopy (THz-TDS)



$$\tilde{t}(\omega) = \frac{\tilde{E}_{sam}(\omega)}{\tilde{E}_{ref}(\omega)}$$

$$= t_{vs} \cdot t_{sv} \exp\left[i \frac{(\tilde{n}(\omega) - 1)\omega d)}{c} \right]$$

$$= t_{vs} \cdot t_{sv} \exp\left[i \frac{(n-1)\omega d)}{c} \right] \exp\left[-\frac{\kappa \omega d)}{c} \right]$$

Complex refractive index

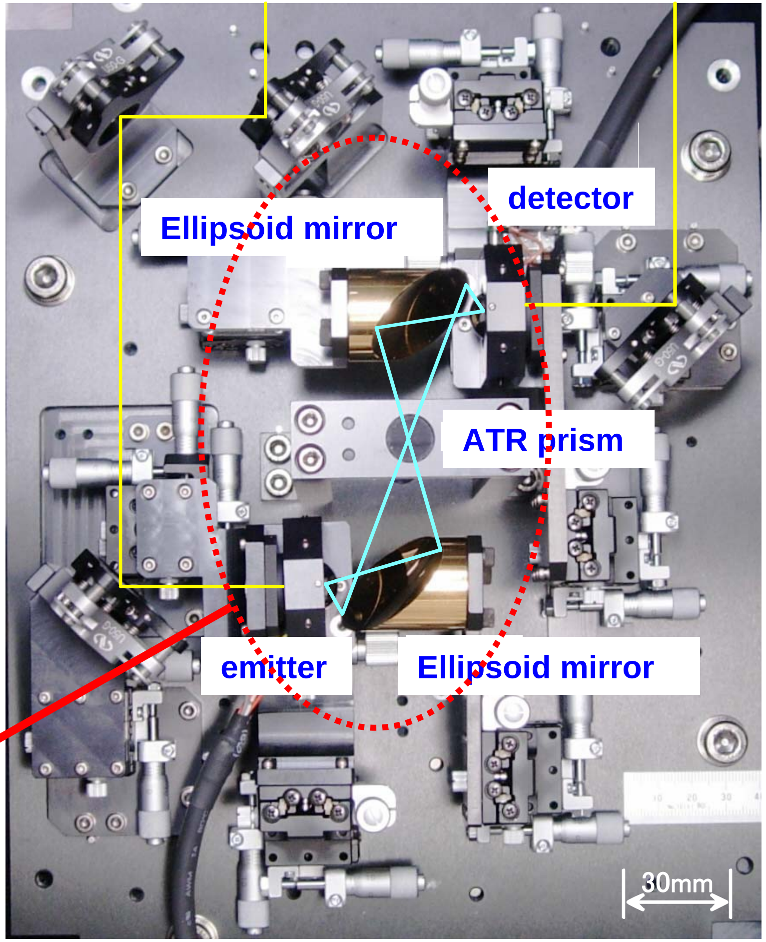
$$\tilde{n} = n + i\kappa$$

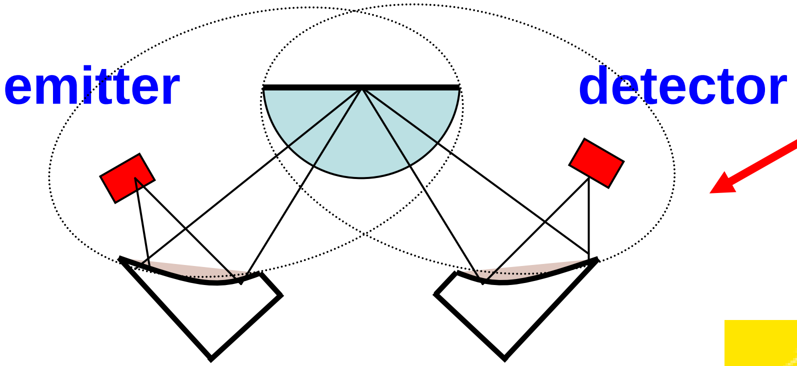
t_{vs}, t_{sv}: Fresnel coefficients

$$t_{vs} = \frac{2}{\tilde{n}(\omega) + 1} \qquad (vacuum \rightarrow sample)$$

$$t_{sv} = \frac{2\tilde{n}(\omega)}{\tilde{n}(\omega) + 1} \qquad (sample \rightarrow vacuum)$$

Compact THz-ATR spectrometer





Double-ellipsoid mirrors with a semispherical ATR prism.

