Coherent THz Pulses from the NSLS SDL Photo-injected Linac and Applications in Materials Science

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Outline

• What are coherent THz pulses? What are they used for?

• The NSLS Source Development Lab photo-injected linac and accelerator-based THz.

• Some possible applications of strong coherent THz pulses in condensed matter physics.

• Electro-optic sampling, jitter and strong E-field effects.

• Demonstration of strong-field effects in superconductors.
Talks I am missing at Brookhaven ...

Subject: NSLS2 seminar talks
Date: Mon, 10 Jan 2011 17:44:02 -0500
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There will be seminar talks tomorrow and Wednesday.

1. Tuesday, January 11, 2011
   "Super KEK-B Status"
   Makoto Tobiyama, High Energy Accelerator Research Organization, KEK
   Hosted by Sam Krinsky, Weixing Cheng
   1:30 PM, NSLS-II Seminar Room, Bldg. 817

2. Wednesday, January 12, 2011
   "Compact ERL Project Status at KEK"
   Takashi Obina, High Energy Accelerator Research Organization, KEK
   Hosted by Sam Krinsky, Weixing Cheng
   1:30 PM, NSLS-II Seminar Room, Bldg. 817
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Coherent THz pulses

- Single or few cycle EM waves having $\leq 1$ ps duration & spectral content in the $0.1$ to $10 \times 10^{12}$ Hz range $\Rightarrow$ spectral width is typically comparable to the frequency content.

- Produced with ultra-fast lasers (e.g. optical self-rectification in a non-linear material) or as radiation from ultra-short electron bunches.

- Allow coherent detection (of E-field) $\Rightarrow$ magnitude and phase information $\Rightarrow$ direct connection to complex optical response

- Note that 1 THz is about:
  - 300 $\mu$m wavelength
  - 33 cm$^{-1}$
  - 4 meV
  - 50 K

- THz can sense: Intraband electronic transitions, acoustic phonons and soft modes, vibrational/rotational modes of (large) molecules, charge and spin density waves, magnons, cyclotron and spin resonance, pair-breaking in superconductors …
Synchrotron radiation: E-field for a single electron

- Dipole bending magnet radiation: Monopolar current, expect ½ cycle E-field pulse

\[ \delta t \sim \frac{\rho}{2c\gamma^3} \]

\[ \omega_c \sim \frac{2c\gamma^3}{\rho} \]
A “bunch” of 10 electrons
Resulting radiated E-field for 10 electrons

At low frequencies ($\lambda >$ bunch length), $E_{rms}$ scales as $n \Rightarrow I \sim n^2$ (coherent enhancement)

**Coherent Enhancement:**

Difficult to achieve in storage rings due to relatively long bunch length (cm’s and longer).

Linear accelerators are capable of producing much shorter bunches ( <100 fs) with number of electrons $\sim 10^{10}$

$$\frac{dI(\omega)}{d\omega} = \left[ N + N(N-1)f(\omega) \right] \frac{dI(\omega)}{d\omega}$$

where $f(\omega) = \left| \int_{-\infty}^{\infty} e^{i\omega \hat{r} \cdot \hat{r}} / c \ S(r) dr \right|^2$ (Nodvick & Saxon)
Synchrotron radiation waveforms

- Dipole bending magnet radiation: Monopolar current, \( \frac{1}{2} \text{cycle} \) \( E \)-field pulse

- Transition radiation: emitted when electron's Coulomb field crosses abrupt boundary between two different media, usually vacuum and a metal conductor. Bipolar current => Full-cycle \( E \)-field pulse
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National Synchrotron Light Source

User Facilities: 2.8 GeV X-ray and 0.8 GeV VUV/IR storage rings
Source Development Laboratory: 300 MeV photo-injected linac and free electron laser
The NSLS Source Development Lab Photo-injected Linac

X.-J. Wang

300 MeV S-Band Linac (DARPA)

BNL Photo-injector IV

10 m NISUS Wiggler (SDI)

Chicane Bunch Compressor

Ti:Sapphire Laser
Intense THz Pulses at the Source Development Lab

- ~300 fs ~300MeV nC electron bunches
- Dipole chicane compressor
- Off-crest section
- Photocathode e- gun
- 266 nm 8 ps

