Generation and Application of Terahertz Pulses

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Introduction

- Properties of THz radiation/pulses
- Importance of THz science and technique

Cw THz sources

THz detectors, THz imaging

Single-cycle THz sources I.: Photoconductive antennae

Time-domain terahertz spectroscopy (TDTD)

Single-cycle THz sources II.: Optical rectification
  - Optical rectification II.: Tilted-intensity-front-pumping

Single-cycle THz sources III.: Laser plasma

High intensity THz science and technology

Generation and application of THz pulses with extreme high energy and field
Terahertz radiation (T-ray) in the EM spectrum
Correspondence of 1 THz frequency

- $1 \text{ THz} \div 4.15 \text{ meV}$ photon energy ($h \nu$)
- $1 \text{ THz} \div 33.3 \text{ cm}^{-1}$ wave-number
- $1 \text{ THz} \div 300 \text{ µm}$ wavelength ($\lambda$)
- $1 \text{ THz} \div 1 \text{ ps}$ temporal period ($T$)
- $1 \text{ THz} \div 48.1 \text{ K}$ temperature ($T=h \nu/k_B$)
Single-cycle THz pulse
Number of "THz publications"
THz imaging

It is based on the different transmission or reflection of different materials

Metals reflects, some dielectrics absorbs, but most plastics, paper and clothes mostly transmits THz radiation

THz imaging is applicable in security screening!

THz transmission  THz reflexion  Optical and THz images of metal, ceramic and explosive hidden into shoe heel
THz security screening
Organic molecules and crystals have specific THz absorption spectrum. Spectral "fingerprints" of explosives.

Intermolecular vibration in the unit cell of RDX crystal.
Spectral imaging

MDMA, Methamphetamine, aspirin

THz sources

Electronic and optical radiation sources. For a long time „Terahertz gap” existed

cw: FIR molecular lasers, FELs

Single-cycle THz sources are based on fs lasers
## Outline

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  - Optical rectification II.: Tilted-intensity-front-pumping
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- **Generation and application of THz pulses with extreme high energy and field**
Single-cycle (broad-band) terahertz pulse sources
I. Emitters and detectors based on photoconductive antennas

Time domain THz spectrometer (TDTS)

Based on photoconductive switches or photoconductive antennae

\[ h = R_L \frac{n}{n-1} \]

Jepsen: JOSA B 13, 2424 (1996)
The averaged photocurrent is:

\[ j(\tau) = \frac{1}{T} \int_{0}^{T} E(t) g(t + \tau) dt \]

where \( E(t) \) is the THz electric field and \( g(t) \) is the conductivity.
Measured THz pulse-shape

\[ P_{\text{laser}} = 30 \ \text{mW} \]

\[ \Delta t_{\text{laser}} = 100 \ \text{fs} \]

\[ E_{\text{THz}} = 100 \ \text{V/cm} \]

\[ I_{\text{average}} = 10 \ \text{nA} \]
... and the spectrum calculated from that

Broad spectral bandwidth

\[ \Delta v_{\text{FWHM}} \geq v_{\text{mean}} \]
Time domain terahertz spectroscopy (TDTS)

Time domain THz spectrometer (TDTS) is usually based on photoconductive switches (photoconductive antennae)

Set-up for transmission measurement

The sample is placed between the two off-axis parabolic mirrors

Time domain terahertz spectroscopy (TDTS)

But time domain THz spectrometer (TDTS) is sometimes based on optical rectification and electro-optic sampling.

Set-up for transmission measurement

Time domain terahertz spectroscopy (TDTS)

Reflection set-up

And time domain THz spectrometer (TDTS) is sometimes based on THz pulse generation and detection in air plasma

Set-up for reflection measurement

Calculation of the spectra

In both cases the $E(t)$ temporal shape of the THz pulses are measured by sampling, and the complex spectrum of the THz pulse calculated by (fast) Fourier transformation:

$$\tilde{E}(\omega) = A(\omega) \cdot e^{-i\phi(\omega)} = \int E(t) \cdot e^{-i\omega t} dt$$

The $\delta\omega$ spectral resolution is limited by the $T$ temporal scanning range:

$$\delta\omega = \frac{2\pi}{T}$$

The spectral range is limited by the $\delta t$ temporal step of sampling:

$$\Delta\omega = \frac{2\pi}{\delta t}$$

($\Delta\omega$ is decreased by the finite spectral width of the source and the bandwidth of the detection.)

In order to have a smooth calculated THz spectrum 0 padding technique (adding several 0 values to one of both side of the measured waveform) can be used.

