Ultrafast Laser Physics

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Chapter 14: Ultrafast THz science

Goals of these lectures

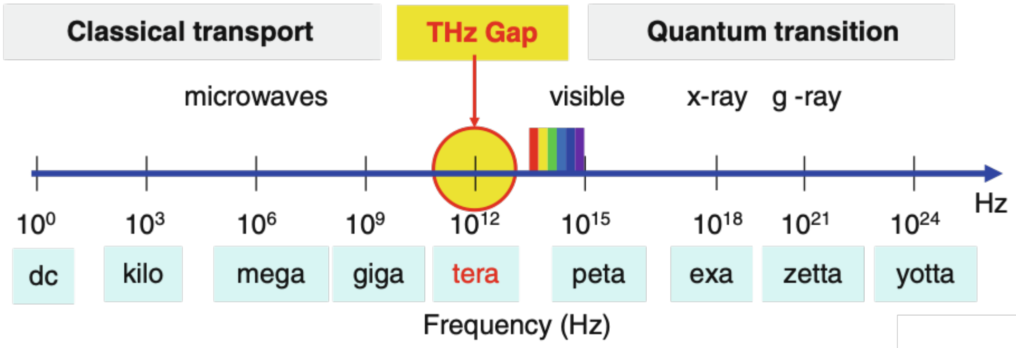
- Explain why THz radiation is relevant & its interaction with materials
- Example of applications
- Generation methods
- Detection methods

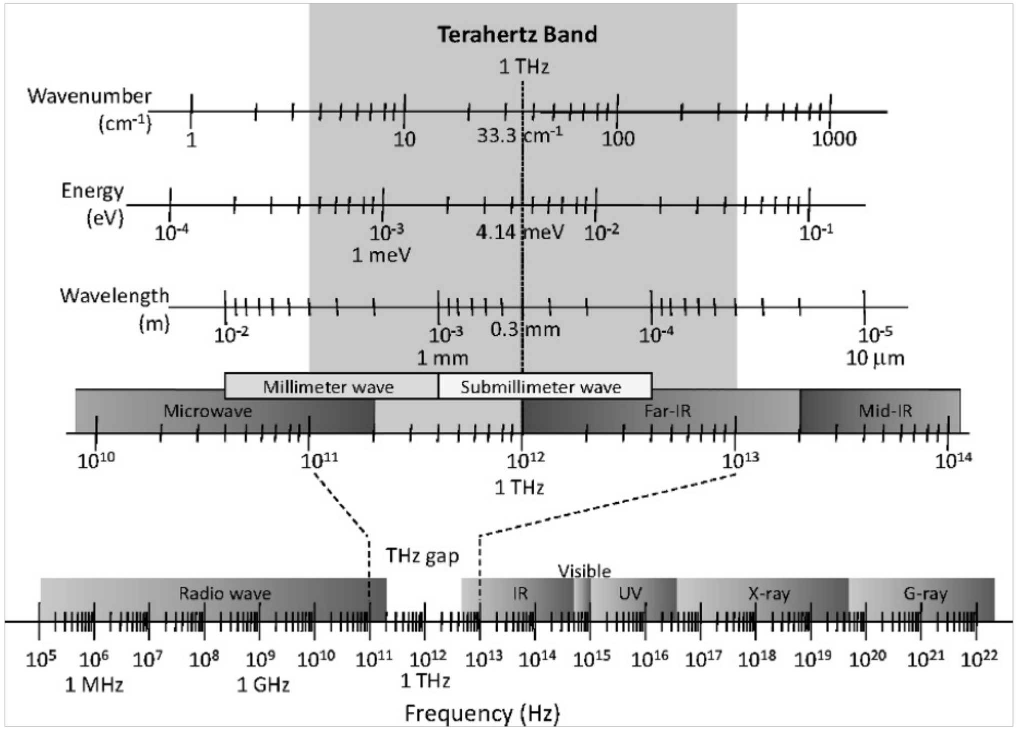
Possible follow-up classes

More in-depth courses on the subject (in case you are interested):

- Terahertz Science and Applications, starting in Fall 2024
- Ultrafast Methods in Solid State Physics, every Spring semester

THz light





Frequency: $\nu = 1$ THz $= 1000$ GHz
Angular frequency: $\omega = 2\pi\nu = 6.28$ THz
Period: $\tau = 1/\nu = 1$ ps
Wavelength: $\lambda = c/\nu = 0.3$ mm $= 300$ μm
Wavenumber: $\bar{k} = k/2\pi = 1/\lambda = 33.3$ cm^{-1}
Photon energy: $h\nu = \hbar\omega = 4.14$ meV
Temperature: $T = h\nu/k_B = 48$ K

Properties of materials in the THz range

Table 1.1. Optical Properties of Condensed Matter in the THz Band

Material Type	Optical Property
liquid water	high absorption ($\alpha \approx 250$ cm^{-1} at 1 THz)
metal	high reflectivity (>99.5% at 1 THz)
plastic	low absorption ($\alpha < 0.5$ cm^{-1} at 1 THz)
	low refractive index ($n \approx 1.5$)
semiconductor	low absorption ($\alpha < 1$ cm^{-1} at 1 THz)
	high refractive index ($n \sim$ 3-4)

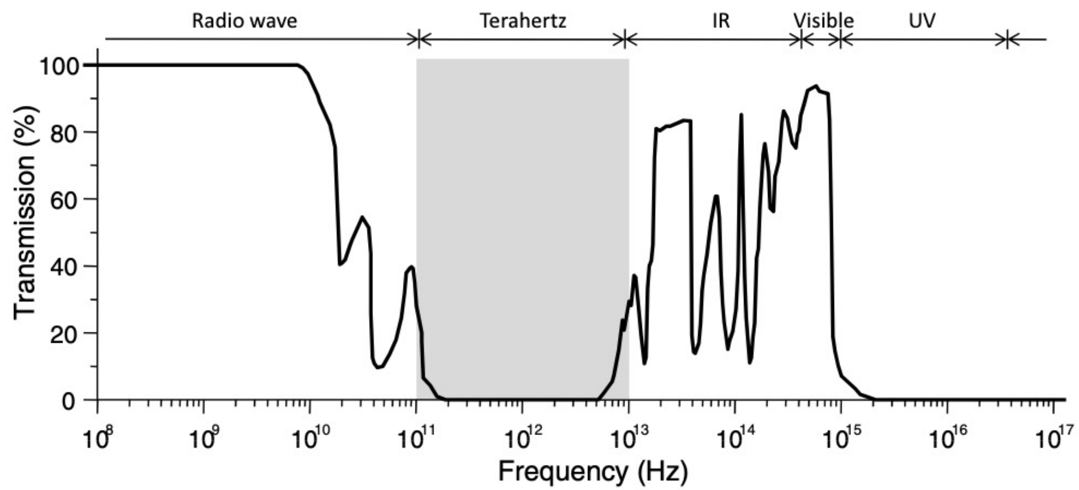


Fig. 1.6. Atmospheric transmission spectrum of electromagnetic waves

Physical phenomena at THz frequencies

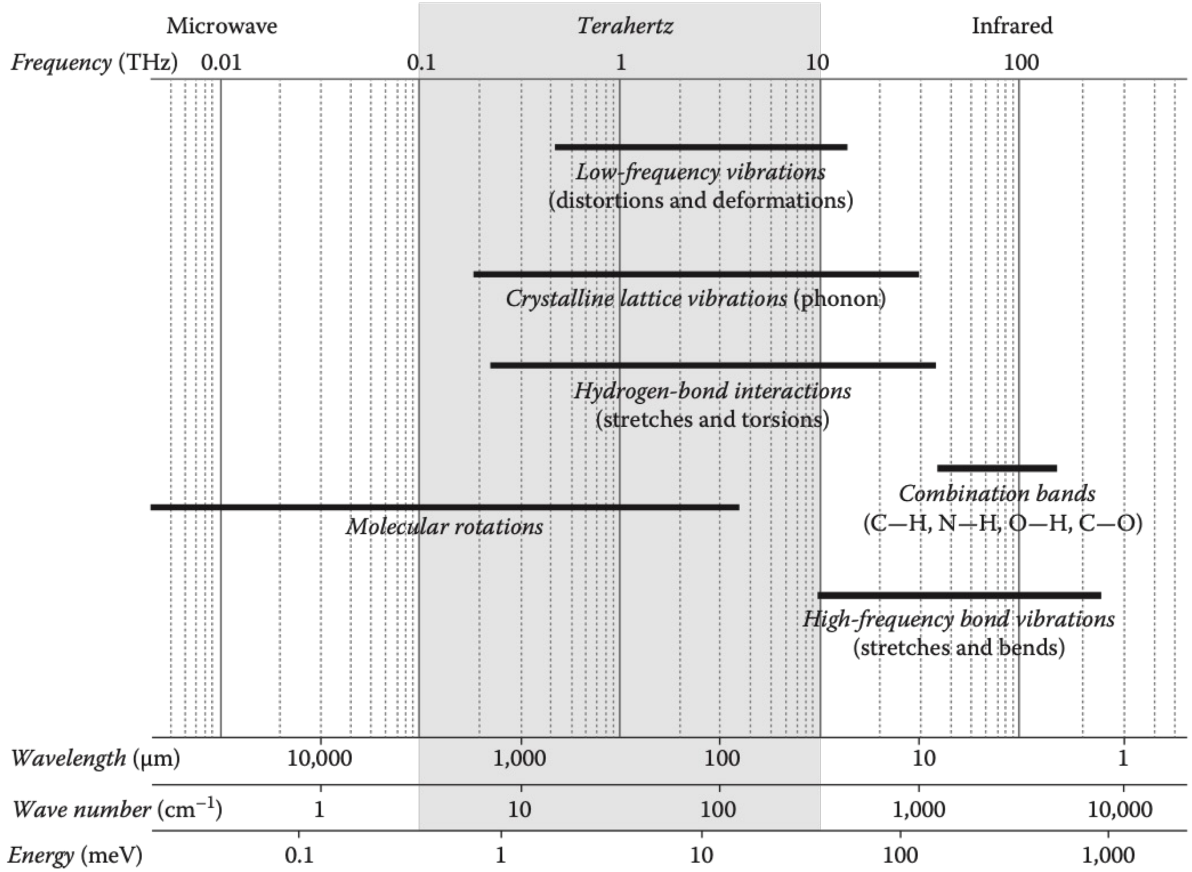


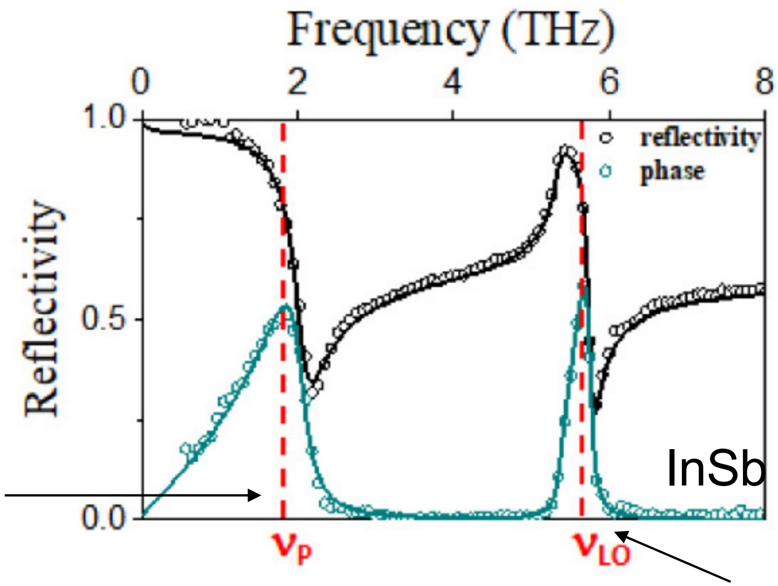
FIGURE 1.1 Characteristic energies in the electromagnetic spectrum around THz frequency region.