**THz Exp’t**

- 800 nm 150 fs pulses

- Coherent THz transition radiation
- 100 μJ per pulse \((10^{17}\) photons) (could be \(4\times\) higher)
  - Single-cycle pulses
  - Spectral content up to 2 THz
- 1 mm spot, would yield intensity of \(3\times10^{13}\) W/m²

\[
E = \sqrt{2(377\Omega)I} = 10^8 \text{ V/m} \quad \text{(MV/cm or 0.1 V/Å)}
\]

- Strong transient \(E\)-field ... and \(B\)-field = \(E/c \approx 0.3\) T

X.-J. Wang, Y. Shen, J.B. Murphy, X.Xi, GLC et al
A Comparison of Dipole and Transition Source Parameters

<table>
<thead>
<tr>
<th>Source Type</th>
<th>Dipole bend</th>
<th>E = 51 MeV ( \rho = 1 \text{ m} )</th>
<th>Transition</th>
<th>E = 51 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \theta_{\text{rms}} )</td>
<td>((3\lambda/4\pi \rho)^{1/3})</td>
<td>60 mrad; ((\lambda=1\text{mm})) 35 mrad; ((\lambda=200\mu\text{m}))</td>
<td>( \sim 1/\gamma )</td>
<td>10 mrad</td>
</tr>
<tr>
<td>( \sigma_{\text{rms}} )</td>
<td>((4\lambda^2 \rho/3\pi^2)^{1/3})</td>
<td>5 mm; ((\lambda=1\text{mm})) 2 mm; ((\lambda=200\mu\text{m}))</td>
<td>( \sim \lambda \gamma )</td>
<td>100 mm; ((\lambda=1\text{mm})) 20 mm; ((\lambda=200\mu\text{m}))</td>
</tr>
<tr>
<td>( Z_0 ) (Rayleigh)</td>
<td>((16\rho^2 \lambda/9\pi)^{1/3})</td>
<td>83 mm; ((\lambda=1\text{mm})) 48 mm; ((\lambda=200\mu\text{m}))</td>
<td>( \sim \lambda \gamma^2 )</td>
<td>10 mm; ((\lambda=1\text{mm})) 2 m; ((\lambda=200\mu\text{m}))</td>
</tr>
<tr>
<td>Multiple cycle?</td>
<td>Undulator (M. Gensch)</td>
<td></td>
<td>Smith-Purcell (Walsh &amp; Kimmitt)</td>
<td></td>
</tr>
</tbody>
</table>
Intense sources of coherent THz pulses: Laser vs Accel.

Strong fields => more than 10 μJ.

Thermal limits => absorbed power > 1W challenging for low T exp’ts..

Not shown: laser spectral range affected by NLO materials (5 to 10 THz difficult).

Not shown: highest energy laser-based THz pulses often have multiple cycles at higher frequencies (A. Leitenstorfer, U. Konstanz)

Accelerator-based sources offer better combination of pulse energy, rep. rate and spectral content for photoexcitation. Stronger fields even at low THz frequencies, single-cycle waveforms.

Plus, can combine THz and X-rays for unique THz pump
Laser Sources of Strong-Field THz via NLO

Difference frequency generation in a solid

**Several-cycle** pulses: $E \sim 100$ MV/cm: useful for some science problems but not all

Strong field with less pulse energy due to smaller volume (diffraction limits spotsize)

A. Sell, A. Leitenstorfer, and R. Huber

5 to 15 THz difficult due to NLO absorption (phonons)

$< 5$ THz requires more pulse energy for strong fields.
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**Science Opportunities with Intense Single-Cycle THz Pulses**

Systems driven by large transient Electric and/or Magnetic Fields.

- **Superconductivity:** dynamics of phase excitations
  - how do vortices initially form at the instant $J$ exceeds $J_{\text{critical}}$?
  - how does SC state recover? (SC magnets, RF cavities)
  - disrupting phase order in cuprates

- **Ferroelectricity:**
  - maximum switching rate, domain wall velocity?