This mathematically increases the value of $T$, resulting more dense calculated spectral points. However, the resolution will not be better.
 Calculation of the spectra

Both for transmission and reflection measurement a reference measurement is needed. In the case of transmission measurement the reference can be a measurement without the sample. In the case of reflection measurement reference is the result of a measurement with a mirror in exactly (with an accuracy of a very small fraction of the wavelength) in the place of the sample.

Jepsen: JOSA B 13, 2424 (1996)
Calculation of the spectra

Simplest case

For transmission measurement the evaluation is simplest if a thinner sample is used as a reference. In this case the effect of the Fresnel loss at the surfaces will be canceled. If the $E_S(t)$ signal and $E_R(t)$ reference waveforms are measured on a shorter temporal length than the round-trip time in the reference ($t_{RT} = 2nd/c$), the (field) absorption spectrum is given by:

$$\alpha(\omega) = \frac{1}{d} \ln \left( \frac{A_R(\omega)}{A_S(\omega)} \right)$$

and the index of refraction is given by:

$$n(\omega) = 1 + \frac{[\phi_S(\omega) - \phi_R(\omega)]c}{d \cdot \omega}$$

where $d = d_S - d_R$.

Both amplitude and phase can be measured.

Both the real and imaginary part of index of refraction and permittivity can be determined.
THz spectra
Typical spectra on the THz range

- Rotation spectral lines of small molecules
- Spectral features of molecular crystals (for example explosives)
- Dynamics of macromolecules, stretching, bending, torsion (typically featureless)
- Absorption of water (featureless)
- Solvation (featureless)
- Quasi-particles in solids: phonons, polarons, excitons, magnons, spin-waves
- Free-carrier absorption

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- Generation and application of THz pulses with extreme high energy and field
Single-cycle terahertz pulse sources II.
Optical rectification. The inverse process: Electro-optic sampling

DFG can be used for generation of THz radiation

Broad spectrum of ultrashort pump pulse contains spectral content with THz frequency difference

Optical rectification (OR)
Nonlinear optical devices

Basic theory

From Maxwell's eqs. a wave equation can be derived: \[ \nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P}{\partial t^2} \]

Consider monochr. plane wave and linear material response: \[ P(\omega) = \varepsilon_0 \chi(\omega) E(\omega) \]

\[ \frac{\partial^2 E(\omega)}{\partial z^2} - \frac{\varepsilon(\omega)}{c^2} \frac{\partial^2 E(\omega)}{\partial t^2} = 0 \quad \text{where} \quad \varepsilon(\omega) = 1 + \chi(\omega) = n(\omega)^2 \]

For nonlinear material response: \[ P(\omega) = \varepsilon_0 \left[ \chi^{(1)}(\omega) + \chi^{(2)}(\omega) E(\omega) + \ldots \right] E(\omega) \]

\[ \frac{\partial^2 E(\omega)}{\partial z^2} - \frac{n(\omega)^2}{c^2} \frac{\partial^2 E(\omega)}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P^{(2)}(\omega)}{\partial t^2} \quad (1) \]

where \[ P^{(2)}(\omega) = \varepsilon_0 \chi^{(2)}(\omega) E(\omega) E(\omega) \]

This wave equation is an inhomogeneous diff. eq. r.h.s. is responsible for wave generation
Optical rectification

\[ P^{(2)}(\omega) = \varepsilon_0 \chi^{(2)}(\omega) E(\omega) E(\omega) \quad E(\omega, t) = A \cos(\omega t - k z) \]

\[ P^{(2)} = \varepsilon_0 \chi^{(2)} A^2 \left[ \cos(\omega t - k z) \right]^2 = \frac{1}{2} \chi^{(2)} A^2 \left[ 1 + \cos(2\omega t - k z) \right] \]

\[ E(\omega, t) = A_1 \cos(\omega_1 t - k_1 z) + A_2 \cos(\omega_2 t - k_2 z) \]

\[ P^{(2)} = \varepsilon_0 \chi^{(2)} \left\{ A_1^2 \left[ \cos(\omega_1 t - k_1 z) \right]^2 + A_2^2 \left[ \cos(\omega_2 t - k_2 z) \right]^2 \right\} + \]

\[ + \varepsilon_0 \chi^{(2)} \left\{ 2 A_1 A_2 \cos(\omega_1 t - k_1 z) \cdot \cos(\omega_2 t - k_2 z) \right\} = \]