Electronic interaction: (semi-)conductors

Drude model with finite relaxation time

plasma frequency

$$\epsilon_r(\omega) = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\omega\gamma} = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

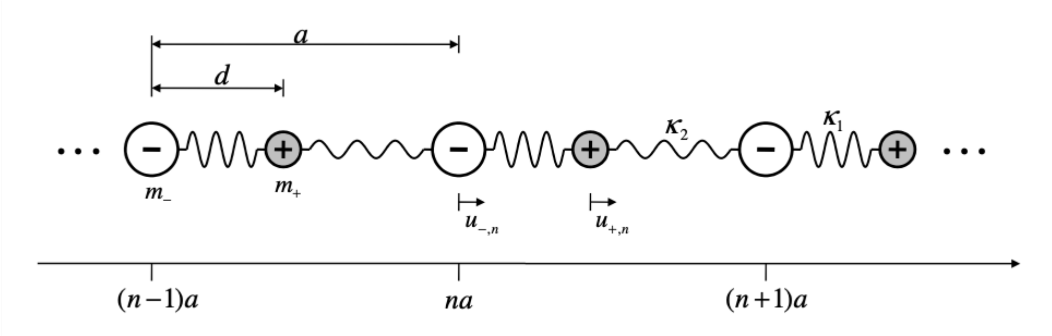


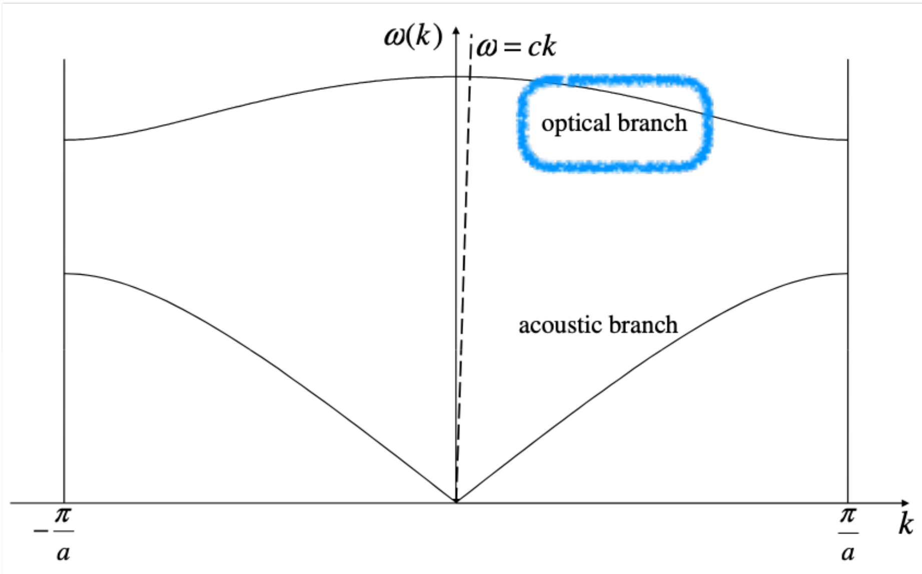
plasma frequency

phonon frequency

S. Houver et al., Opt. Express 27, 10854 (2019)

Lattice interaction in crystals: phonons





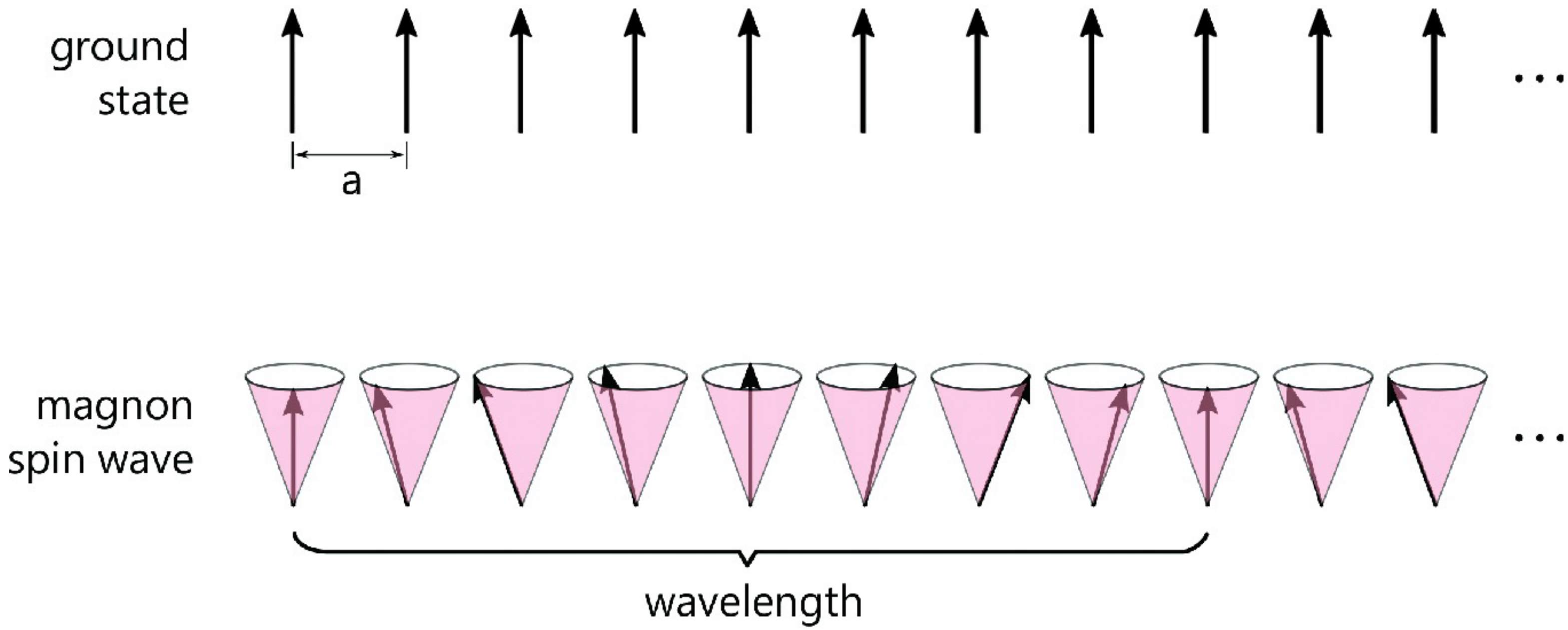
Phonon frequency in ionic crystals

Table 2.1. Transverse Optical Phonon Frequency for Ionic Crystals[a]

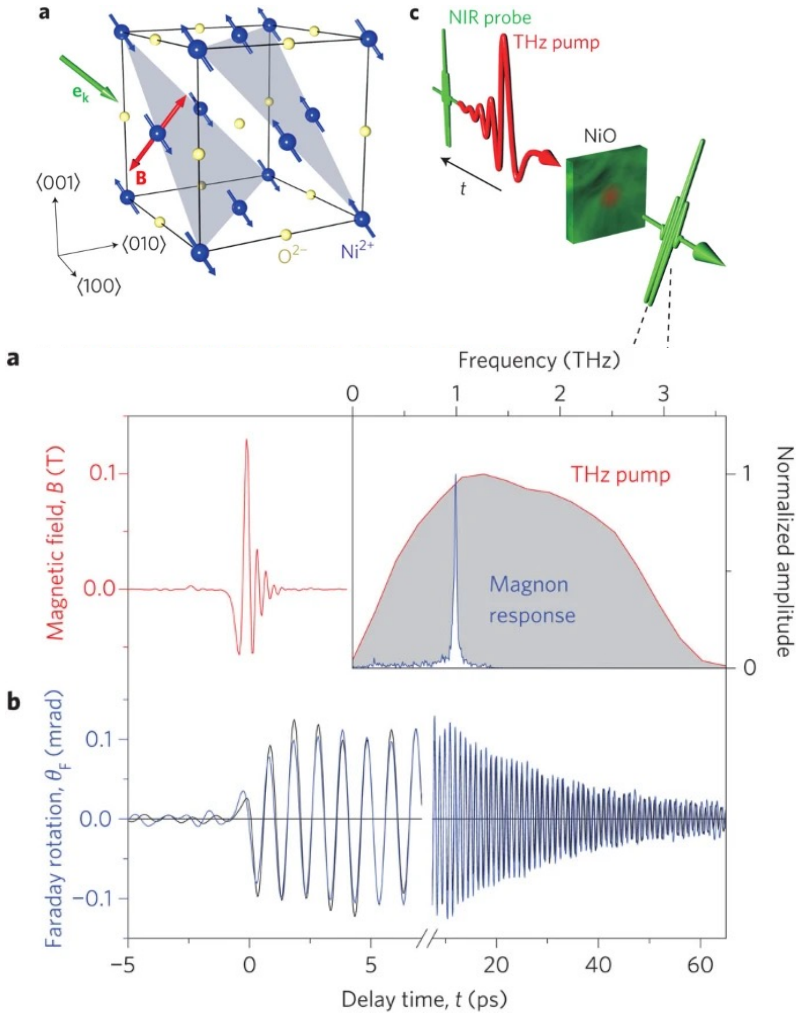
Crystal	$\omega_T/2\pi$ in THz
LiF	9.19
NaF	7.36
KF	5.70
CsF	2.60
LiCl	5.74
NaCl	5.10
KCl	4.47
CsCl	3.14
LiBr	4.76
NaBr	4.06
KBr	3.45
CsBr	2.37

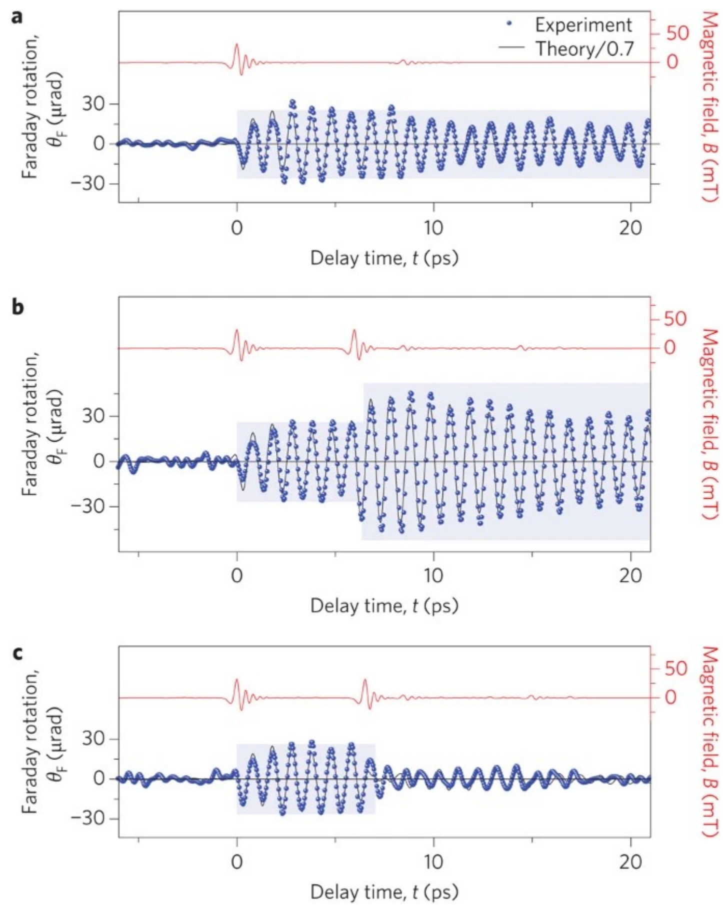
[a]Reference [11]

Magnetic excitations



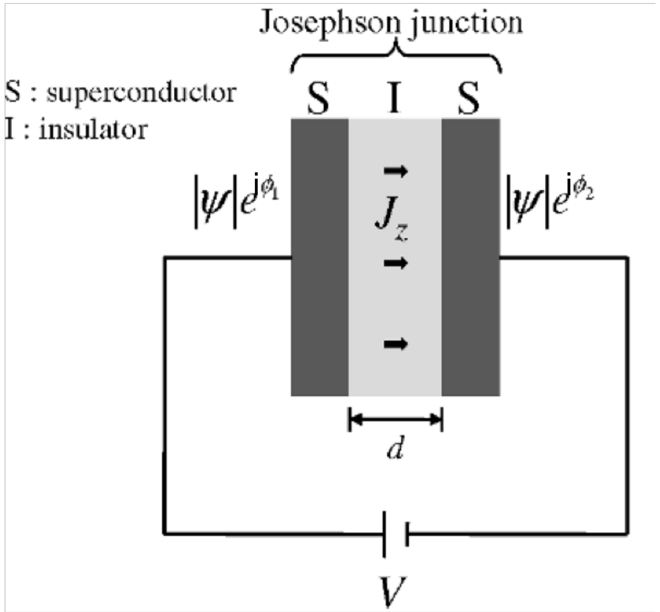
Magnetic excitations





Nature Photonics **5**, 31–34(2011)