Advanced Terahertz Technologies adapted for Optical Communication

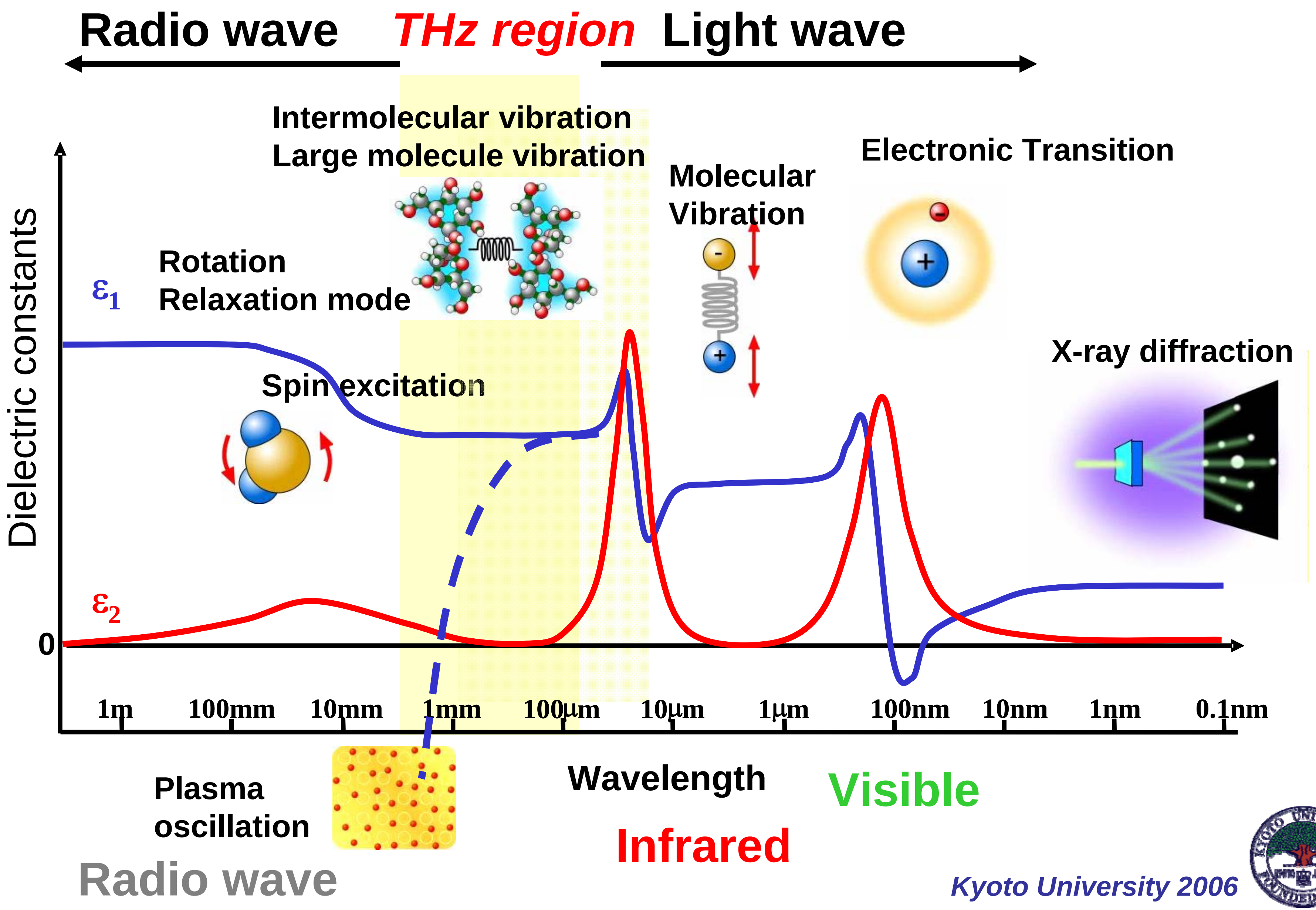
講演内容

- テラヘルツテクノロジーとは何か？
- 時間領域テラヘルツ分光法の基礎
- 時間領域テラヘルツ分光法による物性測定

  有機材料，強誘電体、半導体、超伝導体

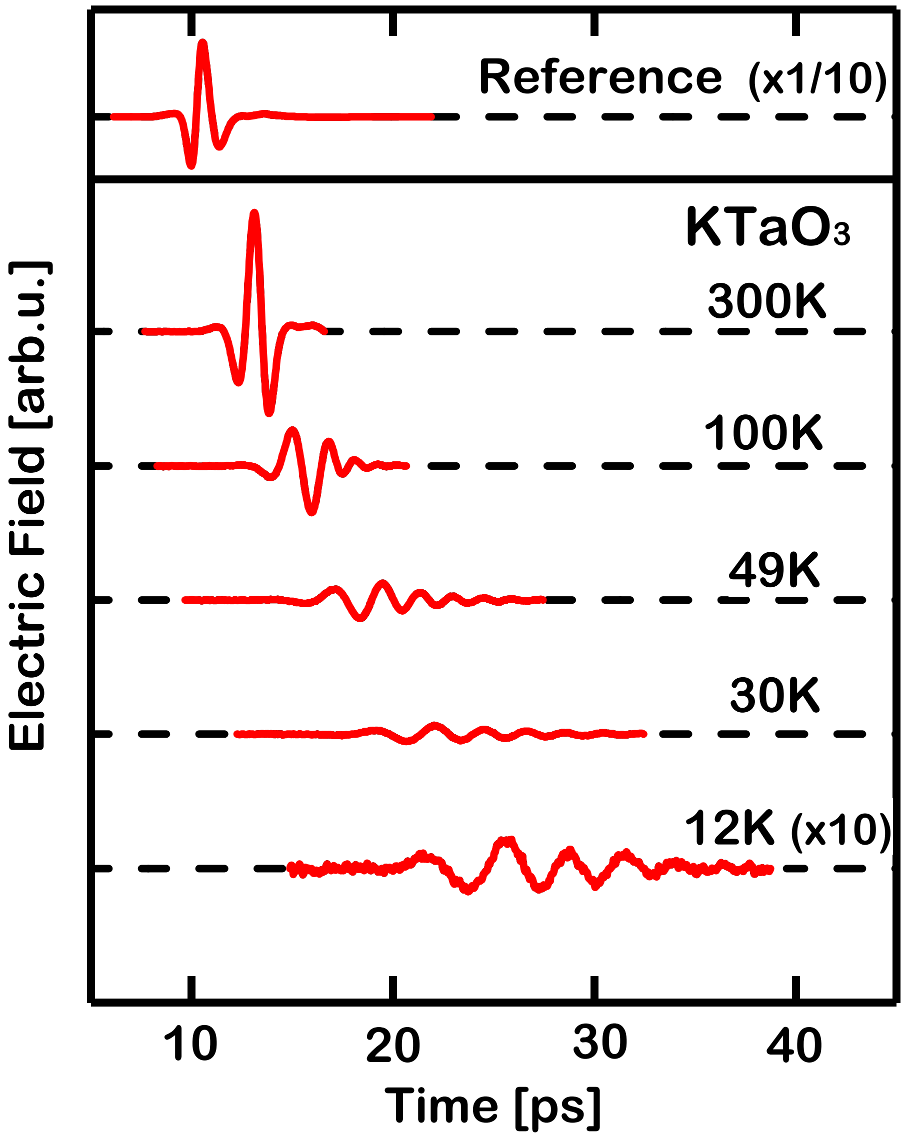
  水、水溶液
- 非線型テラヘルツ分光の必要性と戦略

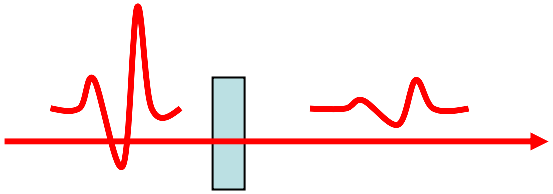
Importance of terahertz frequency region

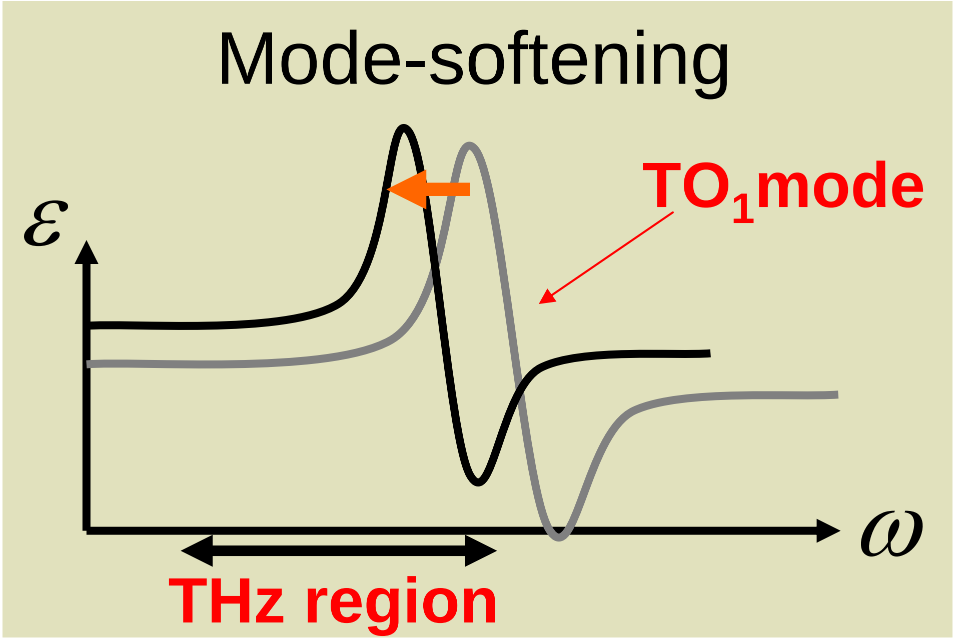


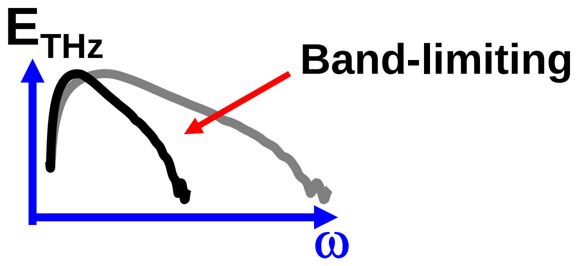
Mode-softening in $KTaO_3$ crystal

Ichikawa, Tanaka *et al*.: Physical Review B 71(2005) 086509.



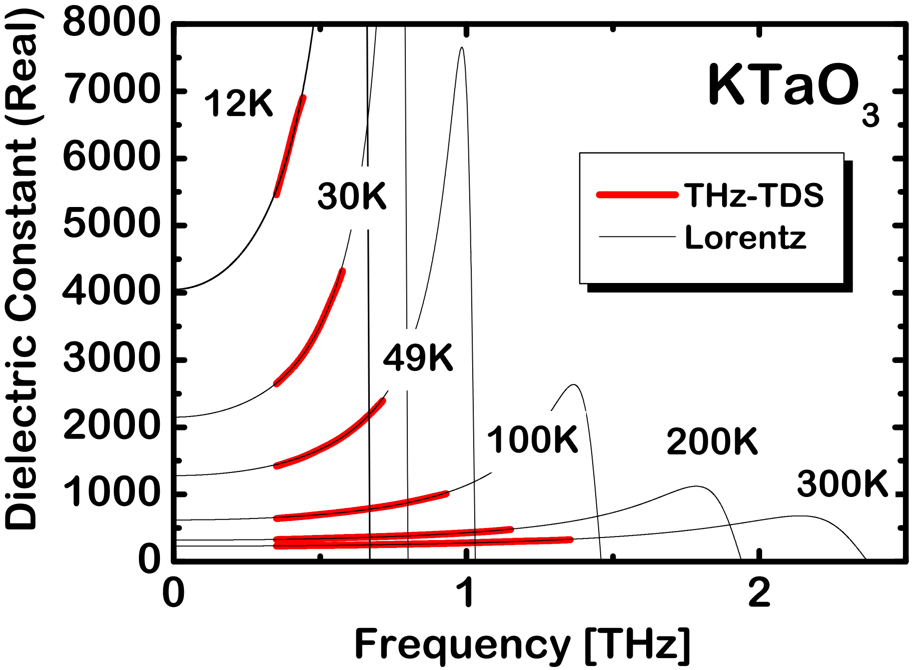


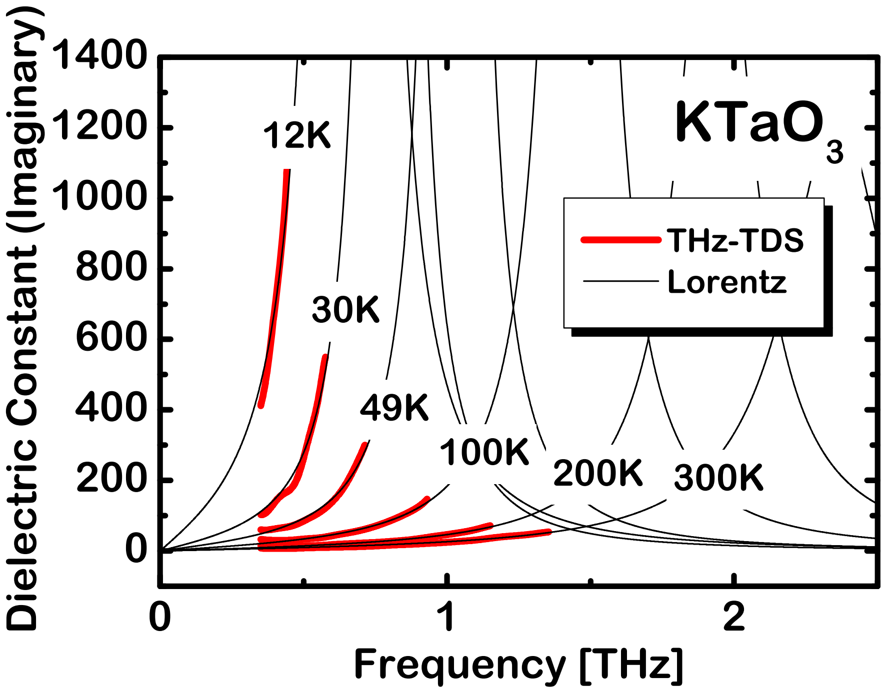




Dielectric function in $KTaO_3$

Ichikawa et al.: Physical Review B 71(2005) 086509.