\[ = \varepsilon_0 \chi^{(2)} \left\{ \ldots 1 \ldots + \ldots 2 \ldots + A_1 A_2 \cos((\omega_1 + \omega_2) t - (k_1 + k_2) z) \right\} + \quad \text{SFG} \]

\[ + \varepsilon_0 \chi^{(2)} \left\{ A_1 A_2 \cos((\omega_1 - \omega_2) t - (k_1 - k_2) z) \right\} \quad \text{DFG, „OR”, THz generation} \]

\[ \omega_{THz} = \omega_1 - \omega_2 \]
Optical rectification

Using ultrashort laser pulses for excitation: \( E(\omega, t) = A(t) \cos (\omega t - kt) \)

\[
P^{(2)} = \varepsilon_0 \chi^{(2)} \left\{ 1 + 2 + A^2(t) \cos((\omega_1 + \omega_2)t - (k_1 + k_2)z) \right\} + \\
+ \varepsilon_0 \chi^{(2)} A^2(t) \cos((\omega_1 - \omega_2)t - (k_1 - k_2)z)
\]

Single-cycle THz pulse

Spectral content is determined by:

- spectral content of pump pulse
- phase-matching bandwidth
- THz absorption

Effect of dispersion in THz range
Optical rectification

That part of the nonlinear polarization which responsible for THz pulse generation is:

\[ P_{THz}^{(2)} = \varepsilon_0 \chi^{(2)} A^2(t) \cos\left( (\omega_1 - \omega_2) t - (k_1 - k_2) z \right) \]

THz pulse:

\[ E_{THz}(\omega_{THz}, t) = A(t) \cos \left( \omega_{THz} t - k_{THz} t \right) \quad \omega_{THz} = \omega_1 - \omega_2 \]

For constructive THz generation \( k_{THz} = k_1 - k_2 \) is needed →

→ Velocity matching (phase matching) is needed:

\[ n_p^{gr} = n_{THz} \]

Without velocity matching generation only at the two surfaces of the nonlinear crystal

Example: generation in 0.5 mm LiNbO₃

**Optical rectification**  
**Velocity matching**

Isotropic crystals: only possibility (if any) for velocity matching is choosing appropriate pump and/or THz wavelength (frequency)

![Graph showing refractive indices of ZnTe](image)

Refractive indices of ZnTe

\[
 n_p^2(\lambda) = 4.27 + \frac{3.01 \cdot \lambda^2}{\lambda^2 - 0.142} \\
 n_{THz}^2(\nu) = \frac{289.27 - 6 \cdot \nu^2}{29.16 - \nu^2}
\]

A. Nahata et al., Appl. Phys. Lett. 61, 2321 (1996)

Anisotropic (birefrigent) crystal: velocity matching by angle tuning

Example: GaSe (see lecture 3)

Other phase/velocity matching methods:
- Cherenkov (small spot size needed → limited THz energy)
- Waveguide (large dispersion, practically no single-cycle generation)
- Tilted-pulse-front pumping (LiNbO$_3$, see lecture 8)
- Quasi-phase matching (no velocity matching, no single-cycle generation, as many cycles as the number of periods in the crystal)
**Optical rectification**

**Efficiency**

In the case of velocity matching and no dispersion

\[
\eta_{THz} = \frac{2\omega^2 d_{eff}^2 L^2 I}{\varepsilon_0 n_v^2 n_{THz} c^3} \cdot \exp\left[\frac{-\alpha_{THz} L}{2}\right] \cdot \frac{\sinh^2\left[\frac{\alpha_{THz} L}{4}\right]}{\left[\frac{\alpha_{THz} L}{4}\right]^2}
\]

Using the contracted matrix

\[
d_{il} = \frac{1}{2} \chi^{(2)}_{ijk}
\]

where \( l = 1 \), \( 2 \), \( 3 \), \( 4 \), \( 5 \), \( 6 \)

for \( jk = 11 \), \( 22 \), \( 33 \), \( 23,32 \), \( 31,13 \), \( 12,21 \)

For \( \alpha_{THz} L \ll 1 \)

\[
\eta_{THz} = \frac{2\omega^2 d_{eff}^2 L^2 I}{\varepsilon_0 n_v^2 n_{THz} c^3}
\]

For \( \alpha_{THz} L \gg 1 \)

\[
\eta_{THz} = \frac{2\omega^2 d_{eff}^2 I}{\varepsilon_0 n_v^2 n_{THz} \alpha_{THz}^2 c^3}
\]
### Optical rectification