Josephson Effect: seeing superconductivity



No bias:

$$J_z = J_0 \sin \Delta\phi$$

$$J_0 \sim \frac{e\hbar}{m^* d}|\psi|^2$$

DC bias:

$$\Delta\phi = \frac{E_1 - E_2}{\hbar}t = \frac{2eV}{\hbar}t$$

(oscillating current)

Oscillating bias:

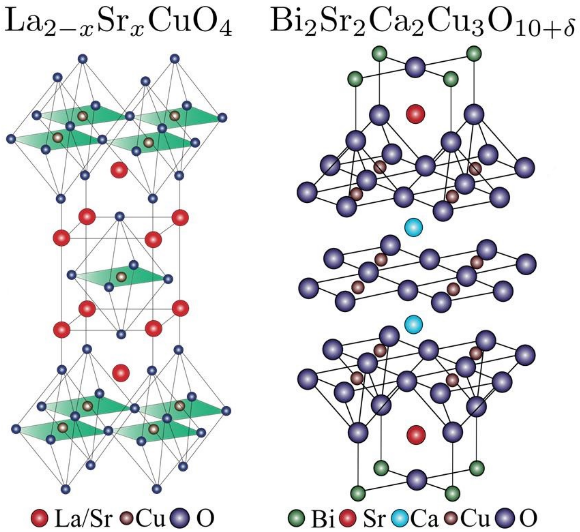
$$E_z(t) = \frac{V(t)}{d} = E_z^0 e^{-i\omega t}$$

$$\epsilon(\omega) = \epsilon_0 \left[1 - \frac{\omega_{JP}^2}{\omega^2}\right]$$

$$\omega_{JP} = \sqrt{\frac{2eJ_0 d}{\hbar\epsilon_0}} \sim \sqrt{\frac{2e^2|\psi|^2}{\epsilon_0 m^*}}$$

High temperature superconductivity

Intrinsic Josephson effect in layered cuprate superconductors at THz frequencies: plasma-like absorption below the superconducting transition



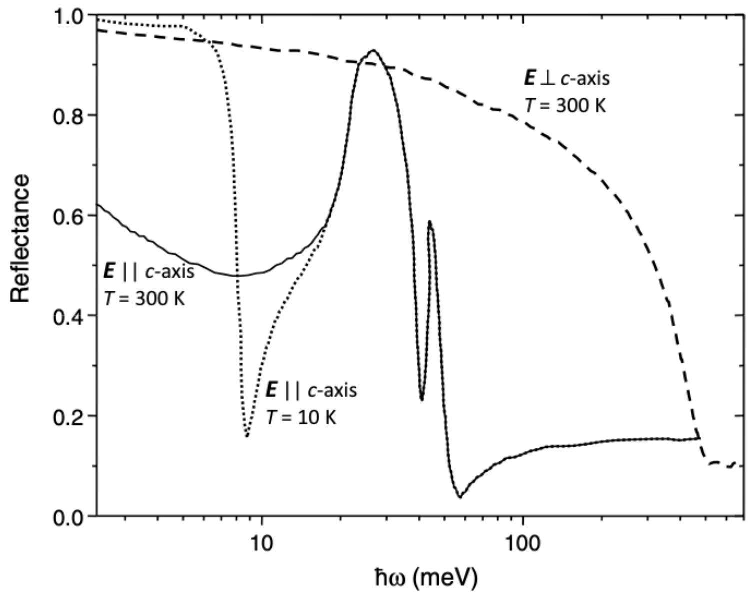
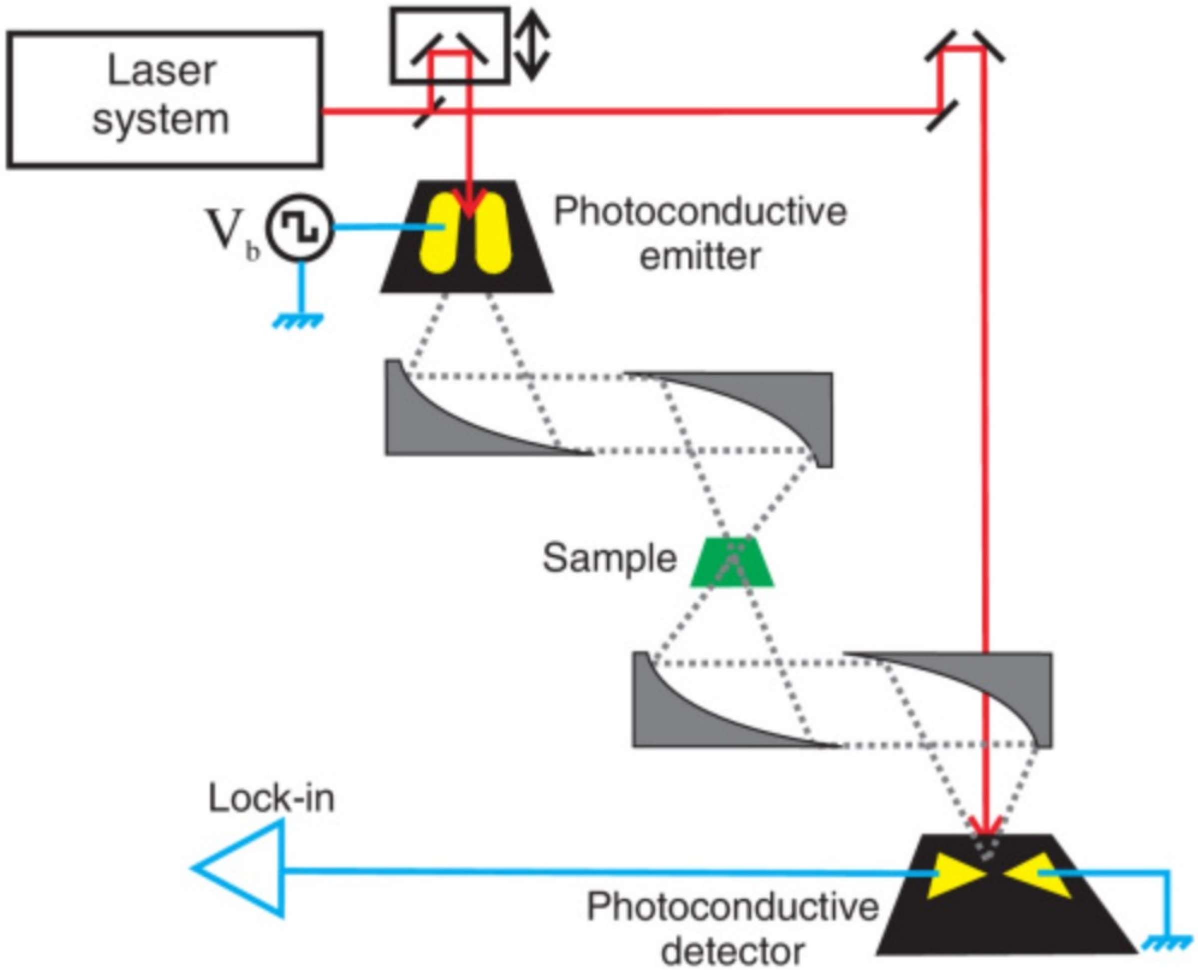


Fig. 8.22. Anisotropic reflectance spectra of a $La_{1.83}Sr_{0.17}CuO_4$ crystal. Dashed line (T = 300 K): the field is polarized in the CuO_2 plane (**E** $\perp$ c-axis). Solid (T = 300 K) and dotted (T = 10 K) lines: the field is parallel to the c-axis (**E** $\parallel$ c-axis). (Data from Ref. [246])

Generating (coherent) THz-frequency pulses

- Photoconductive Switches
- Optical Rectification in nonlinear crystals
- THz FELs
- Plasma generation

Photoconductive (or Auston) switch/emitter

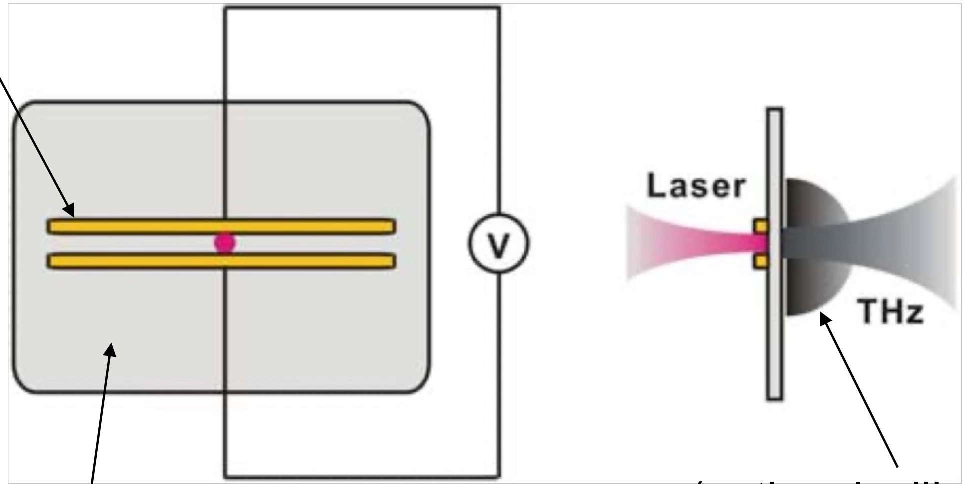


E. Castro-Camus and M. Alfaro, Photon. Res. 4, A36-A42 (2016)

Photoconductive switch

"simple dipolar antenna"

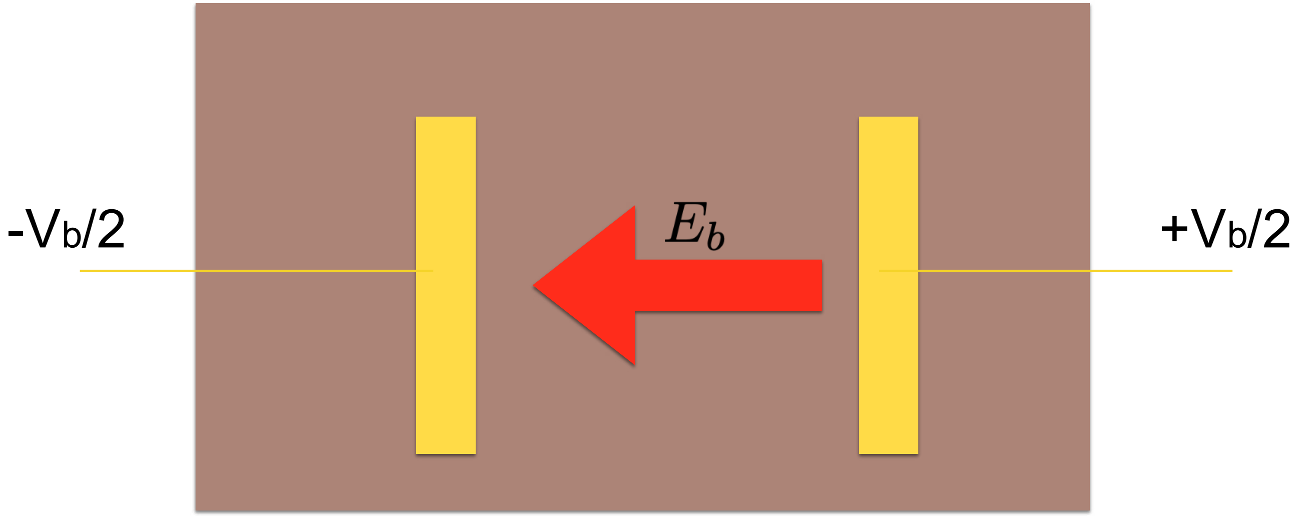
Metal electrodes on surface



(optional: silicon lens)