- **Magnetism:**
  - spin dynamics (precession, relaxation)

- **Dielectrics:**
  - onset of dielectric breakdown

- **Structural distortions:** soft modes, lattice / electron coupling
  - detailed atomic potentials (double well, flat bottom)?

- **Semiconductors:** high-field transport, band structure
  - dynamics of band-bending and internal fields

- **Non-linear optics**

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Some phenomena could be probed with an ultra-fast x-ray pulse.
Strong Transient E-Field: Ferroelectric Switching


Calculations for PbTiO$_3$ 0.5 MV/cm field strength

Measurement idea:
Strong transient E-field to induce domain wall motion on femto-second time scale to sense fundamental limits on switching

$\frac{1}{2}$ cycle pulse desirable for driving polarization state, sense domains by microdiffraction?
**Strong Transient H-Field: Magnetization Switching**

*Figure from C. H. Back, et al., Science 285, 864 (1999)*

*Stohr et al, Stanford / SLAC*

- **Idea:**
  Use strong THz field to affect magnetization state of a thin film on $< 10^{-12}$ s time scale.

- **Ex situ approach**
  Use propagating THz wave external to accelerator. Contrast study at SLAC/SPPS (Stöhr *et al, Nature*) where specimen was placed inside linac and directly exposed to electron beam. **Need “half-cycle” type of THz waveform.**

- **Method:**
  pre-saturate film, expose to THz field pulse, then perform post image analysis (SEMPA)
Electromagnons in Multiferroic RMnO$_3$

Complex coupling of a lattice vibrational mode (electric polarization) and a lattice spin wave (magnon) in a multiferroic oxide.

**Experiment**: Pump the electromagnon with **few cycle THz pulse** matched to mode frequency, measure lattice (diffraction) and magnetization (element-specific XMCD) as function of time

Excitations within a potential well

Can THz waveform shape be controlled to drive system through and into specific quantum states?

Laser slicing (IMS/UVSOR). Variable field EM undulator? Smith-Purcell?
Superconductivity and Dynamics

Ordered, coherent state of paired electrons (Cooper pairs) in a conductor.
– electrons in a pair have opposite momentum and **opposite spin**.
– sea of paired electrons $n_{\text{super}}$ behaves as kind of superfluid.
– gap $\Delta$ forms in the electronic (excitation) spectrum.
– coherent state can be described by a complex order parameter $\psi = \psi_0 e^{i\varphi} \sim \Delta e^{i\varphi}$
  • amplitude $\psi_0 \sim \Delta$ is measure of gap and pair density
  • phase $\varphi$ describes electrodynamics

– supercurrent density $J \sim \nabla \varphi$
– voltage (potential) difference $\phi \sim \partial \varphi / \partial t$

– gap revealed in electron (quasiparticle) tunneling, photoemission, **THz spectroscopy**.

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Pairing driven by spin interactions - occurs for temperature above $T_C$? If so, have non-zero order parameter amplitude for $T>T_C$, long range superconductivity results when phase ordering takes place.

Normally, unable to sense electronic scattering under the SC dome by spectroscopy (superfluid dominates).

Use ultra-fast strong-field THz to disrupt phase, probe under the SC dome: THz to sense transport, x-ray pulse for stripes. Follow recovery (time-resolved).

Regular and random Josephson Junction arrays (weakly connected superconducting islands)

A model system for studying SC phase excitation and relaxation: THz pump and THz probe.
Carbon Wonderland

Graphene, a newly isolated form of carbon, provides a rich lode of novel fundamental physics and practical applications.

By Andre K. Geim and Philip Kim

Graphene is a one-atom-thick sheet of carbon that stacks with other such sheets to form graphite—pencil "lead." Physicists have only recently isolated the material.

The pure, flawless crystal conducts electricity faster at room temperature than any other substance.

Engineers envision a range of products made of graphene, such as ultrahigh-speed transistors. Physicists are finding the material enables them to test a theory of exotic phenomena previously thought to be observable only in black holes and high-energy particle accelerators.