FOMs of a few crystals supposing 1 mm crystal length and 1 THz

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<tr>
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<td>3.73</td>
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<td>4.8</td>
<td>2.10</td>
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<tr>
<td>GaAs</td>
<td>4.18</td>
<td>3.61</td>
<td>0.5</td>
<td>0.78</td>
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Electro-optic effect

Electro-optic effect $\approx$ Pockels effect

**Pockels effect:** static electric field induced change of refractive index (birefrigence)

Pockels effect is used for example for Q-switching of lasers

$$P_i^{(2)}(\omega) = 2 \sum_{j,k} \varepsilon_0 \chi^{(2)}_{ijk}(\omega, \omega, 0) E_j(\omega) E_k(0) = \sum_j \varepsilon_0 \chi^{(2)}_{ij}(\omega) E_j(\omega) \quad \text{Eq. EO}$$

where $\chi^{(2)}_{ij} = 2 \sum_k \chi^{(2)}_{ijk}(\omega, \omega, 0) E_k(\omega)$

Use Pockels effect for the quasi-static field of THz pulse: $0 \rightarrow \omega_{THz}$

Optical rectification:

$$P_i^{(2)}(\omega_{THz}) = \sum_{j,k} \varepsilon_0 \chi^{(2)}_{ijk}(\omega_{THz}, \omega, \omega - \omega_{THz}) E_j(\omega) E^*_k(\omega - \omega_{THz})$$
Electro-optic sampling
Using EO effect for measuring THz pulse shape

The quasi-static field of THz pulse can induce birefrigence through the Pockels effect

\[ I_y = \frac{1}{2} I_0 \]
\[ I_x = \frac{1}{2} I_0 \]

Electro-optic sampling in ZnTe-type materials

In the case of velocity matching the phase retardation difference between the two orthogonal component of the probe is:

\[ \Delta \phi = (n_y - n_x) \frac{\omega L}{c} = \frac{\omega L}{c} n_O r_{41} E_{THz} \]

where \( n_o \) is the refractive index of the optical probe, and \( r_{4,1}=4d_{1,4}/n_o^4 \) is the EO coefficient.

The intensities at the two photodetector are:

\[ I_x = \frac{I_0}{2} (1 - \sin \Delta \phi) \approx \frac{I_0}{2} (1 - \Delta \phi). \]

\[ I_y = \frac{I_0}{2} (1 + \sin \Delta \phi) \approx \frac{I_0}{2} (1 + \Delta \phi). \]

The signal of the balanced photodetector is:

\[ I_s = I_y - I_x = I_0 \Delta \phi = \frac{I_0 \omega L}{c} n_O r_{41} E_{THz} \propto E_{THz} \]
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High intensity THz science and technology

Generation and application of THz pulses with extreme high energy and field
## Optical rectification

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Limitations of previous velocity matching methods:

- **Birefringence**
  possible for GaSe, but decreases the effective nonlinear coefficient, not possible at all for LN

- **Periodic poling (PPLN)**
  only (quasi-) phase matching, no velocity matching, → THz
  pulse duration > 10 ps, reduced efficiency

- **Cherenkov radiation**
  inconvenient emission characteristics, energy can not be scaled-up
Velocity matching methods

**velocity matching by tilting of the pump pulse front**


\[ \mathbf{V}_{vis}^{gr} \]

\[ \mathbf{V}_{THz}^{ph} \]

THz radiation excited along the tilted pulse front propagates perpendicularly to this front \( \rightarrow \) velocity matching condition:

\[ \mathbf{V}_{vis}^{gr} \cdot \cos \gamma = \mathbf{V}_{THz}^{ph} \]
Comparison of Cherenkov-type and tilted-pulse-front velocity matching

Cherenkov

\[ \Delta t_{\text{pump}} < T_{\text{THz}} \]
\[ w_{\text{pump}} < \lambda_{\text{THz}} \]
Cone shaped radiation

Tilted-pulse-front

\[ \Delta t_{\text{pump}} < T_{\text{THz}} \]
No limit for \( w_{\text{pump}} \)
Plane-wave radiation
Comparison of line focusing and tilted-pulse-front velocity matching

Line focusing: the THz wave transversal size increasing → THz energy ~ crystal thickness

Tilted-pulse-front: electric field of the THz pulse increases → THz energy ~ square of crystal thickness
Experimental set-up