Semiconductor substrate, usually "low temperature grown (LT)" GaAs

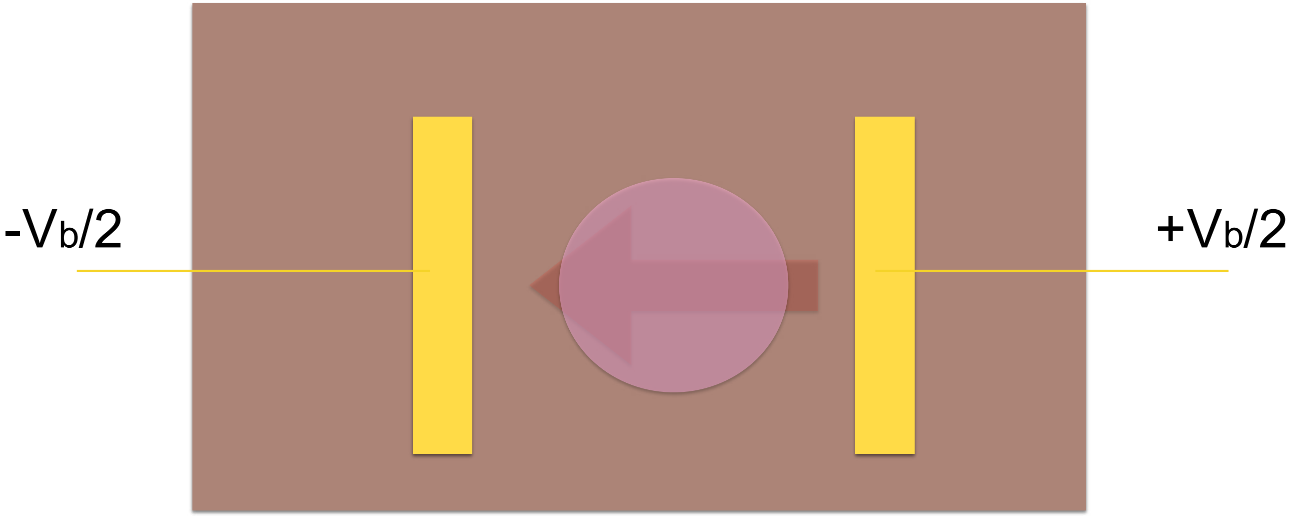
Microscopic mechanism



initially, (almost) no electron-hole pairs exist

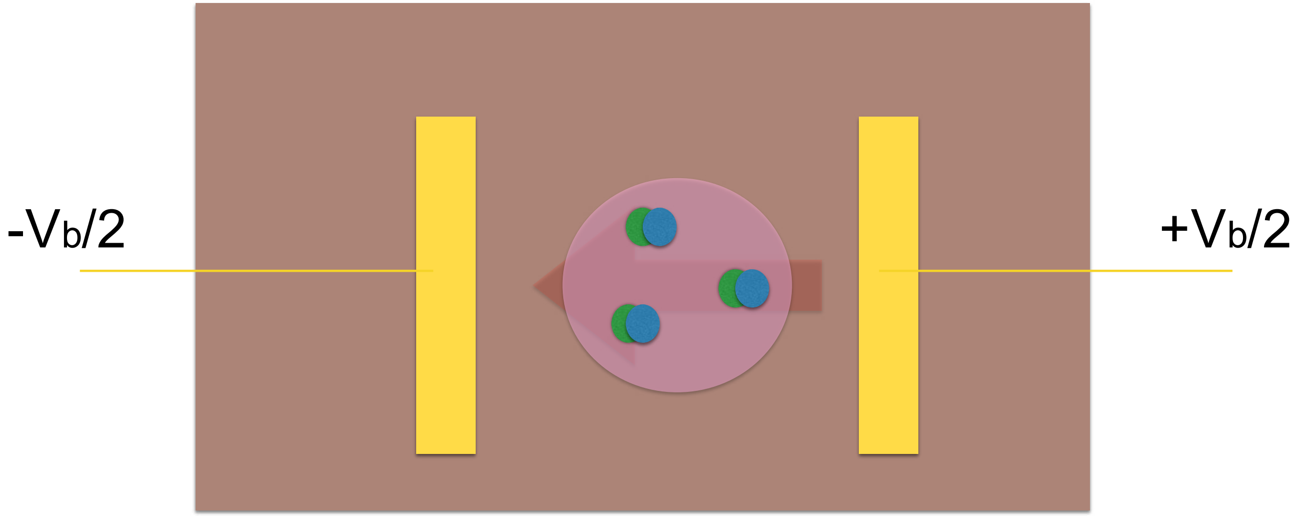
Bias field near surface

Microscopic mechanism



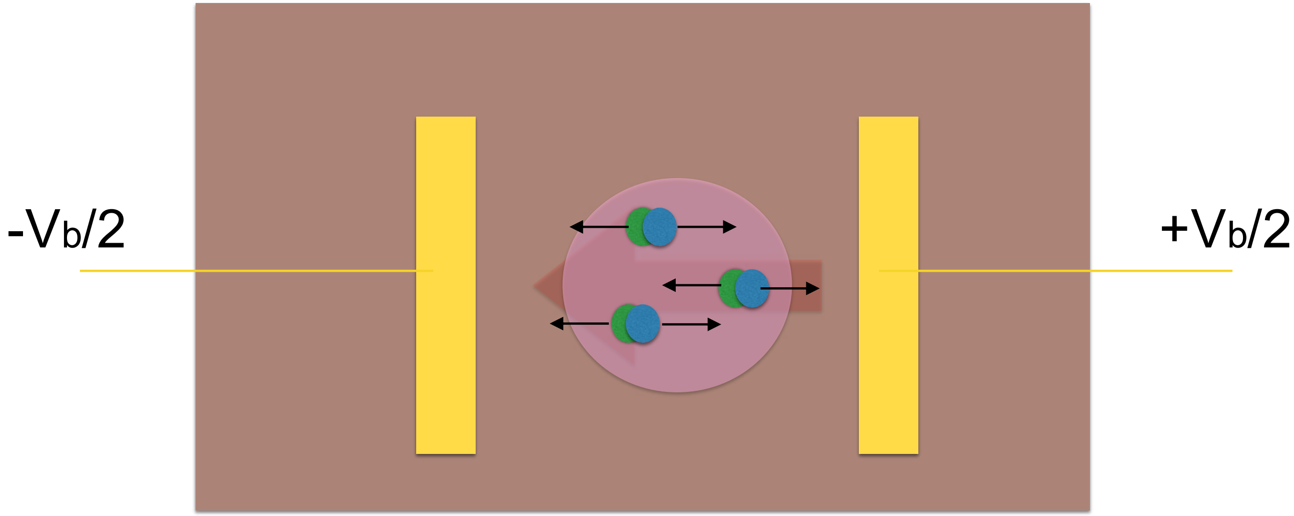
Laser pulse arrives, makes e-h pairs

Microscopic mechanism



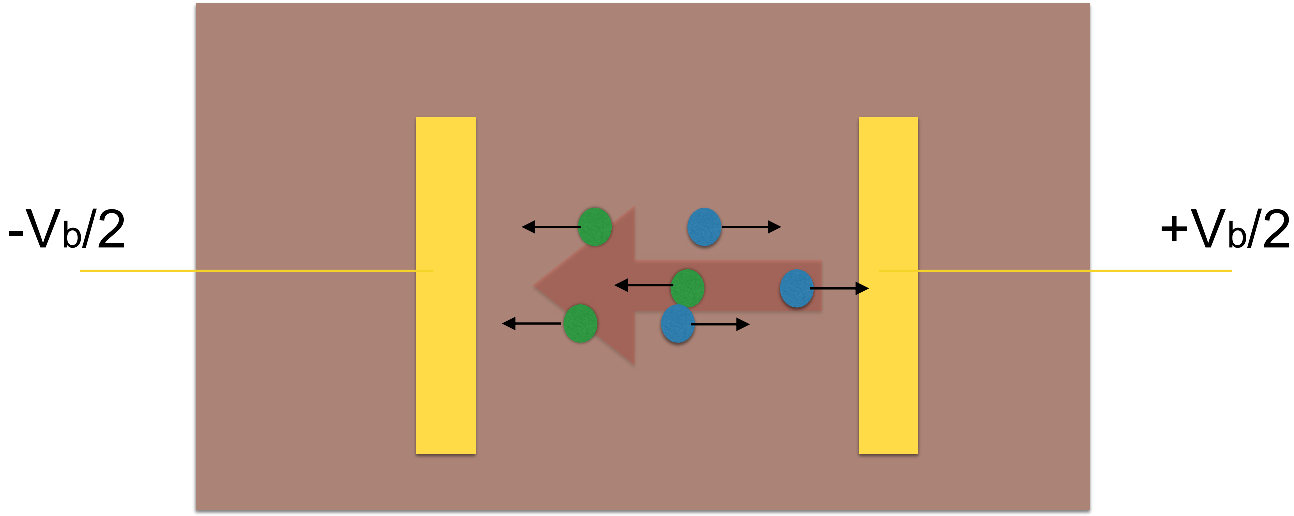
Laser pulse arrives, makes e-h pairs

Microscopic mechanism



Electrons and holes see oppositely directed forces from bias field

Microscopic mechanism



Quickly brought to drift velocity, resulting in a current

Microscopic mechanism

Assume acceleration to drift velocity is fast

$$v_d = \mu E_b$$

“mobility” is proportionality constant between E-field and drift velocity

Also assume hole mobility is much smaller than electron mobility (often true)

$$J(t) = N(t) e v_d = N(t) e \mu E_b$$

density of electrons

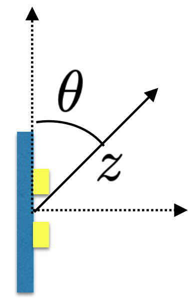
Microscopic mechanism

$$J(t) = N(t)ev_d = N(t)e\mu E_b$$

In the far field (i.e. several wavelengths away from antenna):

$$E_{THz} = \frac{1}{4\pi\epsilon_0}\frac{A}{c^2 z}\frac{\partial J(t)}{\partial t}\sin\theta$$

$$A \equiv \text{ Illuminated gap area}$$

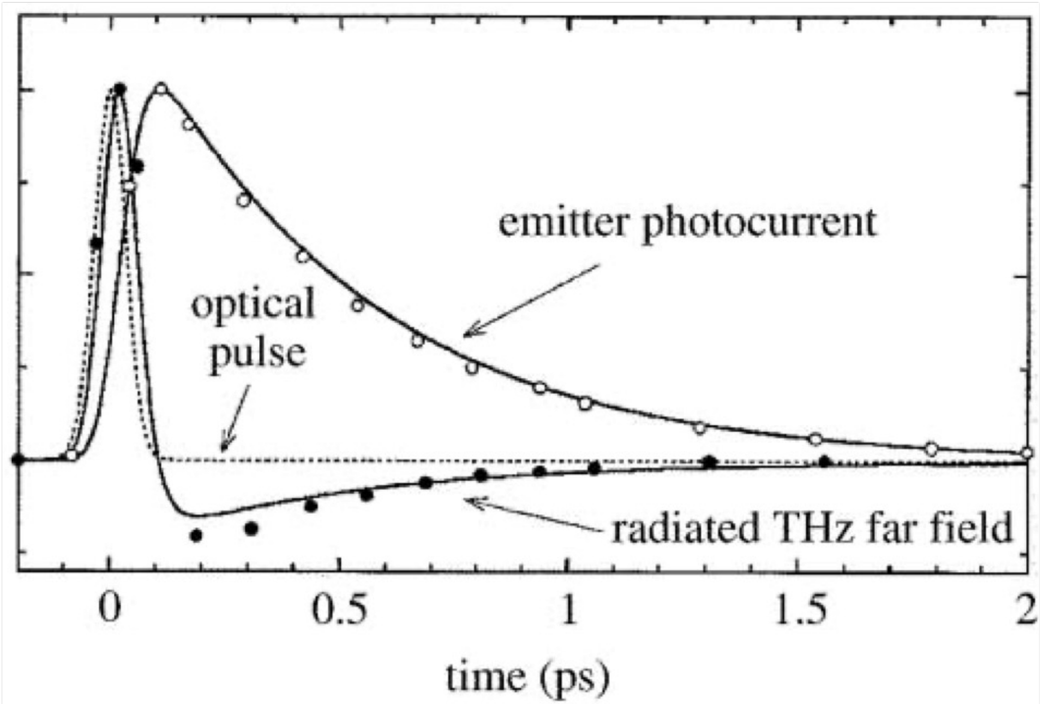


For a complete derivation please refer to "Generation and Detection of Broadband Terahertz Pulses", in Principles of Terahertz Science and Technology by Yun-Shik Lee - Springer (2009)

$$E_{THz} = \frac{Ae}{4\pi\epsilon_0 c^2 z}\frac{\partial N(t)}{\partial t}\mu E_b \sin\theta$$

Microscopic mechanism

Can also use the *decrease* in N(t) to make THz



Assumption

$$E_{THz} = \frac{Ae}{4\pi\epsilon_0 c^2 z} \frac{\partial N(t)}{\partial t} \mu E_b \sin\theta$$

Assumes “instantaneous” acceleration of electrons (holes)

Invalid on time scales comparable to
this acceleration time

Inherent contrast

Acceleration of carriers

$$\frac{1}{\gamma} = \frac{v_d m}{e E_b} = \frac{\mu m}{e}$$

Silicon at 300 K: $\frac{1}{\gamma} \sim 1 \text{ ps}$!!!