A single Lorentz Model

$$\tilde{\varepsilon}(\omega) = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)\Omega_0{}^2}{\Omega_0{}^2 - \omega^2 - i\gamma_0\omega}$$

Fitting parameters

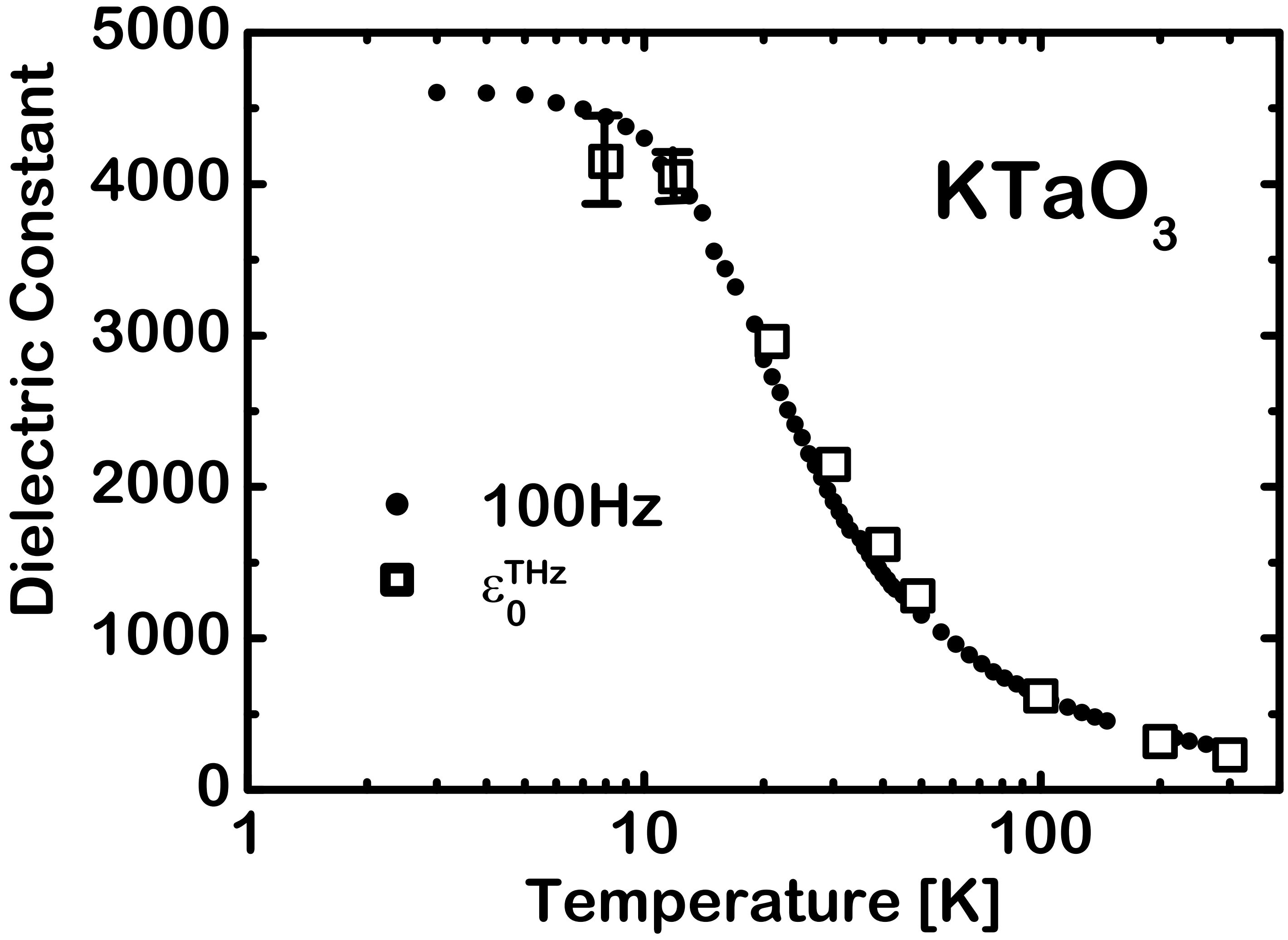
Ω_0 : Mode frequency

γ_0 : Damping

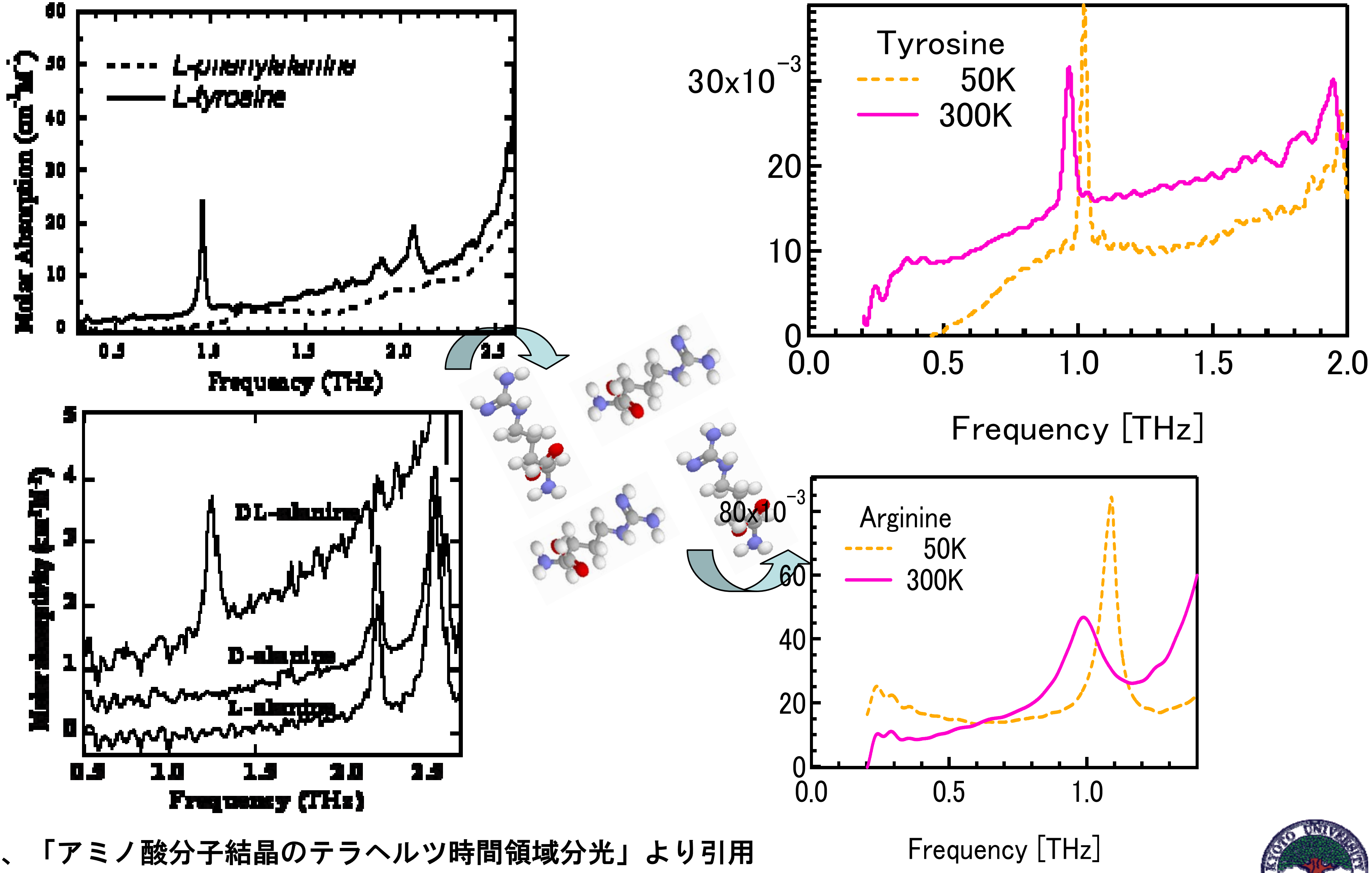
ε_0 :Static dielectric constant

Static dielectric constant

Ichikawa et al.: Physical Review B 71(2005) 086509.



アミノ酸結晶の吸収

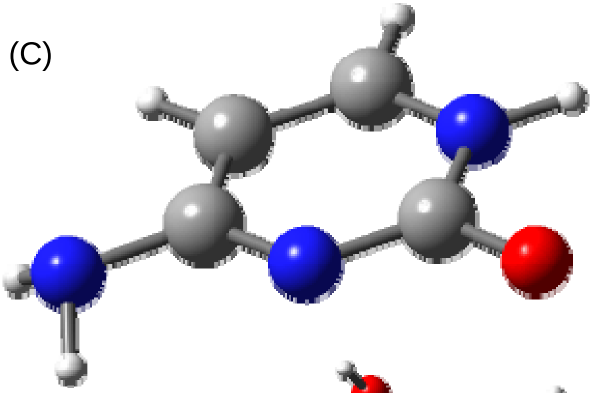


谷他、「アミノ酸分子結晶のテラヘルツ時間領域分光」より引用

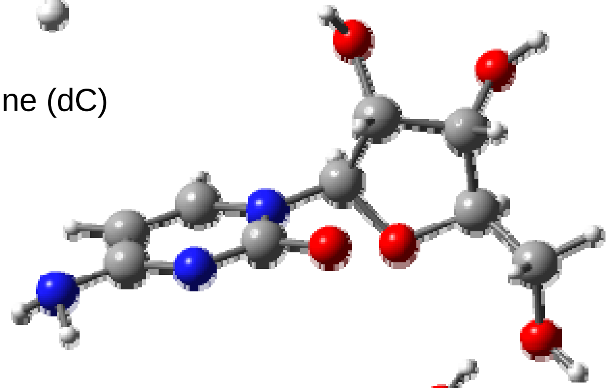
http://www.technova.co.jp/teratech/saizensen/tera_saizensen1.html

isolated bases → DNA: first step

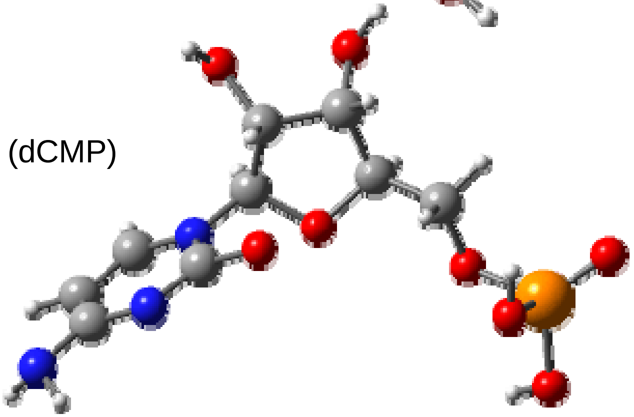
cytosine (C)