LiNbO$_3$

THz

2 mm

He bolometer

Mira/Rega
200 kHz 3 μJ

2000 l/mm

L

λ/2
Experimental results

THz pulse energy at (77/300 K): 400/100 pJ
peak electric field: 7 kV/cm, photon conversion efficiency: 3.4%
Scaling-up the THz pulse energy

\[ E_{\text{THz}} = 240 \text{ nJ}, \quad \eta_{\text{ph}} = 10 \%, \quad \text{electric field (unfocused) } 0.1 \text{ MV/cm} \]
Classification of THz pulses by peak electric field and energy

- **Linear (TDTS) THz spectroscopy** ($E_{\text{max}} \approx 100 \text{ V/cm} \rightarrow 10 \text{ fJ energy}$)

- **High field THz pulses** ($E_{\text{max}} \approx 100 \text{ kV/cm} \rightarrow 1 \text{ µJ energy}$)
  THz pump – probe measurement, nonlinear THz optics

- **Extreme high field THz pulses** ($E_{\text{max}} \approx 100 \text{ MV/cm} \rightarrow 10 \text{ mJ energy}$)
  enhancement of HHG, particle manipulation, etc.
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High intensity terahertz science and technology

- Nonlinear terahertz optics
- Nonlinear spectroscopy (THz pump – THz/NIR probe measurement)
- Keldysh parameter
- THz photoemission
- THz Kerr-effect
- THz streaking
THz pump – THz probe set-up

Following the carrier dynamics by THz pump – THz probe measurement

Following the carrier dynamics by THz pump – THz probe measurement

n-type Ge, $n_c = 5 \times 10^{14}$ cm$^{-3}$

$$\alpha_c(\omega) = \frac{\varepsilon_b \omega_p^2 \gamma}{nc(\omega^2 + \gamma^2)} = \frac{e^2 N_c \gamma}{\varepsilon_0 n c^* (\omega^2 + \gamma^2)} = \frac{e N_c \gamma^2 \mu}{\varepsilon_0 n c (\omega^2 + \gamma^2)}$$

$$\alpha(t) \propto e^{-t/\tau_r} \quad \tau_r = 2.7 \text{ ps}$$

n-type GaAs, $n_c = 8 \times 10^{15}$ cm$^{-3}$

$$\gamma_{S3} = 2 \text{ ps}^{-1}, \quad \gamma_{3S} = 20 \text{ ps}^{-1}, \quad \gamma_{32} = 9 \text{ ps}^{-1}$$

$$\alpha(t) \propto \sum_i \mu_i n_i(t) \quad \tau_r = 1.9 \text{ ps}$$

n-type Si, $n_c = 5 \times 10^{14}$ cm$^{-3}$

$$\alpha(t) \propto e^{-t/\tau_{r1}} + e^{-t/\tau_{r2}} \quad \tau_{r1} = 0.8 \text{ ps}, \quad \tau_{r2} = 24 \text{ ps}$$

Impact ionization in InSb

Undoped sample, $N=5 \times 10^{13}$ cm$^{-3}$, $T=80$ K
Used model (avalanche ionization): Appl. Phys. A 29, 125 (1982), $\tau_e=7$ ps, $\varepsilon_0=1.3$ eV

Zener effect (tunneling) plays any role?

According to Phys. Rev. B 82, 075204 (2010), YES it does
Terahertz manipulation of excitons


Excitons in carbon nanotubes


THz free induction decay from tyrosine
The so-called Kaldysh parameter can be used to describe the strength of nonlinearity.

Keldysh parameter: \( \gamma = \sqrt{\frac{W_B}{2U_P}} \) where \( W_B \) is the binding energy, and \( U_P = \frac{e^2 E_{\text{THz}}^2}{4m\omega_{\text{THz}}^2} \)

is the cycle-averaged kinetic energy of the wiggling electron, the so-called ponderomotive potential.

Perturbative nonlinear optics for \( \gamma >> 1 \)  Strong field nonlinear optics for \( \gamma << 1 \)

For example for the Hirori experiment \( \gamma = \sqrt{\frac{19.6\text{meV}}{2 \cdot 64\text{meV}}} \approx 0.4 \)

The THz field dominates the excitonic Coulomb field.