LT-GaAs: $\frac{1}{\gamma} \sim 0.1 \text{ ps}$

Difference is due to high mobility of Silicon

<u>Inherent tension</u>: higher mobility leads to slower transients, but higher currents ⇒ For photoconductive switches low temperature grown GaAs is a better THz source

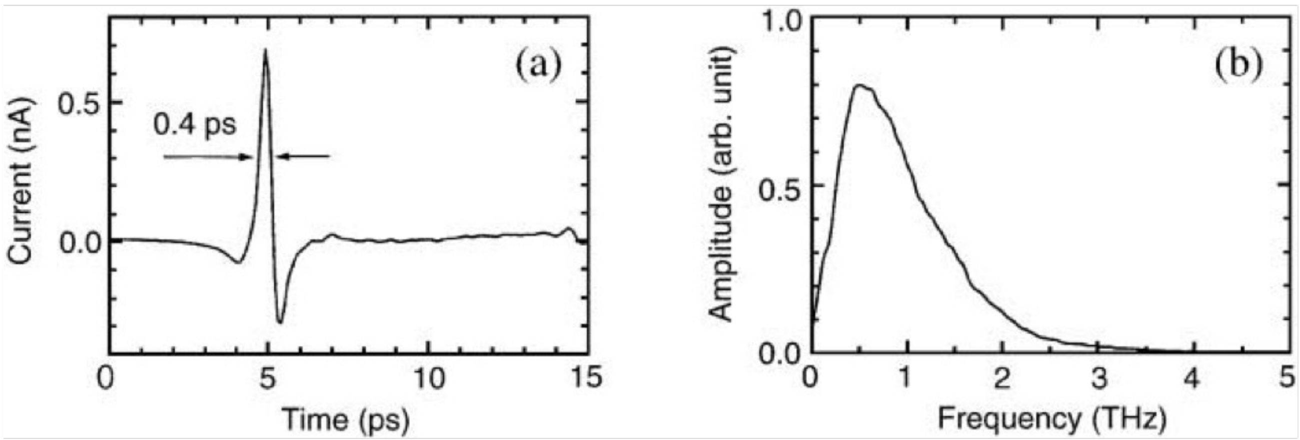
PC switch performance

At low rep rates, can get up to ~ 1 μJ

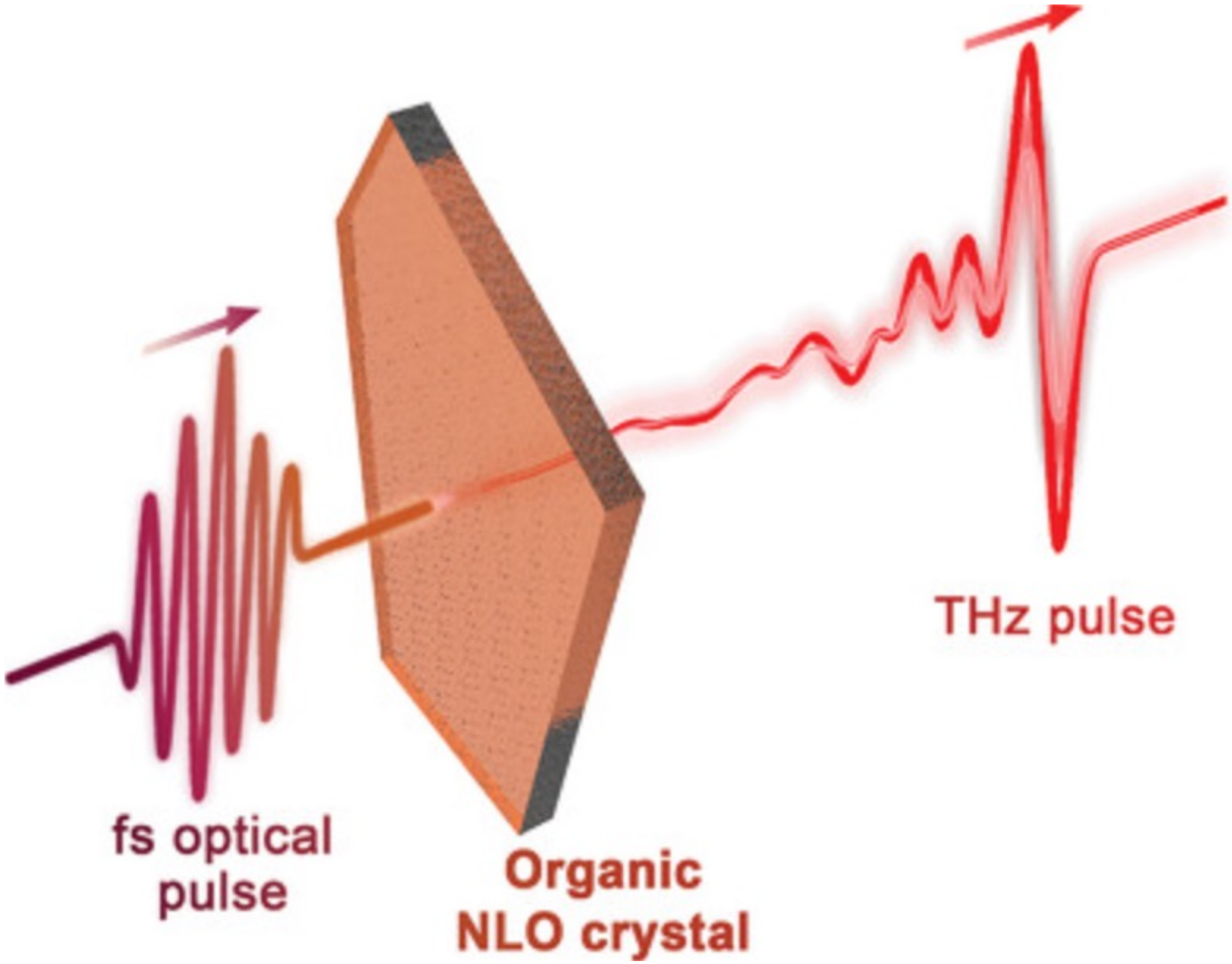
Usually limited peak fields (<10 kV/cm)

Broadband, single cycle, ~ 1 THz center freq.

Usually limited to rather low frequency region (< 4 THz)



Optical rectification

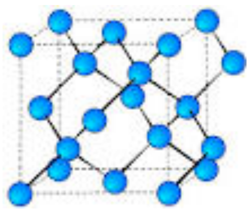


Kim, et al. *Adv. Optical Mater.* 2021, 9, 2101019

Optical rectification

$$P_i(t) = \sum_j \chi^{(1)}_{ij} E_j(t) + \sum_{jk} \chi^{(2)}_{ijk} E_j(t) E_k(t) + \dots$$

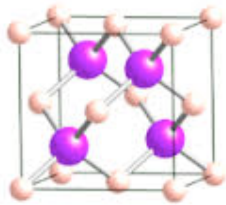
zero in inversion-symmetric materials



Diamond structure:

inversion symmetric

Diamond, Si, Ge,…



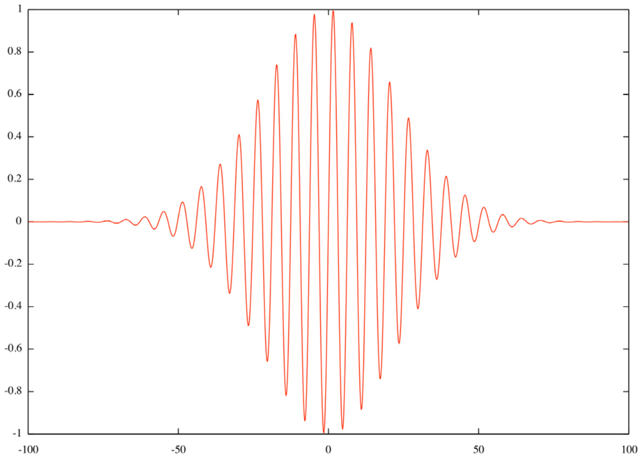
Zinc blende:

non-inversion symmetric

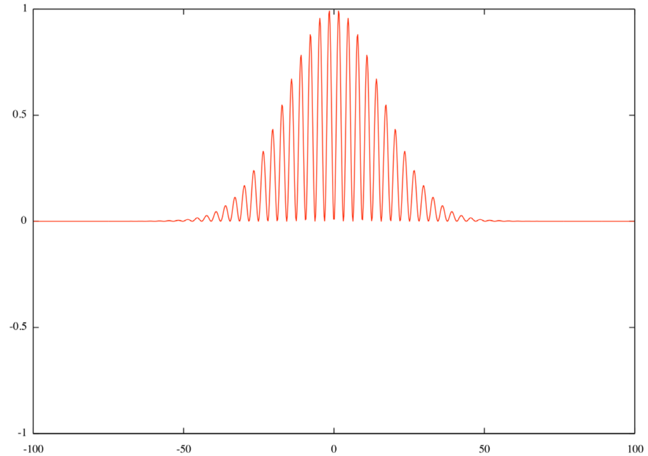
GaAs, InSb, …

Optical rectification

$$P_i(t) = \sum_j \chi^{(1)}_{ij} E_j(t) + \sum_{jk} \chi^{(2)}_{ijk} E_j(t) E_k(t) + \ldots$$



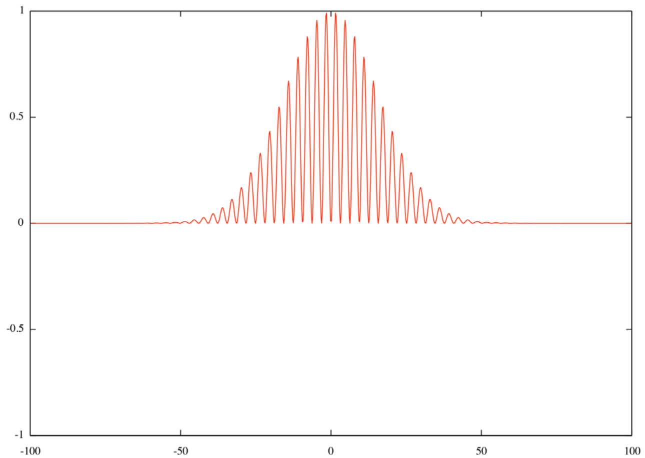
$E(t)$



$E^2(t)$

Optical rectification

$$P_i(t) = \sum_j \chi_{ij}^{(1)} E_j(t) + \boxed{\sum_{jk} \chi_{ijk}^{(2)} E_j(t) E_k(t)} + \dots$$



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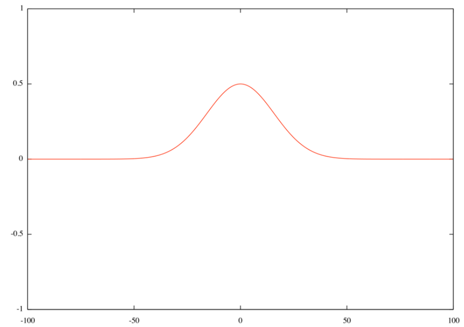
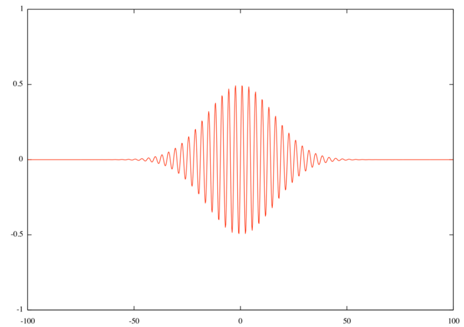


THz

(DFG)