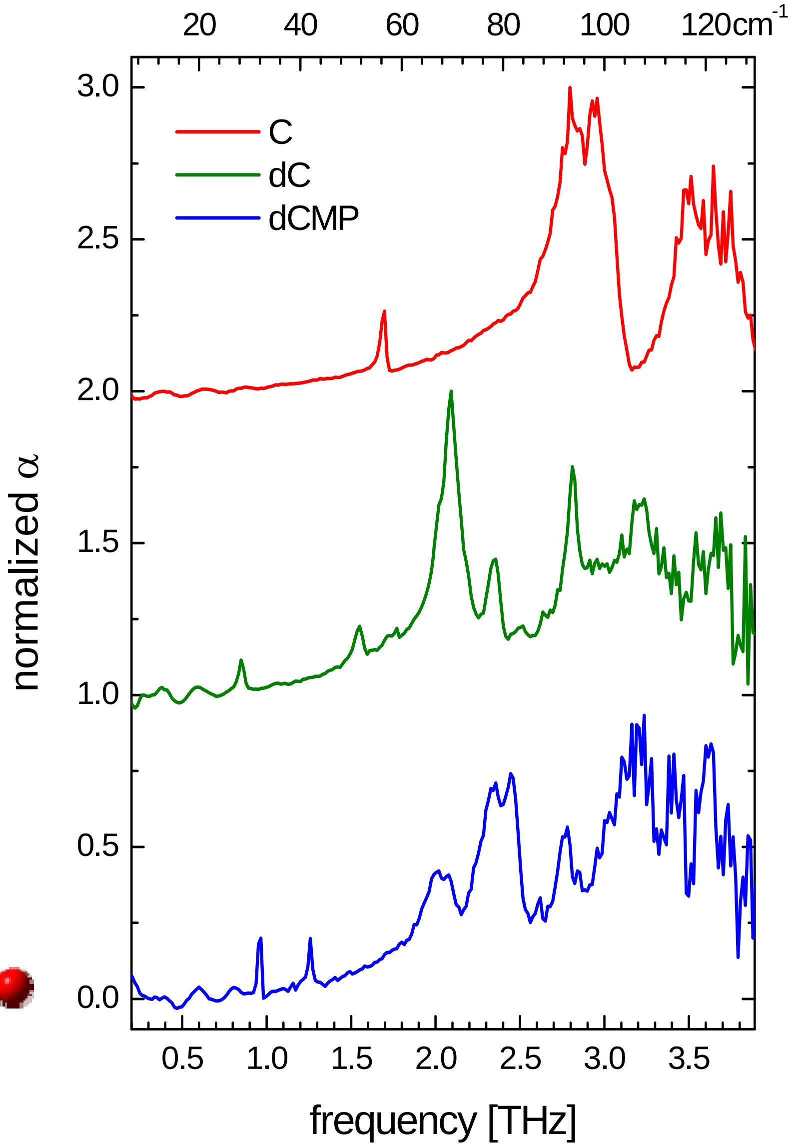


deoxycytidine (dC)



deoxycytidine-
monophosphate (dCMP)





B. M. Fischer, M. Walther, and P. Uhd Jepsen, *Phys. Med. Biol.* **47** 3807 (2002)

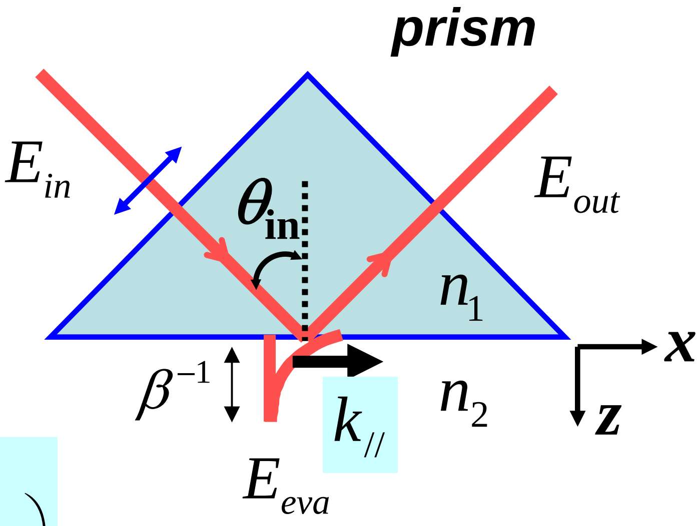
Courtesy of Peter Uhd Jepsen

Evanescent wave in ATR

Internal total reflection condition

Evanescent wave for *p*-polarization

$$\theta_{in} > \theta_c = Sin^{-1}\left(\frac{n_2}{n_1}\right)$$



$$\begin{pmatrix} H_x \\ H_y \\ H_z \end{pmatrix} = \begin{pmatrix} 0 \\ \underline{B} \\ 0 \end{pmatrix} \exp(-\beta z) \exp i\left(\frac{n_1 k \sin\theta_{in}}{n_2} x - \omega t\right)$$

TM mode

$$E_{eva} \quad \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \begin{pmatrix} A \\ 0 \\ C \end{pmatrix} \underline{\exp(-\beta z)} \exp i\left(\frac{n_1 k \sin\theta_{in}}{n_2} x - \omega t\right)$$

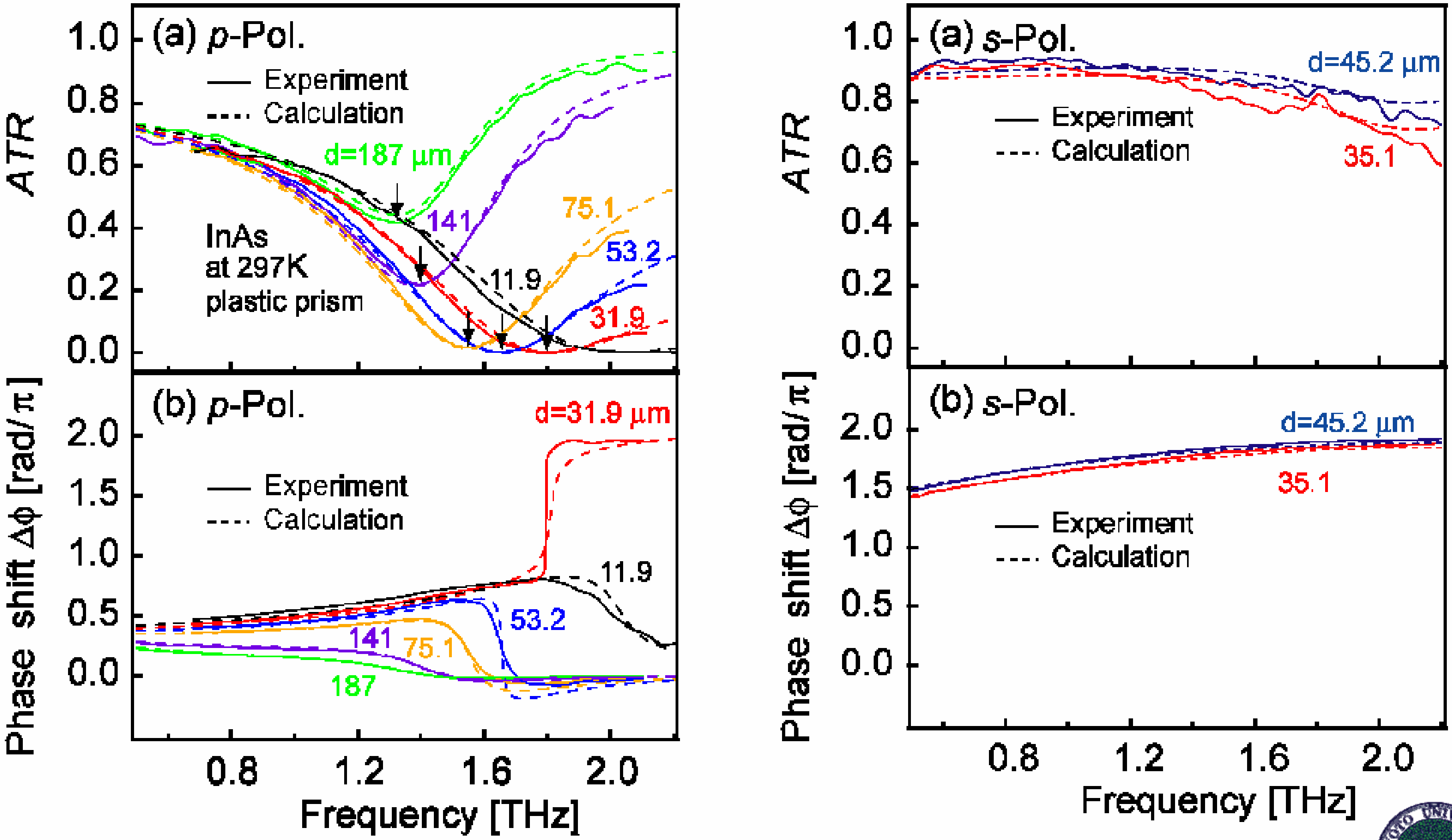
exponentially decaying away from the interface waves

TM mode with large *k*//

Enable to excite surface modes.