The amplitude of the wiggling motion of an electron: \( a_w = \frac{e E_{\text{THz}}}{m\omega_{\text{THz}}^2} \)
Photoemission

Cs. Lombosi et al., ELI Workshop 2014
Photoemission

Photocurrent vs. peak THz field

Photocurrent vs. THz field polarization

Cs. Lombosi et al., ELI Workshop, Szeged 2014
Coherent control of soft mode

Lattice anharmonicity

SrTiO$_3$ is a ferroelectric crystal soft mode at 1.5 THz

300-nm-thick SrTiO$_3$ on 0.5 mm MgO

Tilted-pulse-front pumped THz source $E_{\text{THz, max}}$ = 80 kV/cm

Increasing $E_{\text{THz}}$ increasing amplitude, decreasing period

$$t(\omega) = \frac{E_{t}^{\text{Sam}}(\omega)}{E_{t}^{\text{Ref}}(\omega)}$$

Effective susceptibility:
\[
\chi_{\text{eff}}(\omega) = \frac{c(n+1)(t(\omega)-1)}{i\omega t(\omega)d}
\]

Resonance peak shifts to higher frequency for increased THz field. The bandwidth decreases.

Resonance peak shifts to higher frequency for increased temperature, but the bandwidth also increases.

The THz pulse drives the soft mode, but not the other modes.
Lattice anharmonicity

Numerical simulation

\[
\frac{d^2 Q}{dt^2} + \Gamma \frac{dQ}{dt} + \Omega^2 Q + \lambda Q^3 = \frac{e^* E_{\text{film}}(t)}{M}
\]

\[
E_{\text{film}}(t) = \frac{1}{n+1} \left( 2E_i(t) - \frac{d}{\varepsilon_0 c} \frac{\partial P(t)}{\partial t} \right)
\]

\[
e^* = 7.37 \text{ e}, \quad M = 18.6 m \text{ (proton)}
\]

\[
\chi_{\text{eff}}(\omega) = \frac{P(\omega)}{E_{\text{film}}(\omega)}
\]

\[
\Gamma = \Gamma_0 + \alpha Q^2
\]

\[
\lambda = 35 \text{ pm}^{-2}\text{THz}^2,
\]

\[
\alpha = -3.5 \text{ pm}^{-2}\text{THz}
\]
Coherent control of antiferromagnetic spin-waves

Coherent control of antiferromagnetic spin-waves

THz streaking
Characterization of ultrashort electron bunches or X-ray pulses

THz field streaks the electron bunch created by the X-ray (HHG) beam

The momentum of electrons born at time $t$ will be changed by the THz field by:

$$
\Delta \vec{p}(r, t) = e \int_{t}^{\infty} E_{THz}(r, t') dt' = eA_{THz}(r, t)
$$

where $\vec{A}_{THz}$ is the THz vector potential.

The change of the kinetic energy of electrons having velocity parallel to the THz field is:

$$
\Delta W_\parallel(t) \approx \sqrt{\frac{2W_0}{m}} \cdot eA_{THz}(t)
$$

B. Schütte et al., Electron wave packet sampling …, Opt. Express 19, 18833 (2011)
THz streaking
Characterization of ultrashort electron bunches or X-ray pulses

The electric field of a linearly chirped (chirp rate: c) Gaussian XUV pulse is:

\[ E_{\text{XUV}}(t) = E_X^0 \exp^{-2\ln 2 \left(\frac{t}{\tau_X}\right)^2} \cdot \exp^{i(\omega_0 t + ct^2)} \]

If the XUV pulse duration is much shorter than the THz, the temporal shape of the THz pulse can be measured as shown left.

If the XUV is much shorter than the THz pulse and the delay chosen such that the slope of \( A_{\text{THz}} \) is approximately linear the streaking induce a linear energy chirp of the electron bunch lengthening it to:

\[ \sigma_{\text{Xst}} = \sqrt{\sigma_{\text{X0}}^2 + \tau_X^2 \left(s^2 \pm 4cs\right)} \]

where \( s = \frac{\partial W}{\partial t} \approx \sqrt{\frac{2W_0}{m}} e^{E_{\text{THz}}}(t) \) and \( \pm \) corresponds to the two observation direction.

Neon 2p electron kinetic energy spectra vs. delay between ionizing XUV and streaking THz

B. Schütte et al., Electron wave packet sampling …, Opt. Express 19, 18833 (2011)
By measuring the field-free spectrum and the streaked spectra in two opposite directions, the XUV pulse duration and its linear chirp rate can be determined.

Photoelectron spectra measured at the left (left column) and right (right column) detector, respectively. (a,b) field free case, (c,d, e,f) using THz streak field. In the case of (e,f) a negative chirp of the pump pulse of the HHG generation increased the negative chirp of the harmonic pulse.