+



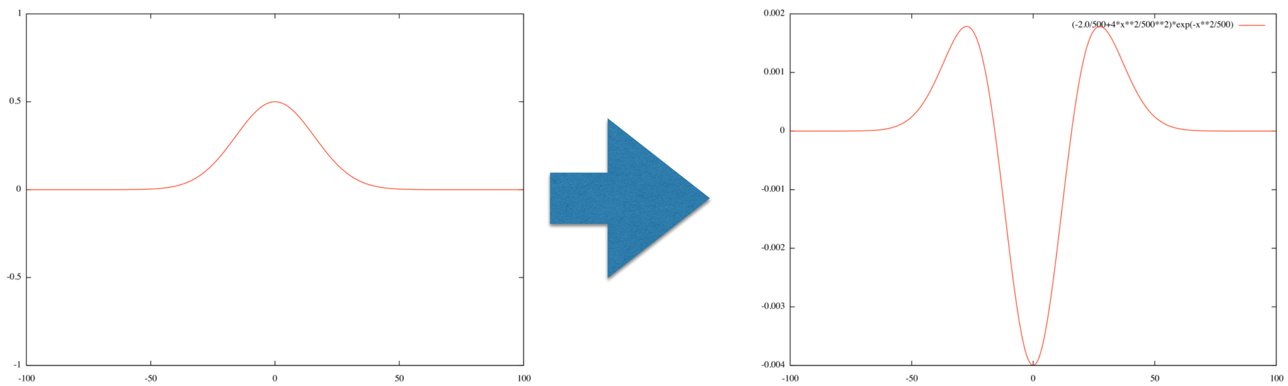
Second harmonic

(SFG)

Optical rectification

$$P_i(t) = \sum_j \chi_{ij}^{(1)} E_j(t) + \sum_{jk} \chi_{ijk}^{(2)} E_j(t) E_k(t) + \ldots$$

Far field: $$E_{THz} \sim \frac{\partial^2 P(t)}{\partial t^2}$$



Phase matching

Process is equivalent to DFG mixing for wavelengths within wavepacket

For two frequencies, DFG phase matching conditions:

$$\omega_1 - \omega_2 = \Omega_{THz}$$
$$k_1 - k_2 = k_{THz}$$

$$\frac{\omega_1 - \omega_2}{k_1 - k_2} = \frac{\Omega_{THz}}{k_{THz}} \implies \frac{\partial \omega}{\partial k} = \frac{\Omega_{THz}}{k_{THz}}$$

$$\implies v_g(\mathrm{pump}) = v_p(\mathrm{THz})$$

Phase matching

Question: what happens if phase matching conditions satisfied but

$$v_g(\mathrm{pump}) \neq v_g(\mathrm{THz}) \text{ ?}$$

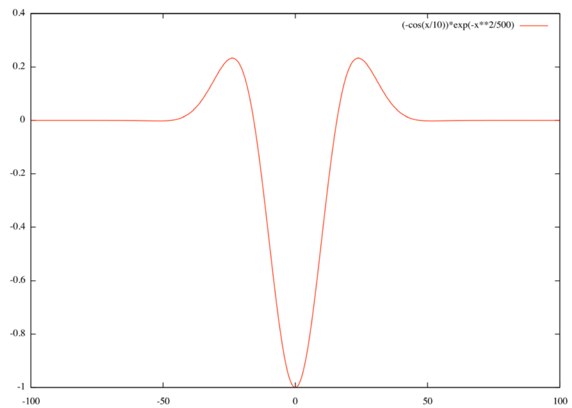
Phase matching

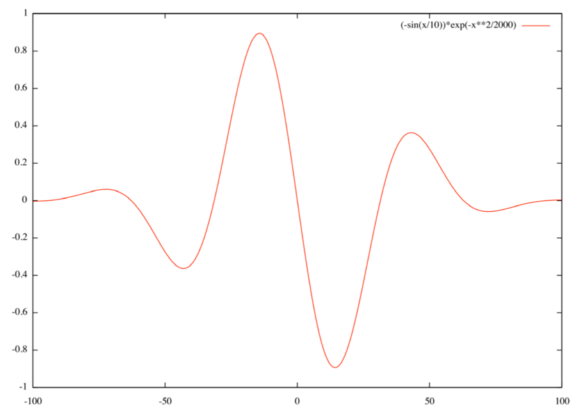
$$v_g(\mathrm{pump}) \neq v_g(\mathrm{THz})$$

$$v_g(\mathrm{pump}) = v_p(\mathrm{THz})$$

$$v_p(\mathrm{THz}) \neq v_g(\mathrm{THz})$$

carrier-envelope phase changes + pulse lengthens





Optical rectification: phase matching

$$\Longrightarrow \; v_g(\text{pump}) = v_p(\text{THz})$$

"Accidentally" true in some materials:

ZnTe and GaP at 800 nm pump

DAST, OH1, DSTMS at ~ 1.5 microns pump

Possible using refractive index ellipse in GaSe at 800 nm

Optical rectification: general requirements

- Material must be transparent at both pump and THz wavelengths
- Material cannot possess a center of inversion symmetry
- Second-order susceptibility should be high
- Phase matching over all THz frequencies in pulse (need low dispersion of permittivity); phonons will limit this

ZnTe: $\omega_{TO} \approx 5\ \mathrm{THz}$

GaP: $\omega_{TO} \approx 11\ \mathrm{THz}$

Achievable outputs with optical rectification

Inorganic compounds

- Cheap and resilient materials
- Mostly transparent for >600 nm
- Sustain high optical pumping intensity
- Phase matched at 800 nm / 1 μm (central wavelength of commercial Ti:Sa and finer lasers)
- Two-photon absorption
- Limited nonlinear tensor χ^2

Typical THz fields : 10-40 kV/cm

Typical BW: up to 8 THz (material dependent)

Organic compounds

- NOT Cheap and NOT resilient
- Sustain limited optical intensity
- Mostly transparent for >1200 nm (OPA needed)
- Phase matched >1.2 μm
- Large nonlinear tensor χ^2

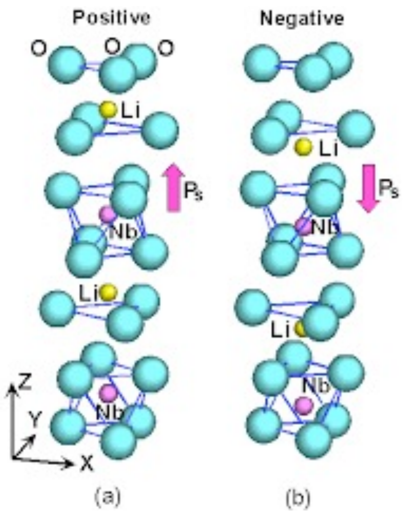
Typical THz fields : >300 kV/cm

Typical BW: up to 8 THz. Usually low frequency phonon present: efficient emitters >1.5 THz (material dependent)

How to “cheat” at phase matching

Tilted pulse front

LiNbO3



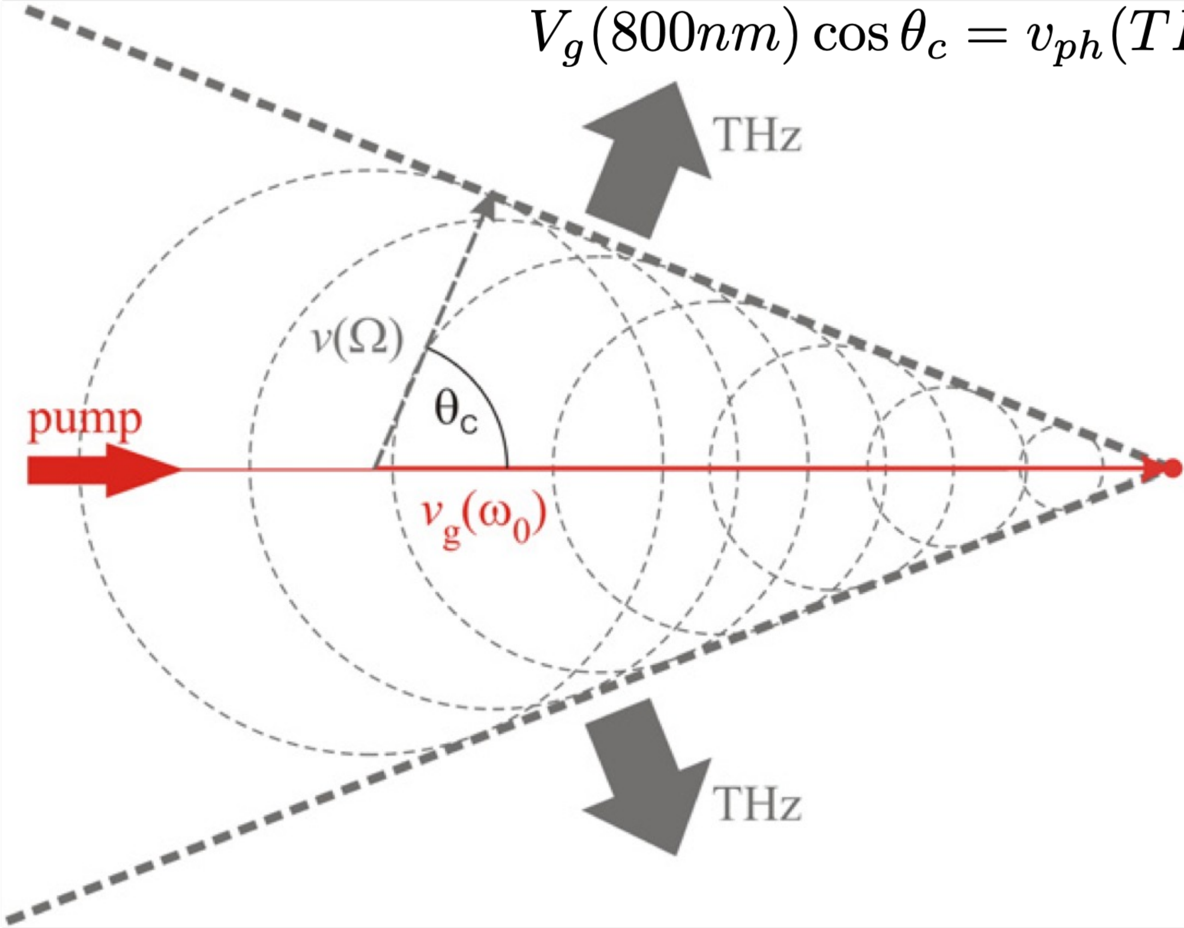
Good: robust, transparent, reasonable nonlinear coefficient

Bad: $v_{ph}(THz) \ll v_g(800nm)$
(mismatch by about a factor of 2)