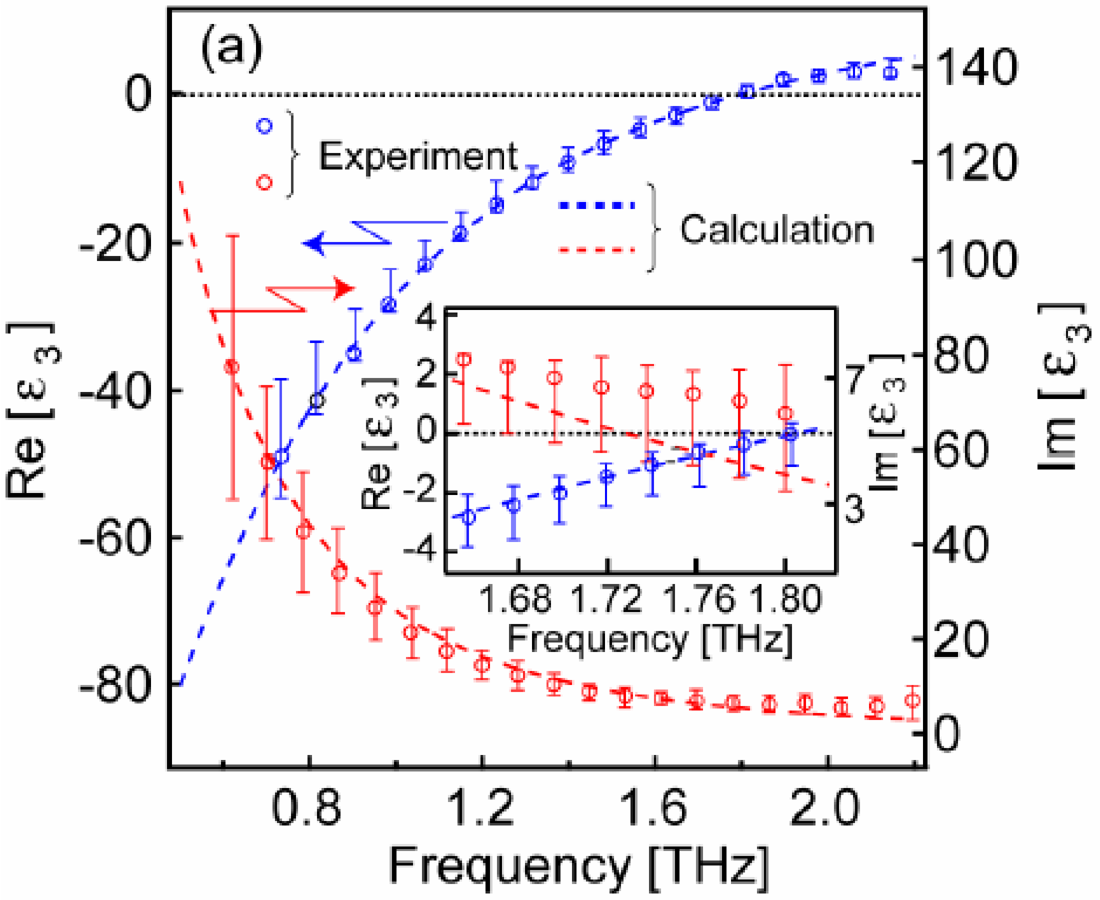
Surface plasmon in semiconductor with THz-ATR

H. Hirori, M. Nagai, and K. Tanaka, Optics Express, 13, (26), 10801-10814 (2005).

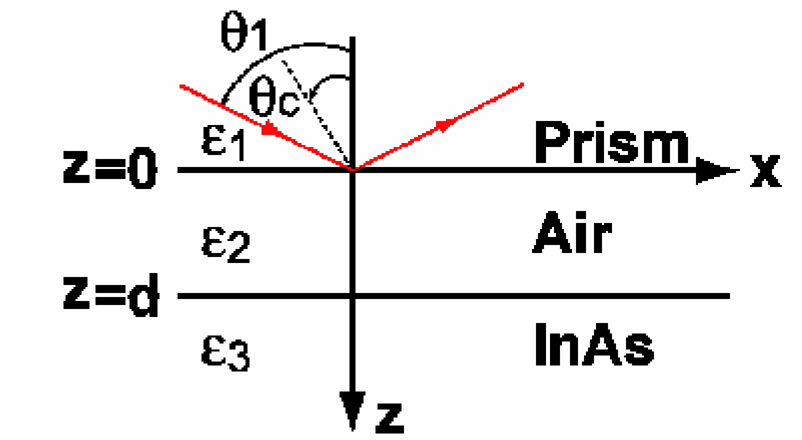


Dielectric constant in doped semiconductor

InAs n-type $n \sim 10^{17}$



Attenuated Total Reflection



$$\varepsilon(\omega) = \varepsilon_b \left(1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \right),$$

$$\omega << \gamma$$

$$\varepsilon(\omega) = i \frac{\varepsilon_b \omega_p^2}{\gamma} \frac{1}{\omega} = i \frac{\sigma}{\omega}$$

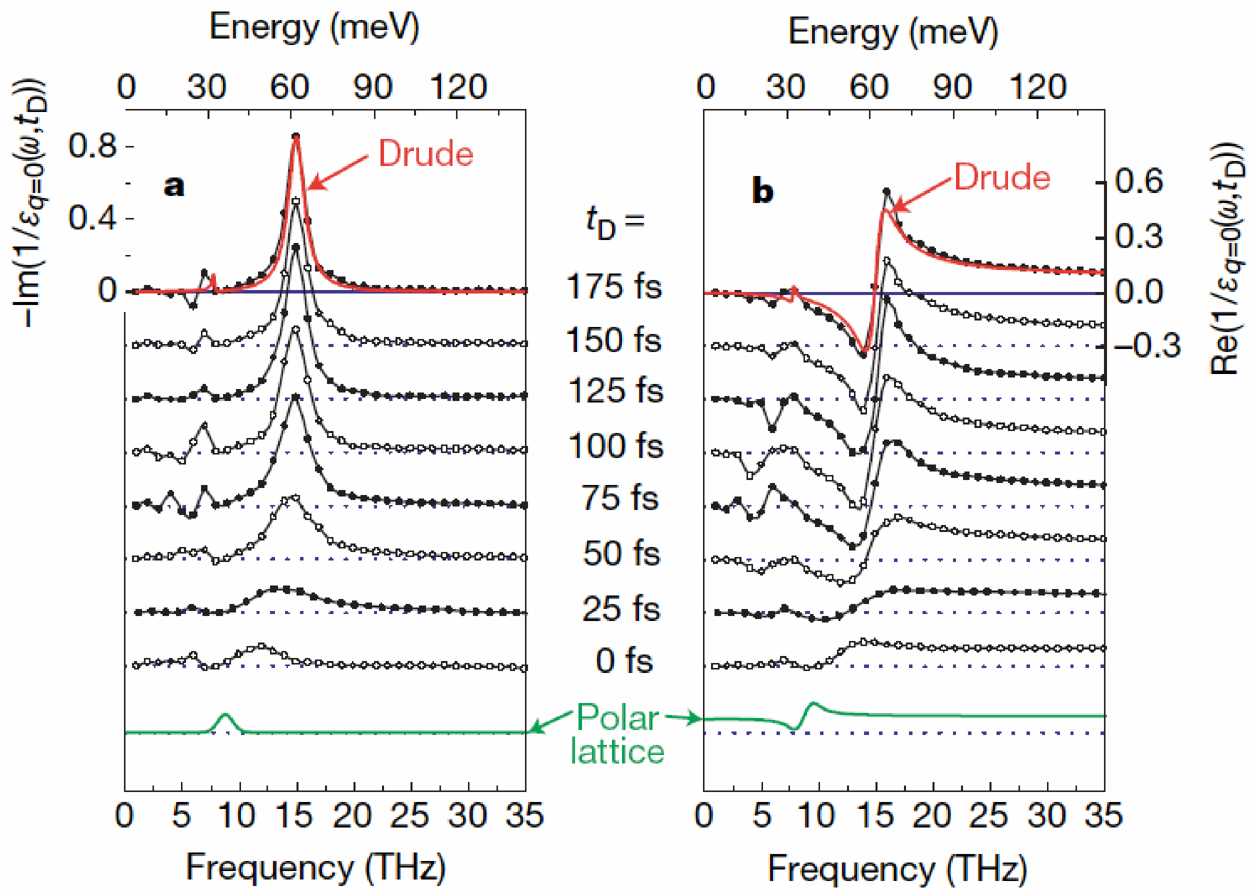
H. Hirori, M. Nagai, and K. Tanaka, Optics Express, 13, (26), 10801-10814 (2005).

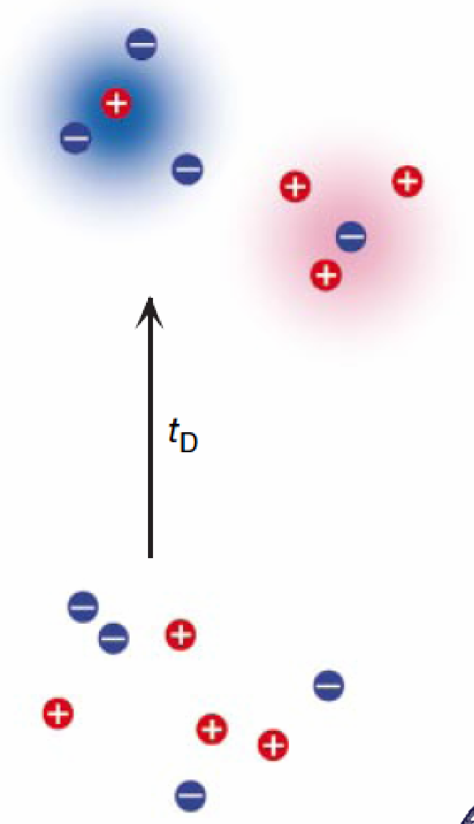
Pump and probe spectroscopy

"How many-particle interactions develop after ultrafast excitation of electron-hole plasma "