Consider the humble pencil. It may come as a surprise to learn that the now common writing instrument at one time topped the list of must-have, high-tech gadgets. In fact, the simple pencil was once even banned from export as a
Graphene as a Non-linear Optical Material

Graphene Nanoplatelet films are thick (0.35 μm), have large area (cm²) and show Dirac-particle behavior. What is electromagnetic response for a system of (nearly) mass-less particles moving at a “fixed” speed?


For $\Omega = 2\pi \times 10^{12}$ rad/s (1 THz), $\tau = 300$K, $v = 10^6$ m/s => 8 kV/cm but, with electronic scattering at rate $\Gamma$, need factor of $\Gamma/\Omega$ more field => 50 to 200 kV/cm
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**THz Characterization: Standard Electro-Optic Methods**

Coherent detection setup for measuring THz waveforms using Pockels Effect: “THz Electro-Optic switch” (Zhang et al, Heinz et al)

\[
E_{\text{laser}} \sim \cos \left( \frac{2\pi n}{\lambda_0} z - \omega_0 t + \Delta \phi_E (t) \right)
\]

where

\[
\Delta \phi_E (t) = \left( \frac{2\pi L}{\lambda_0} \right) \Delta n [E_{\text{THz}} (t)]
\]

Electro-optic material (ZnTe) acts as a “variable waveplate”

\[
E(t) = E(t_0) + \frac{dE}{dt} (t - t_0) + \frac{1}{2} \frac{d^2 E}{dt^2} (t - t_o)^2 + \ldots
\]

Result: Detector signal gives instantaneous THz E-field

Works well for low jitter and high rep. rates.
Coherent detection setup for measuring THz waveforms using Pockels Effect: “THz Electro-Optic switch” (Zhang et al, Heinz et al)

\[ E_{laser} \sim \cos \left[ kz + \Delta \phi_E (t) - \omega t \right] \quad \text{where} \quad \Delta \phi_E (t) = \left( \frac{2\pi L}{\lambda_0} \right) \Delta n [E_{THz} (t)] \]

Electro-optic material (ZnTe) acts as a “variable waveplate”

Result: Snapshot of instantaneous THz E-field.

\[ E(t) = E(t_0) + \frac{dE}{dt} (t - t_0) + \frac{1}{2} \frac{d^2 E}{dt^2} (t - t_0)^2 + \ldots \]
EO sampling of SDL Linac THz Pulses: Spatio-temporal Map

Temporal-spatial E-field profile of coherent radiation pulse @ 5X source demagnification

Note: opposite sides are asymmetric, as shown (radial polarization)

E-field along horizontal plane

Jitter (~ 150 fs) limits ability to extract detailed waveforms & spectra.

Need a “single-shot” method

Simulation/2 (assumes stronger focus)
Single-Shot Electro-Optic Method

Use chirped sampling laser to encode waveform’s entire time-dependence onto different wavelengths of laser in a single pulse. Avoids need for multiple sampling.

Single-Shot EO Sampling of SDL THz Pulse using Chirped Laser

The graph shows the comparison between the THz pulse with and without THz ON. The peak intensity is labeled as 'peak is too high!' with annotations indicating a factor of 2x and 1x for comparison.

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Single-Shot EO Sampling of SDL THz Pulse: Higher intensity

![Graph showing spectral intensity against frequency and wavelength with two traces: one for 'No THz' and another for 'THz ON'.]
Non-linear Optics with Strong THz Pulses at SDL

“Simple” EO setup to observe time-dependent phase modulation (no initial laser chirp)

\[ E_{\text{laser}} \sim \cos \left[ kz + \Delta \phi_{E}(t) - \omega t \right] \quad \text{where} \quad \Delta \phi_{E}(t) = \left( \frac{2\pi L}{\lambda_0} \right) \Delta n[E_{\text{THz}}(t)] \]

Without polarizers, sense only time-dependent components

\[ E(t) = E(t) + \frac{dE}{dt}(t - t_o) + \frac{1}{2} \frac{d^2E}{dt^2}(t - t_o)^2 + \ldots \]

so, if \( \phi(x, t) = kx - \omega t - \beta t^2 \) then \( \omega_{\text{inst}} = \omega + \beta t \rightarrow \text{spectral shifting & chirping} \)

Calculated Phase Modulation Effects

\[ \phi(t) = \eta E(0) + \left[ \eta \left( \frac{dE_{\text{THz}}}{dt} \right) - \omega \right] t + \left[ \eta \left( \frac{d^2E_{\text{THz}}}{dt^2} \right) \right] t^2 + \ldots \]

Other details: Lensing from spatial variation of \( n(t) \) (time-dependent gradient index lens)

500 kV/cm field

0.5mm thick ZnTe

Laser spectra

Spectral Intensity [arb.]