How to “cheat” at phase matching

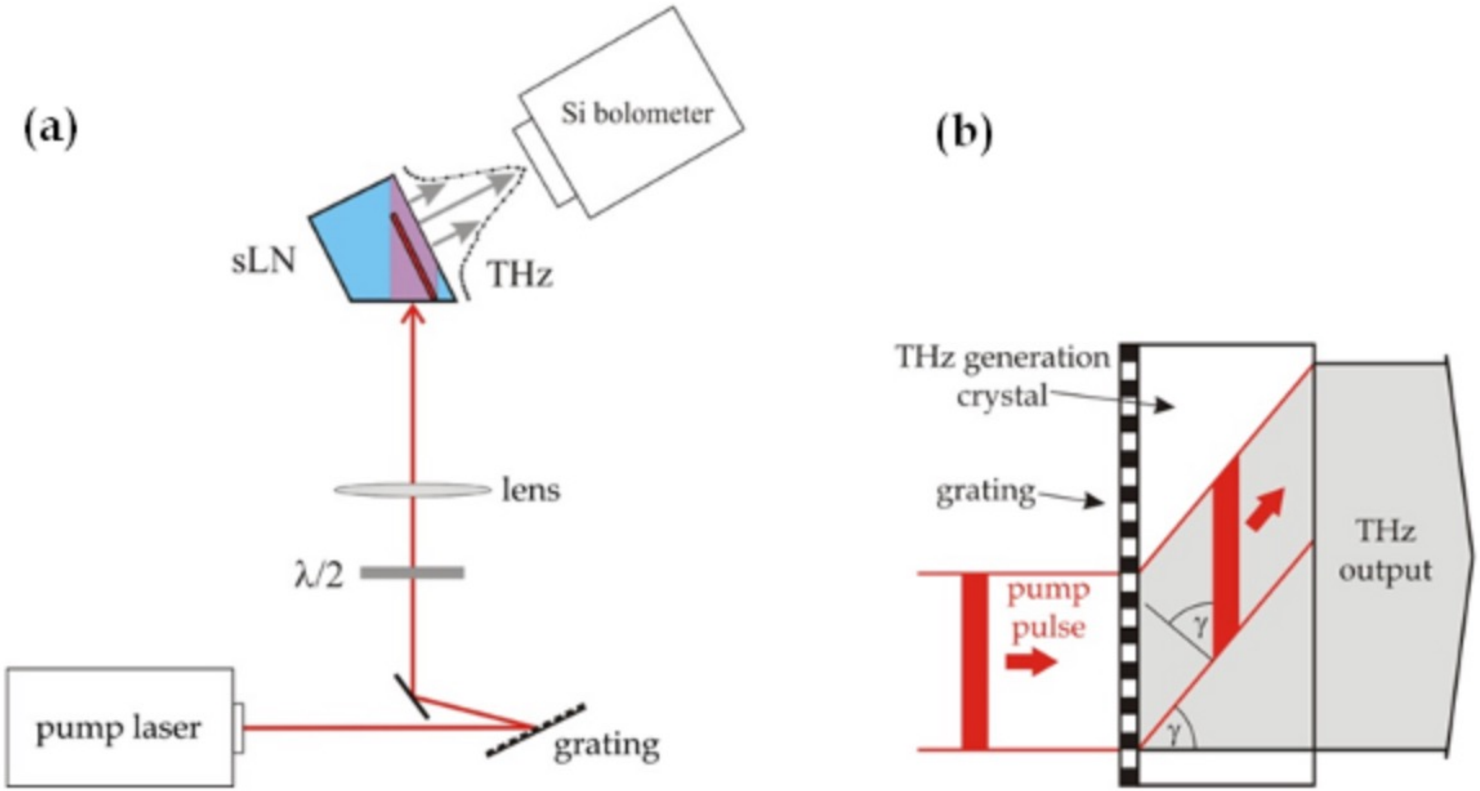
Tilted pulse front

$$V_g(800nm)\cos\theta_c = v_{ph}(THz)$$



How to “cheat” at phase matching

Tilted pulse front, via gratings



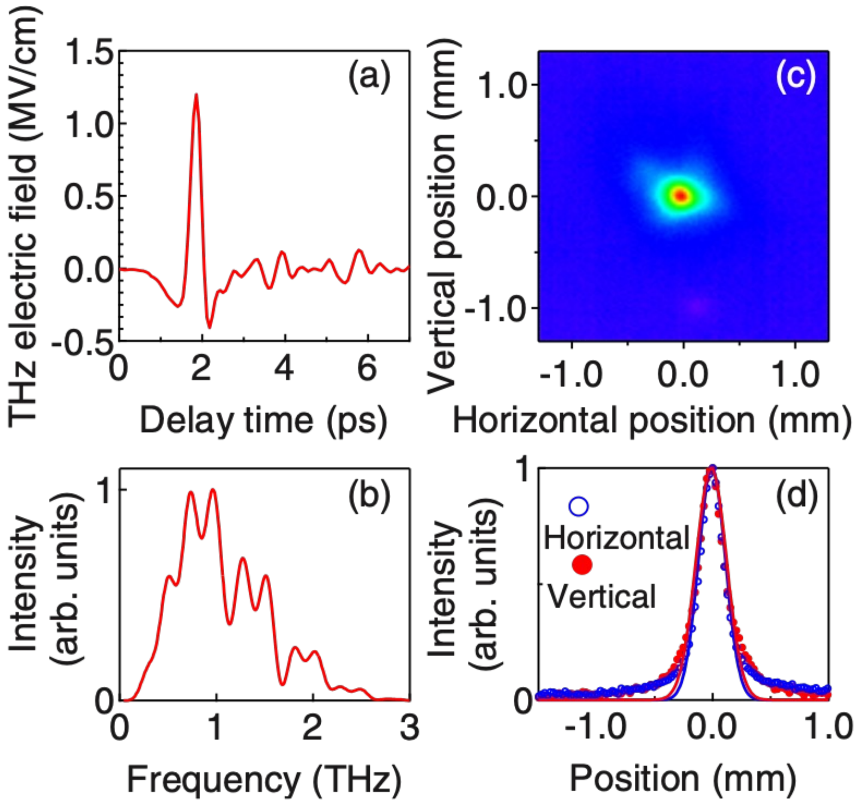
Fülöp, et al. Applications of Tilted-Pulse-Front Excitation, in *Recent Optical and Photonic Technologies*, IntechOpen 2010

How to “cheat” at phase matching

Single-cycle terahertz pulses with amplitudes exceeding 1 MV/cm generated by optical rectification in LiNbO 3

H. Hirori, A. Doi, F. Blanchard, and K. Tanaka

Citation: Applied Physics Letters **98**, 091106 (2011); doi: 10.1063/1.3560062



Nowadays >1 MV/cm routinely achieved

Rather limited BW (<2THz)

Accelerator-based THz pulse sources

- By far the most expensive option, by many orders of magnitude
- Delivers the brightest pulses

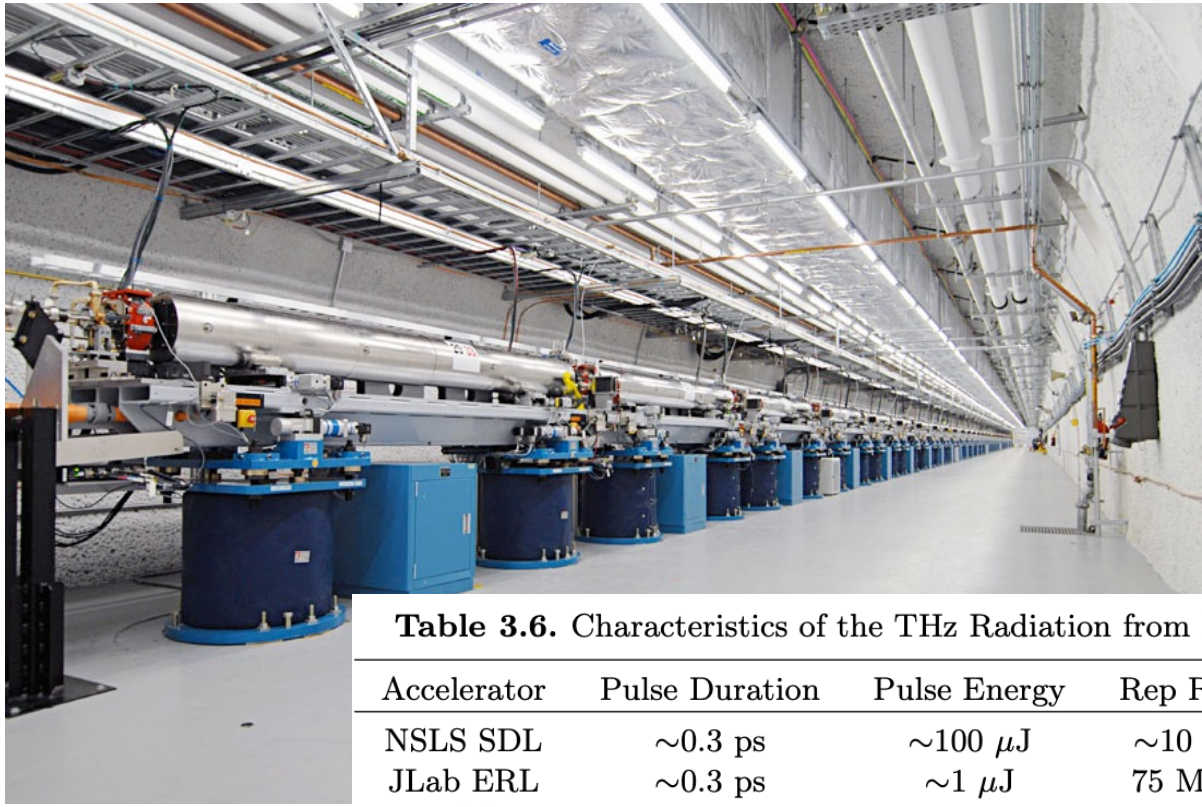


Table 3.6. Characteristics of the THz Radiation from Electron Accelerators

Accelerator	Pulse Duration	Pulse Energy	Rep Rate	Average Power
NSLS SDL	~0.3 ps	~100 μJ	~10 Hz	~1 mW
JLab ERL	~0.3 ps	~1 μJ	75 MHz	~20 W
BESSY	~1 ps	~1 nJ	500 MHz	~1 W

(from Lee, Ch.3 “Terahertz Radiation from Electron Accelerators”)

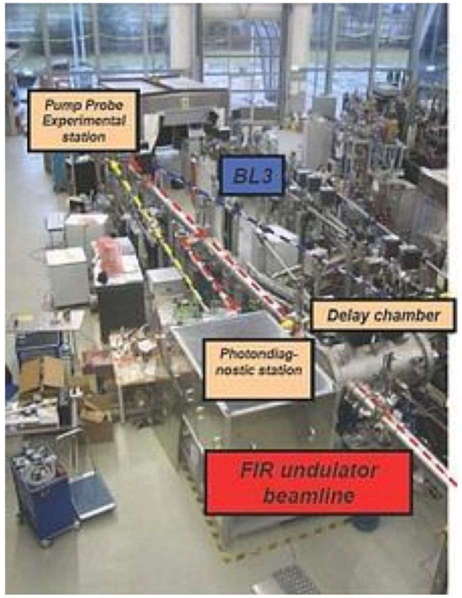
e.g FLASH at Hamburg

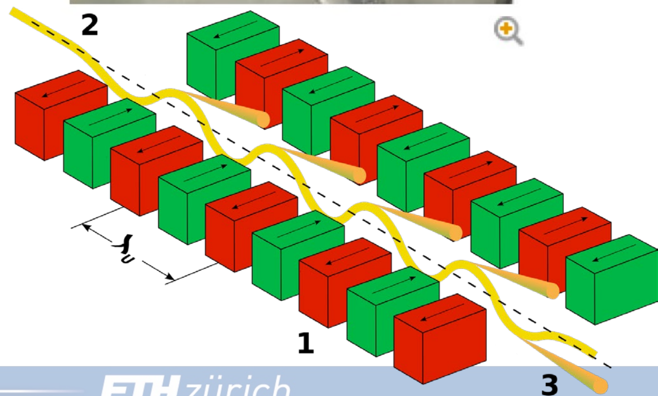
THz Undulator Beamline @ BL3

The THz undulator beamline has been designed to provide coherent femtosecond (fs) – picosecond (ps) THz pulses for pump-and-probe experiments with the fs VUV pulses from FLASH. First light has been delivered into the experimental hall in November 2007. The THz pulses are generated by a purpose built undulator implemented in series to the VUV undulators. The design of the beamline allows to overlap a VUV pulse and a THz pulse generated by the same electron bunch at the end of BL3. The pulses should therefore be naturally synchronized. The expected pulse energies lie within the microjoule regime.