R. Huber, F. Tauser, A. Brodschelm, M. Bichler, G. Abstrelter and A. Leitenstorfer

Nature. Vol.**414** (2001) 286

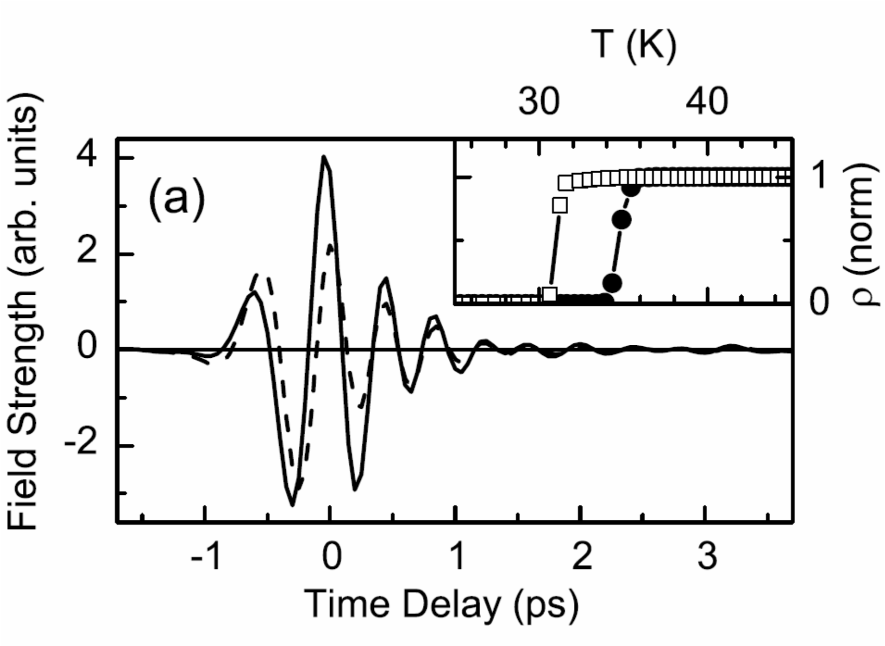




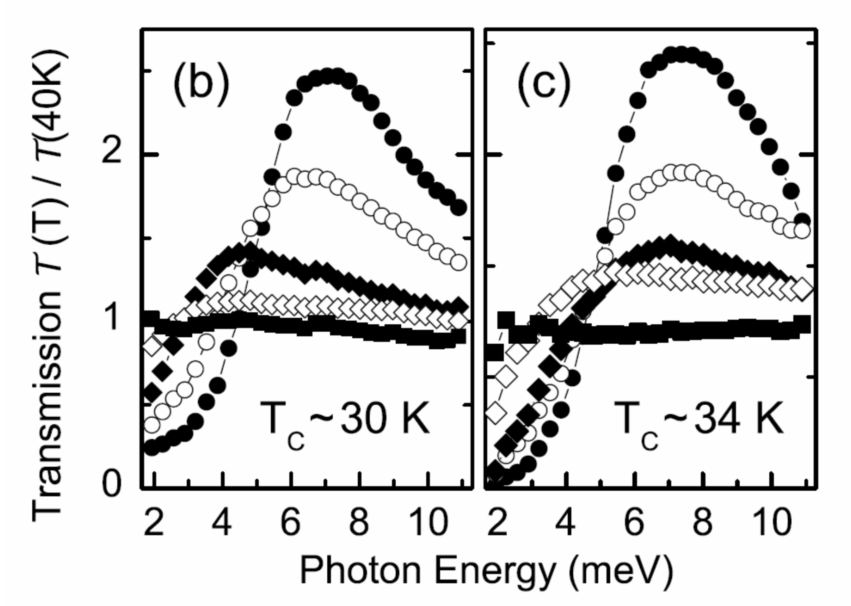
Superconducting gap

“Far-infrared optical conductivity gap in superconducting MgB_2 films”

R. A. Kaindl, M. A. Carnahan, J. Orestein, D. S. Chemla, H. M. Christen, H. Y. Zhai, M. Paranthaman and D. H. Lowndes Phys. Rev. Lett. 88 (2002) 027003



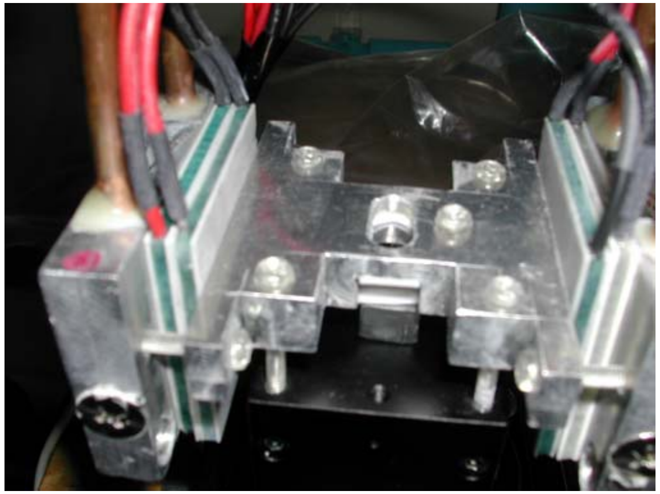
100nm MgB_2薄膜における透過波形
6K（実線）と40K（破線）
Insetは100nm MgB_2薄膜（□）と200nm MgB_2薄膜（●）の抵抗の温度変化

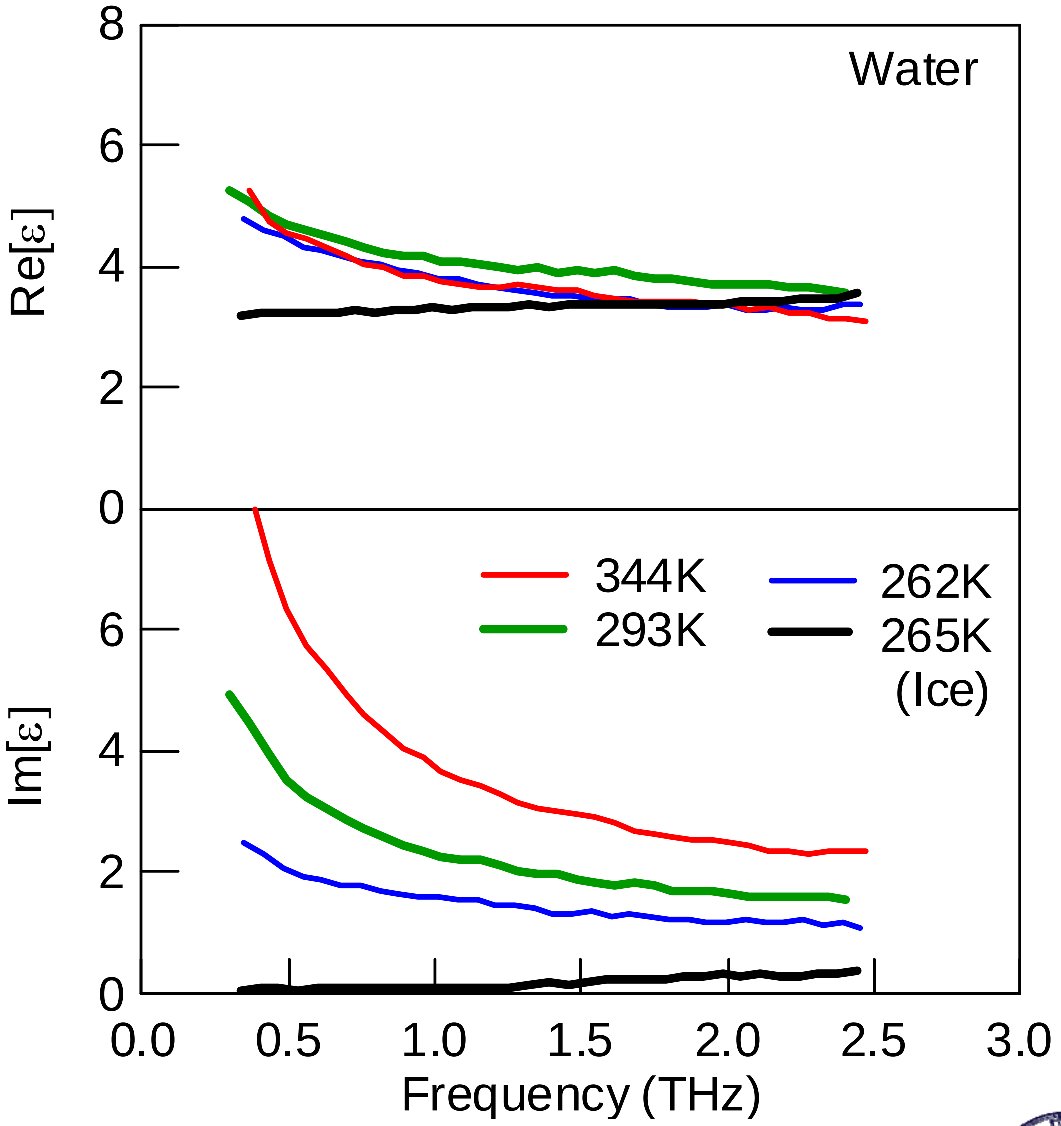


40Kの値で規格化した透過率
(b)膜厚100nm MgB_2
6K（●）, 20K（○） 27K（◆）, 30K（◇）, 33K（■）
(c)膜厚200nm MgB_2（6K, 20K 25K, 30K, 36K)
6K（●）, 20K（○） 25K（◆）, 30K（◇）, 36K（■）