The large and small peaks corresponds to the 59th and 61st harmonic, respectively.
Outline

- Introduction
  - Properties of THz radiation/pulses
  - Importance of THz science and technique
- Cw THz sources
- THz detectors, THz imaging
- Single-cycle THz sources I.: Photoconductive antennae
- Time-domain terahertz spectroscopy (TDTs)
- Single-cycle THz sources II.: Optical rectification
  - Optical rectification II.: Tilted-intensity-front-pumping
- Single-cycle THz sources III.: Laser plasma
- High intensity THz science and technology
- Generation and application of THz pulses with extreme high energy and field
Classification of THz pulses by peak electric field and energy

- Linear (TDTs) THz spectroscopy \( (E_{\text{max}} \approx 100 \text{ V/cm} \rightarrow 10 \text{ fJ energy}) \)
- High field THz pulses \( (E_{\text{max}} \approx 100 \text{ kV/cm} \rightarrow 1 \text{ µJ energy}) \)
  - THz pump – probe measurement, nonlinear THz optics
- Extreme high field THz pulses \( (E_{\text{max}} \approx 100 \text{ MV/cm} \rightarrow 10 \text{ mJ energy}) \)
  - enhancement of HHG, particle manipulation, etc.
Velocity matching by tilting of the pump-pulse-front

Tilted-Pulse-Front Pumping (TPFP) Setup

\[
\nu_{\text{vis}}^{gr} \cdot \cos \gamma = \nu_{\text{THz}}^{ph}
\]


\[
\tan \gamma = -\frac{n}{n_g} \frac{\lambda}{\lambda} \frac{d\varepsilon}{d\lambda}
\]

2 mm

LiNbO_3

THz

He bolometer

lens

\(\lambda/2\)

~100 fs typical

fs laser

2000 l/mm
Longer Pump Pulse Duration for Longer THz Generation Length

Pulse front tilt:
\[ \tan \gamma = -\frac{n}{n_g} \frac{d\varepsilon}{d\lambda} \]

GVD parameter:
\[ D = \frac{d(\nu_s^{-1})}{d\lambda} = \frac{\lambda}{c} \left[ n \left( \frac{d\varepsilon}{d\lambda} \right)^2 - \frac{d^2 n}{d\lambda^2} \right] \]

Martínez et al., JOSAA, 1984
Fülöp et al., Opt. Express, 2010
Generation of THz pulses with extremely high energy

1. Using pump pulses with optimal duration


\[
\tan \gamma = \frac{\lambda \cdot d\theta}{d\lambda} \\
D_v = \frac{d\nu^{-1}}{d\nu} = \frac{\lambda}{c} \left[ \frac{d\theta}{d\lambda} \right]^2 - \frac{d^2 n}{d\lambda^2}
\]


2. Decreasing THz absorption via cooling the LiNbO₃ nonlinear crystal


3. Using contact grating setup


>20 mJ THz energy and >10 MV/cm peak electric field is predicted for 200 mJ pump pulse energy
Sub-mJ THz pulse energy for close to optimal pump duration

\[ y = ax^{1.53} \]

\[ y = ax^{1.40} \]

\[ y = ax^{1.67} \]

Fülöp et al., Opt. Express (2014)
• Tilted-pulse-front-excitation without imaging

• Pump spot diameter can be increased

• For semiconductors: smaller tilt, longer pump wavelength

• ZnTe, with 5 cm beam diameter:
  \( W_{\text{THz}} = 2.2 \text{ mJ} \), \( I_{\text{THz, focus}} = 630 \text{ GW/cm}^2 \),
  \( E_{\text{THz, focus}} \approx 17 \text{ MV/cm} \)

Fülöp et al., Opt. Express, 18, 12311 (2010)
THz pulses with MV/cm electric field generated in DSTMS

Optical rectification of 95 fs, 30 mJ, 1230 nm pulses of Cr:forsterite (Cr:Mg$_2$SiO$_4$) laser in collinear geometry. Pump fluence: 10 mJ/cm$^2$.

Low-pass filter is used with cut-off fr. at 10 THz.

Spectrum determined from the linear autocorrelation (inset) measured with a Michelson interferometer and THz detector.

THz pulses with MV/cm electric field generated in DSTMS

Efficiency up to 3 %, THz energy up to 0.9 mJ!

Focusing down to 260 μm the calculated THz field strength is 40 MV/cm.

THz pulses with MV/cm electric field generated in air-plasma
Efficiency scaling with pump wavelength


For 1850 nm pump from the 0.6 μJ THz energy, from the 85 μm focused THz spot size and from the sub-ps THz pulse duration 4.4 MV/cm field amplitude is calculated.