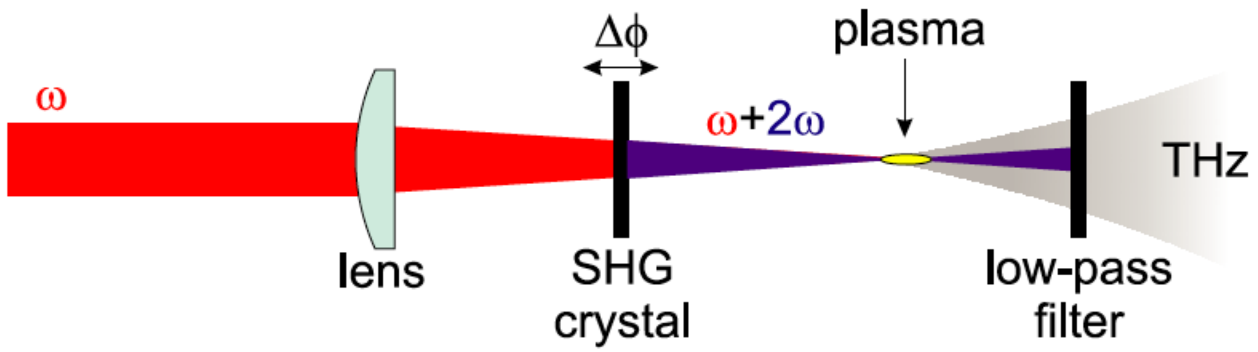
THz radiation with the following parameters is available for pump-and-probe experiments at BL3:

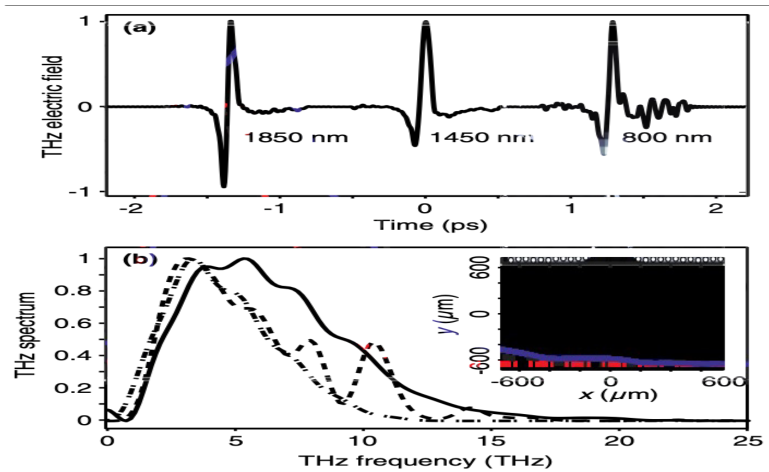
- tunable: 10--230 μm; up to 100 μJ/pulse; ~10% bandwidth,
- broadband at 200 μm, up to 10μJ/pulse; ~100% bandwidth
- synchronized and phase stable to X--ray pulses (down to 5 fs)
- delivered to the experiment via vacuum beamline as:
- (i) ultra--high vacuum (~10--8 mbar), shorter delay between THz and X--ray (~4 m path difference), can accommodate up to 0.3 m wide setup;
- (ii) high vacuum (pressure ~10--6 mbar), longer delay between THz and X--ray (~7 m path difference); can accommodate up to 2 m wide setup

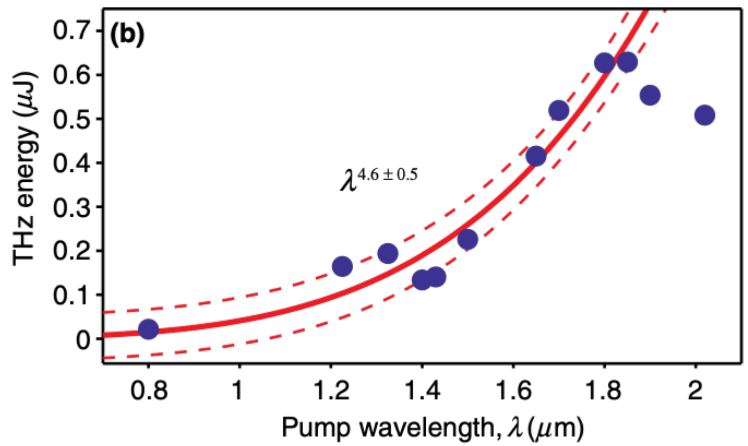




Gas-plasma generation







THz E-fields as large as 4 MV/cm, extremely broad BW (up to 15/20 THz)…

… but rather noisy/unstable, due to the intrinsic chaotic generation process

Clerici et al. Phys Rev Lett 110, 253901 (2013)

THz detectors

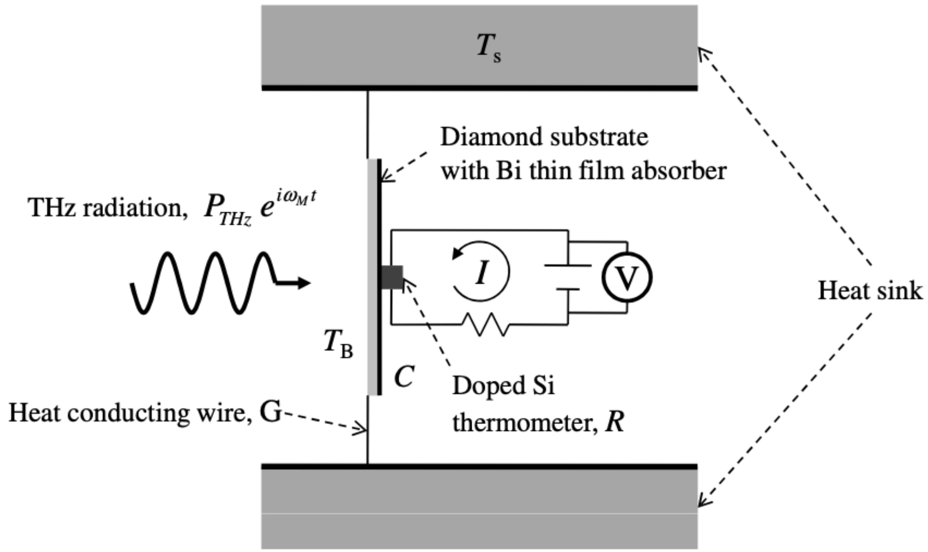
Two main types:

- Thermal detectors
  - Absorb photons, detect change in T
- Phase sensitive
  - Rely on electric-field induced changes in some material

Thermal detectors

- Absorb THz photons in reservoir that is inefficiently thermally coupled to environment
- Measure ΔT

- Advantage: fairly flat spectral response (depends on absorber)
- Disadvantage: slow, sometimes needs cooling



Phase sensitive detectors

- Measure electric field vs. time
- As name suggests, gives phase information that is lost in thermal detection
- Most examples of phase sensitive detectors look like pulse generators operated “in reverse”:
  - Photoswitches
  - EO sampling
  - ABCD detection

Photoswitch

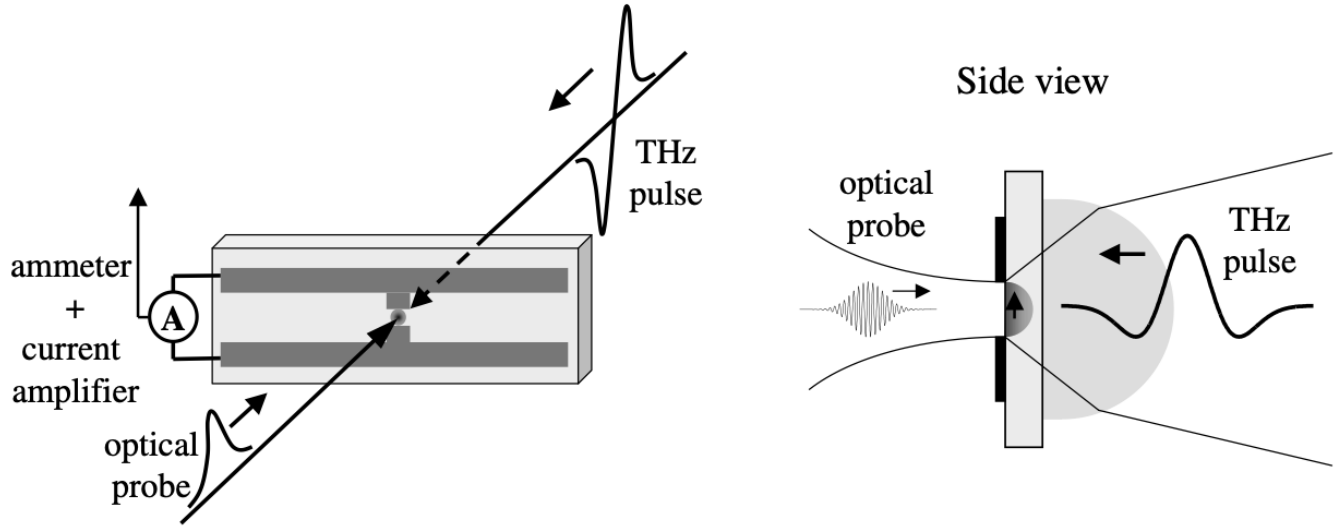


Fig. 3.24. Schematic representation of THz pulse detection with a PC antenna

- Measure E-field via current across antenna
- Similar bandwidth limitations as generator

Electro-optic sampling

Electro-optic effect: modulation of the refractive index (dielectric constant) via application of an external (slowly-varying) E-field

$$\epsilon_{ij} = \delta_{ij} + \chi^{(2)}_{ijk} E_k$$

(only possible in materials without inversion symmetry)

- Applied E-field changes dielectric tensor
- New tensor may have different symmetry
- Can measure changes to dielectric tensor induced by THz field using higher frequency light (e.g. 800 nm)

EO sampling: simple example

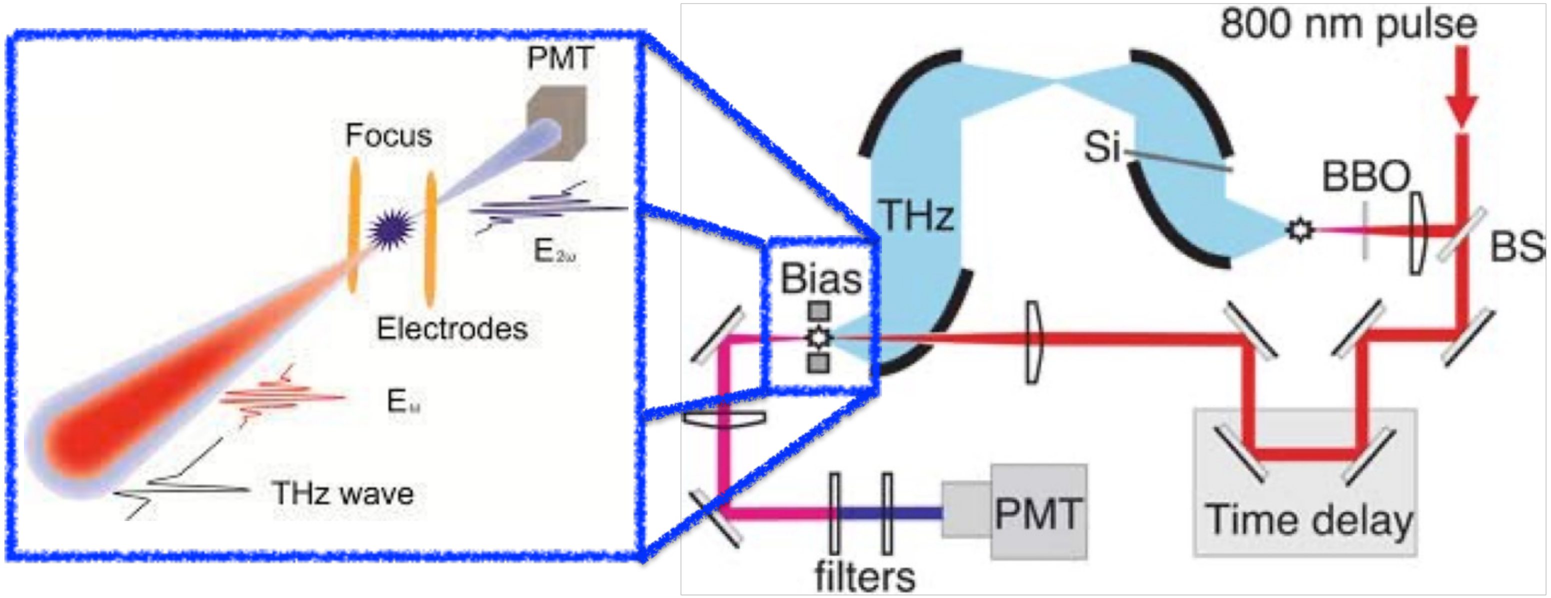
$$n_x = \sqrt{1 + a + cE_0}$$
$$n_y = \sqrt{1 + a - cE_0}$$

$$E_{opt}^{(in)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix} \longrightarrow E_{opt}^{(out)} = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{ikDn_x} \\ e^{ikDn_y} \\ 0 \end{pmatrix}$$

phase difference of DcE_0 …beam becomes slightly elliptical

ABCD (Air Break-down Coherent Detection)

- Four Wave Mixing effect where the THz E-field is used as bias on gas
- Uses third order susceptibility to generate SHG
- Very broad bandwidth, but more noisy than other methods



Jianming Dai and X.-C. Zhang, THz Wave Air Photonics