TD-ATR spectroscopy in water

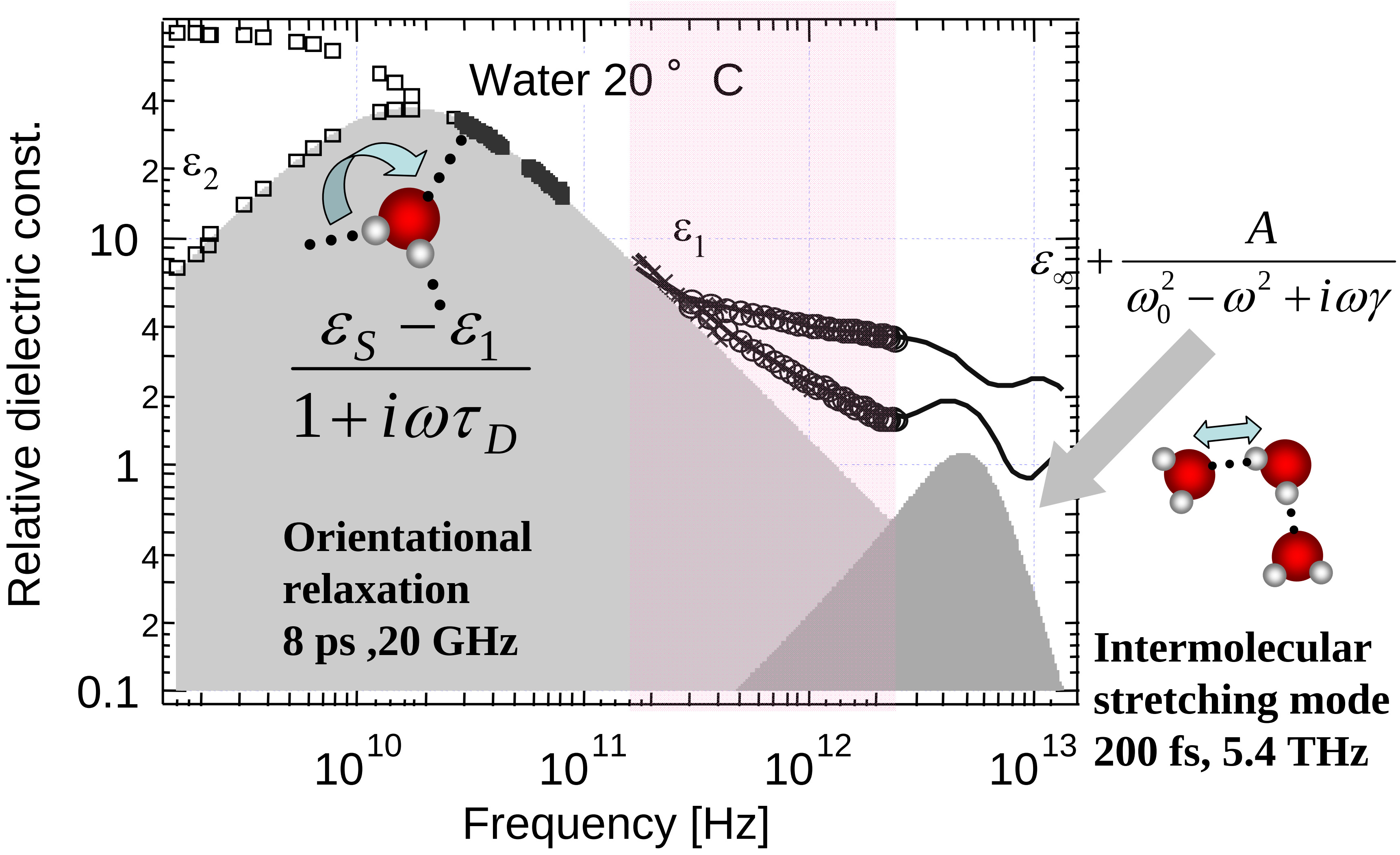
Temperature-controlled
Si ATR prism





Nagai, et al. , Int. J. IRMMW, **27,** 505 (2006).

Dielectric Constants in Water



$$\frac{\varepsilon_S - \varepsilon_1}{1+i\omega\tau_D}$$

Orientational relaxation
8 ps ,20 GHz

$$\varepsilon_\infty + \frac{A}{\omega_0^2 - \omega^2 + i\omega\gamma}$$

Intermolecular stretching mode
200 fs, 5.4 THz

講演内容

- テラヘルツテクノロジーとは何か？
- 時間領域テラヘルツ分光法の基礎
- 時間領域テラヘルツ分光法による物性測定

  有機材料，強誘電体、半導体、超伝導体

  水、水溶液
- 非線型テラヘルツ分光の必要性と戦略

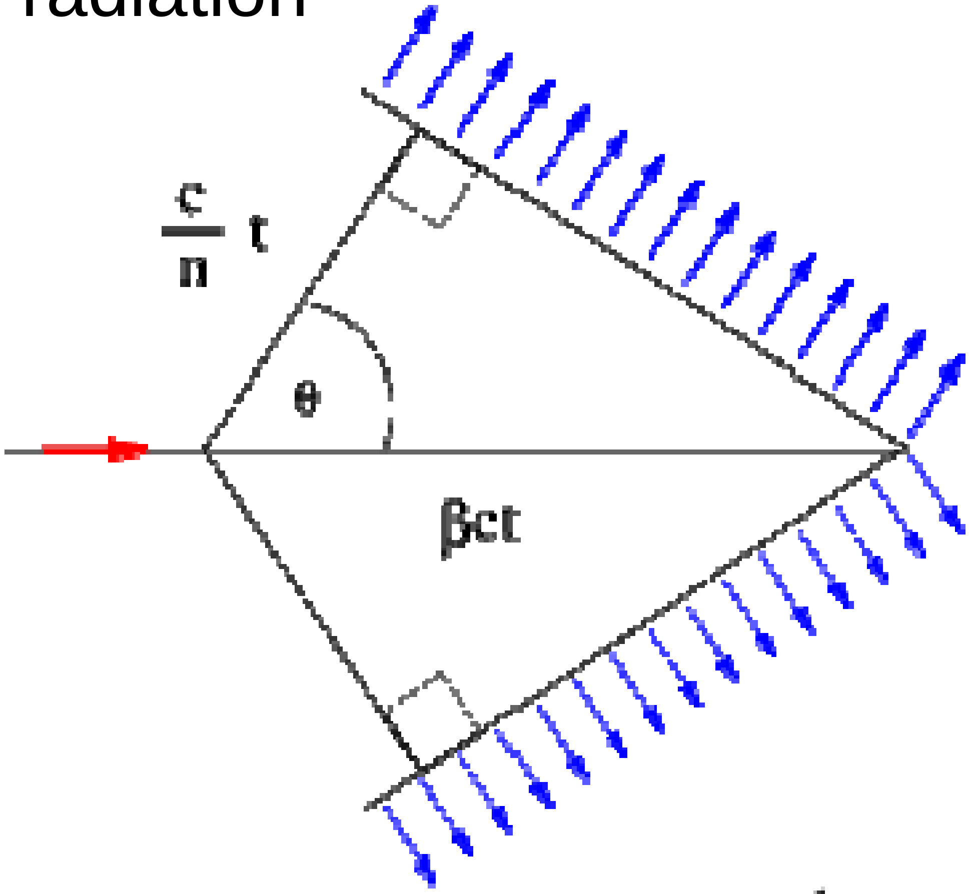
Non-linear THz spectroscopy

State of the art of the THz pulse power using fs laser

	Pump power		THz power		
Ti: Al_2O_3 laser		/pulse	average	**/pulse**	Electric field (V/cm)
Amplifier@1KHz	650 mW	**650 μJ**	0.1μW	**125 pJ**	**1 KV/cm**
Oscillator with magnetic field (Sarukura 1.7T, @80MHz)	1500 mW 650 mW	**18.75 nJ** **8.125 nJ**	650 μW 110 μW	**8.125pJ** **1.375pJ**	**0.25 KV/cm** **0.1 KV/cm**

Non-linear THz spectroscopy

- Cherenkov radiation



Cherenkov angle $\cos\theta = \frac{1}{n\beta}$

Non-linear THz spectroscopy

- **Basic idea of <u>velocity matching</u> by pulse front using Cherenkov effect with non-linear crystal**

A. G. Stepanov, J. Hebling and J. Kuhl *et al.,* Appl. Phys. Lett. 83, 3000 (2003).

$$\theta_c = \arccos(v_{\mathrm{THz}}/v_{\mathrm{vis}}),$$

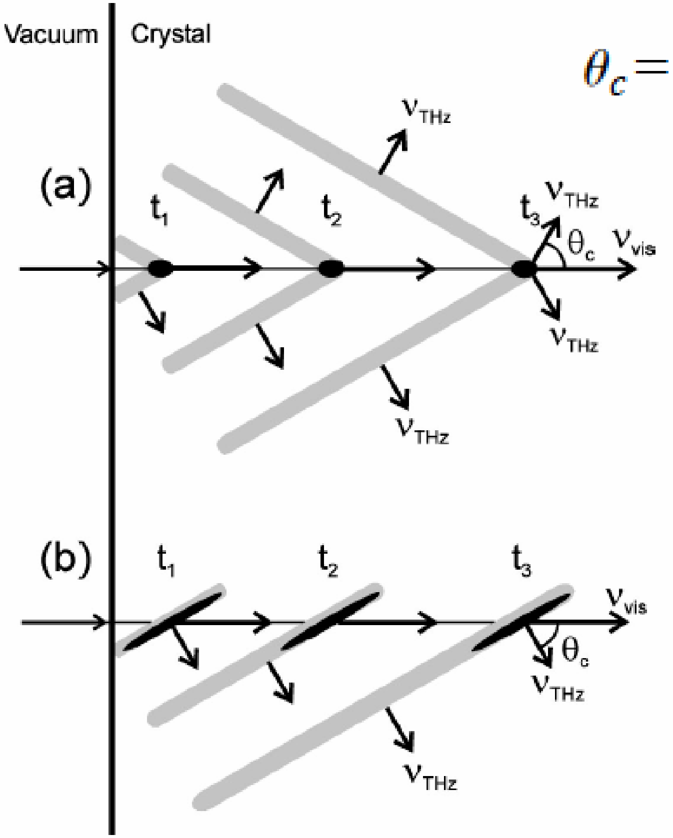


FIG. 1. Schematic illustration of the THz generation by tightly focused (a) and front tilted (b) femtosecond laser pulses propagating in an electro-optical crystal. Black ellipses and gray areas depict laser and generated THz pulses, respectively, at three different instants of time ($t_1 < t_2 < t_3$).