Broad spectrum peaking at 5 THz
Applications of THz pulses having extreme high electric field strength

• Increasing the cut-off frequency of HHG ($E_{THz} = 30 \div 100$ MV/cm)

• Quasi-phase-matched attosecond pulse generation ($E_{THz} = 2 \div 6$ MV/cm)

• Ultrashort X-ray pulse generation by THz driven undulators (and FEL’s)

• Electron acceleration

• Proton acceleration (hadron therapy)
Increasing the cut-off frequency of HHG

HHG in the presence of THz electric field

- Combined THz + IR fields
  \[ E(t) = E_0 \cos(\omega_{IR}t) + E_1 \cos(\omega_{THz}t) \]

- THz broken the symmetry of IR field
  Hong et al., Opt. Expr. 2009

- One as pulse per IR cycle

- Spectrum consists both odd and even harmonics
  Lewenstein, PRA 1994

Increasing the cut-off frequency of HHG

1560 nm

Both odd and even harmonics → only one as pulse per IR period

\[ I_{IR} = 2 \times 10^{14} \text{ W cm}^{-2} \]

\[ E_1 = 4 \times 10^9 \text{ V/m} \]

\[ E_{THz} = 0 \ldots 40 \text{ MV/cm} \]

Quasi-phasematched attosecond pulse generation


800 nm → 1,5 ÷ 2 μm: $\lambda_{\text{MH}} < 4 \text{ nm}$ (víz ablak)

Ne, $L = 2 \text{ mm}$,
$p = 25 \text{ Torr}$

IR: 800 nm,
20 fs, 0,2 mJ

$f = 1,6 \text{ THz}$

$E_{\text{THz}} = 5 \text{ MV/cm}$

$W_{\text{THz}} = 2\div5 \text{ mJ}$

$\lambda_{\text{MH}} = 7,2 \text{ nm}$
Electron manipulation by laser

11, 030704 (2008) undulator
12, 101302 (2009) deflection, focusing

1 GV/m = 10 MV/cm peak field strength is needed!
Motivation for THz-pulse-driven particle acceleration

Suggestion of laser-driven (non-plasma) particle accelerators (dielectric grating, photonic crystal, etc.): Dielectric laser accelerators (DLA)

Dielectric double grating accelerator


Demonstration of acceleration in DLA

J. McNeur, today 11.30

Up to 300 MeV/m acceleration gradient was demonstrated
Motivation for THz-pulse-driven particle acceleration

Disadvantages of short wavelength and short oscillation period of laser used for driving DLA:

1. small gap (400 nm) between dielectric grating $\rightarrow$ small bunch charge and low throughput

2. different parts of the bunch see different field phases $\rightarrow$ both acceleration and deceleration


100÷500 times longer wavelength of THz pulses can solve both problem
THz driven DLA for relativistic electron acceleration


Computer Simulation Technology (CST)
Optimized parameters of THz DLA (Depend on the used material)

- Material: Silicon \( (n = 3.41) \)
- \( A = B = \frac{\lambda}{2} \)
- We optimized \( C, D \) and \( L \) parameter.
  \[
  \rightarrow C = 0.1 \lambda \\
  \rightarrow D = 0.167 \lambda \\
  \rightarrow L = 0.28 \lambda \\
  \]
  \( \lambda = 0.9 \text{ mm (0.33 THz)} \)
The evanescent THz wave accelerator setup

Material requirements

- High optical damage threshold
- Large index of refraction
- Low absorption
- Low dispersion

Cooled LiNbO$_3$ (LN)

Acceleration and monochromatization of a proton bunch

Parameter setting for optimization:

\[
l_{\text{acc}} = 1.25 \text{ cm} \\
\nu_{\text{THz}} = 0.2871 \cdot c \\
\phi_0 = 3 \text{ rad}
\]

- Input: \( E_0 = 1.71 \text{ MV/cm} \)
- Output: \( \omega/2\pi = 0.5 \text{ THz} \)

Proton energy (MeV) vs. ordinal number of the proton (i)
Acceleration and monochromatization of a proton bunch in multi-stages at 0.5 THz frequency

$E_0 = 1.71 \text{ MV/cm}$

$\omega/2\pi = 0.5 \text{ THz}$

$d = 50 \text{ } \mu\text{m}$

$42 \rightarrow 52 \text{ MeV}$

monochromatization rate: 10%
Estimation of needed time for hadron therapy

1 shot typically has $10^9$ proton/MeV at 40 MeV
This means $10^9 \times 10\% \times 70$ MeV = 0.00175 J energy / single shot

Tens of Gy, required for hadron therapy is reached in a couple of minutes