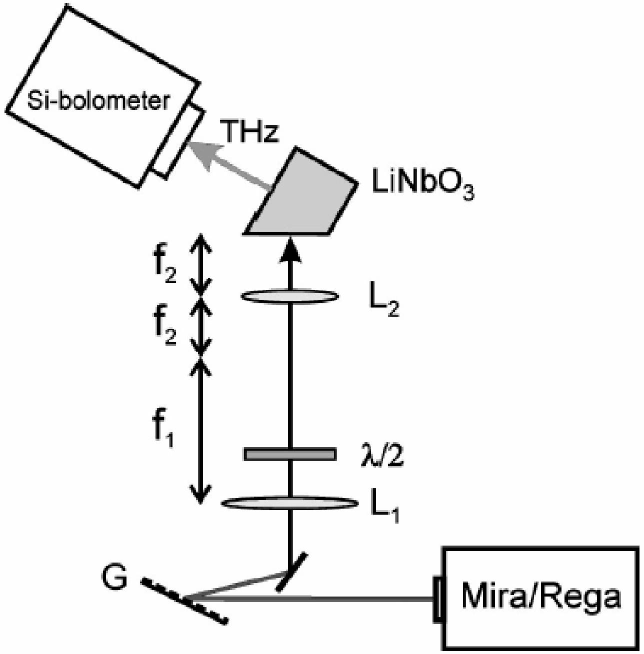
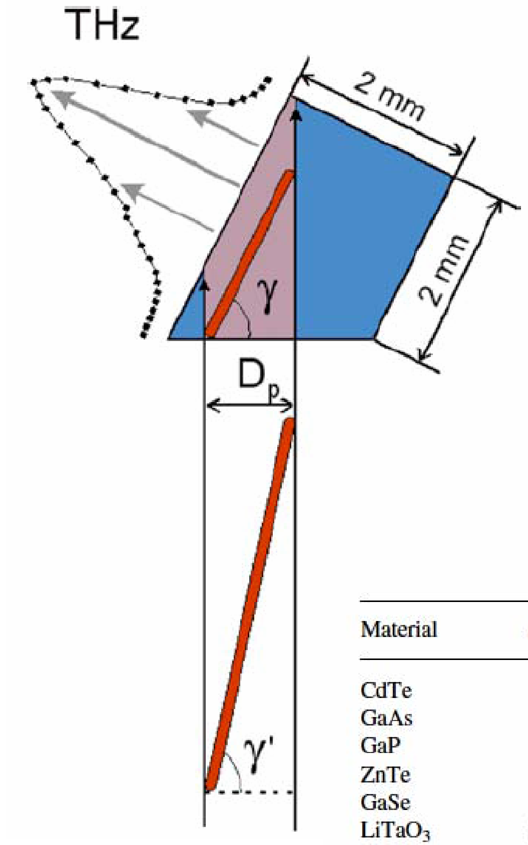


FIG. 2. Experimental setup used for the THz generation by femtosecond laser pulses with tilted pulse fronts.

Non-linear THz spectroscopy

- **High power single cycle THz generation using tilted femtosecond light sources**

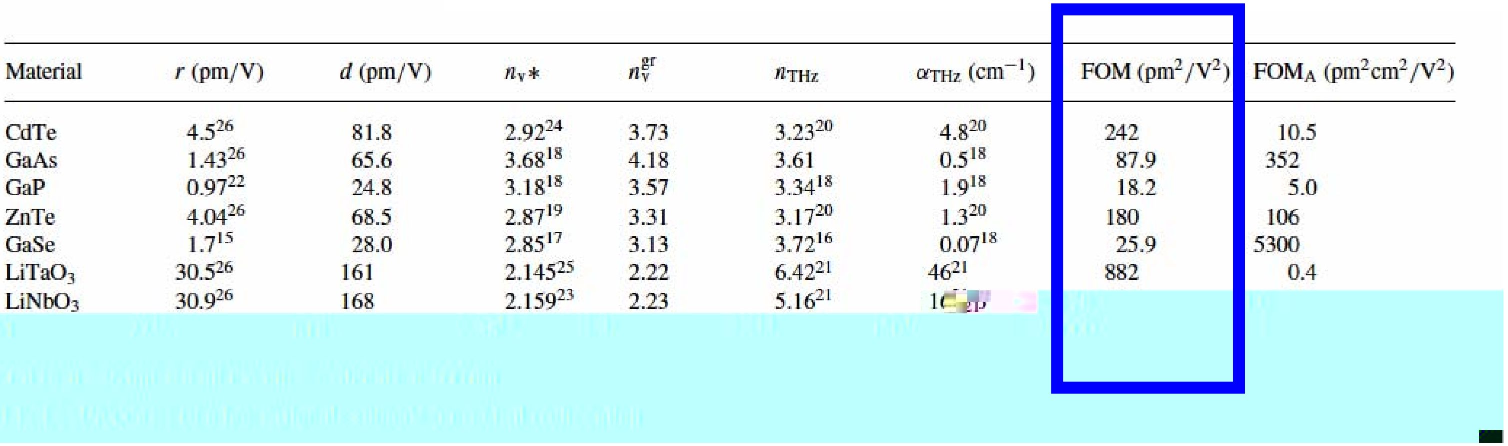
J. Hebling and J. Kuhl *et al.*, Appl. Phys. B 78, 593-599 (2004).



$$v_{vis}^{gr} \cdot \cos\gamma = v_{THz}^{ph} \cdot$$

$$v_{vis}^{gr} >> v_{THz}^{ph}$$

$$n_{v}^{gr} << n_{THz}$$



Material	r (pm/V)	d (pm/V)	$n_{v}*$	n_{v}^{gr}	n_{THz}	α_{THz} (cm^{-1})	FOM (pm^2/V^2)	FOM$_A$ (pm^{2}cm^2/V^2)
CdTe	4.5[26]	81.8	2.92[24]	3.73	3.23[20]	4.8[20]	242	10.5
GaAs	1.43[26]	65.6	3.68[18]	4.18	3.61	0.5[18]	87.9	352
GaP	0.97[22]	24.8	3.18[18]	3.57	3.34[18]	1.9[18]	18.2	5.0
ZnTe	4.04[26]	68.5	2.87[19]	3.31	3.17[20]	1.3[20]	180	106
GaSe	1.7[15]	28.0	2.85[17]	3.13	3.72[16]	0.07[18]	25.9	5300
LiTaO$_3$	30.5[26]	161	2.145[25]	2.22	6.42[21]	46[21]	882	0.4
LiNbO$_3$	30.9[26]	168	2.159[23]	2.23	5.16[21]	[illegible]	[illegible]	[illegible]

Non-linear THz spectroscopy

- **High power single cycle THz generation using tilted femtosecond light sources** : Recent status

A. G. Stepanov and J. Heebling *et al.*, Optics Express 13 5762 (2005).

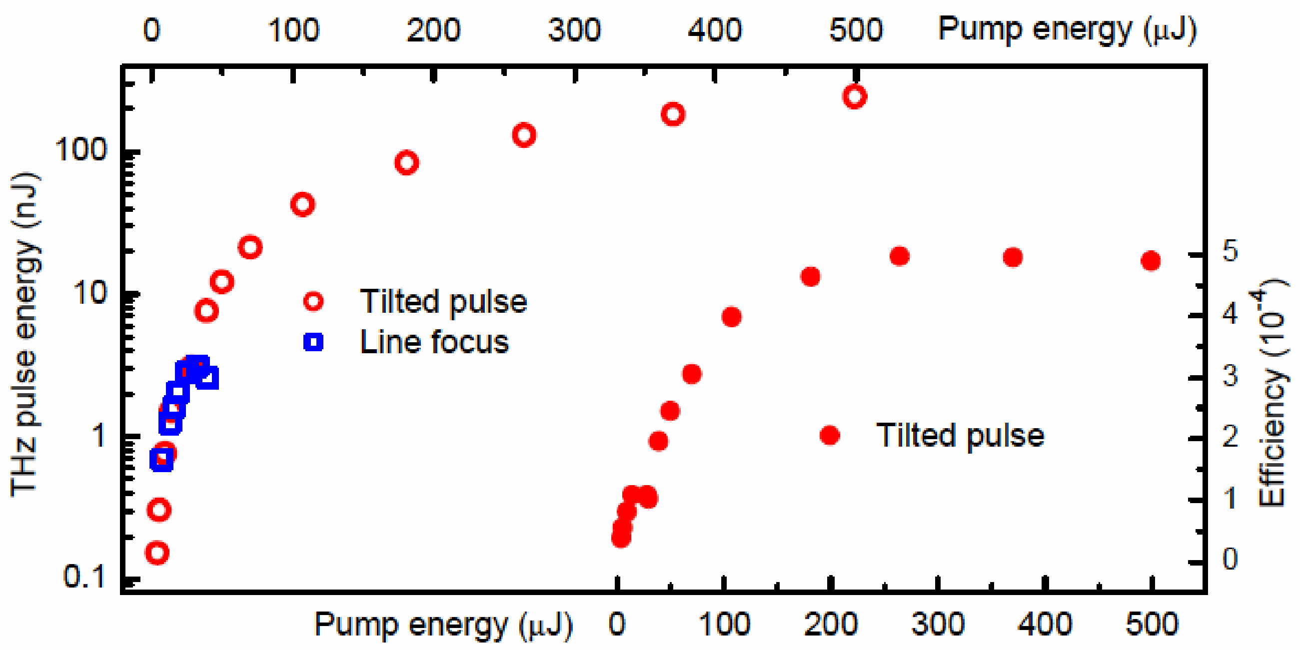


Fig. 1. Measured energy of THz pulses generated by the tilted pulse front (red circles) and line focusing (open blue squares) set-ups versus the energy of the 780 nm pump laser pulses (upper left part). Energy conversion efficiency versus the pump energy for the tilted pulse front set-up (lower right part).

Energy efficiency
5x10-4
Quantum Efficiency:
10%

P_{THz}= 100 nJ/pulse, 10 nJ/cm^2, 10 MW/ cm^2, |E_{THz}|～30kV/cm=3MV/m