Wavelength [nm]

Frequency [THz]
**Measured Phase Modulation with SDL Linac Coherent THz**

Electro-optic measurements of SDL THz pulses.
35 μJ pulses, 2mm focus, 0.5mm ZnTe.

~ 130 fs (FWHM) unchirped laser sampling pulse, no polarization analysis.

Probably still a mixture of effects
phase modulation (2\textsuperscript{nd} and 3\textsuperscript{rd} order NLO)
dynamic lensing that affects coupling into spectrometer’s optical fiber.

**Related Effects:**
Lensing due to parabolic refractive index gradient

\[ n^2 = n_0^2 \left(1 \pm \alpha^2 y^2 \right) \implies f \approx \frac{1}{n \alpha^2 d} \]

\[ 2\Delta n = \pm n_0^2 \alpha^2 y^2 \]

\[ \Delta n = \frac{n_0^3}{4} r_{41} E_{THz} \]

\[ \alpha^2 \sim 0.0023 \text{ for } E = 1 \text{ MV/cm} \implies f \sim 400 \text{ mm for } d = 0.5 \text{mm} \]
EO Detection of Bunch Coulomb Field (inside linac)

X. Yan et al (PRL '00)
I. Wilke et al (PRL '02)
H. Loos et al (PAC '03)

Full calculation with modulation effects
THz Compression of an Ultra-Fast Laser Pulse


Set up

165 fs laser pulse, compressed to 45 ps (factor of 3.7)
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  • phase $\phi$ describes electrodynamics

  – supercurrent density $J \sim \nabla \phi$
  – voltage (potential) difference $\phi \sim \partial \phi / \partial t$

– gap revealed in electron (quasiparticle) tunneling, photoemission, THz spectroscopy.
Response of a Superconductor to a low $\omega$ THz Pulse

Low $\omega$ response is almost purely inductive. E-field accelerates superfluid (current density $J$).

$$J \approx \sigma_n \omega_g \int_{-\infty}^{t} E(t') \, dt'$$

Current can potentially exceed $J_C$

- How does the superconducting state fail? How quickly? What controls the recovery?
- How can (or should) one measure the behavior?
Model Calculation: Finite Difference Time Domain Technique

- FDTD starts with discrete formulation of Maxwell’s equations. (K. Yee – ‘66)

\[ \nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} \quad \nabla \times \vec{E} = \frac{\partial \vec{B}}{\partial t} \n\]

- Dielectric response included through displacement.

\[ \vec{D}(t) = \varepsilon_\infty \varepsilon_0 \vec{E}(t) + \varepsilon_0 \int_0^t \vec{E}(t - \tau) \chi(\tau) d\tau \quad \text{also } B(t) \text{ as function of } \chi_m(\tau) \]

- Solve numerically
  - Recursive convolution method for materials where loss is described by exponential damping (e.g., Lorentzian) (Luebbers, Hunsberger and Kunz - ’91).
  - Drude model dielectric response: describes a normal metal (\( \tau \) small) or a superconductor (\( \tau \) large)
  - Successfully used for time-resolved THz spectroscopy (where \( \omega_p \) and/or \( \tau \) are themselves time-dependent). (Beard and Schmuttenmaer - ’01)

\[ \sigma(\omega) = \frac{\omega_p^2 \tau \varepsilon_0}{(1 - i \omega \tau)} \quad \chi(t) = \omega_p^2 \tau \left[ 1 - e^{-t/\tau} \right] \theta(\tau) \]
FDTD Calculation for Transmission

\[ \omega \ll \frac{1}{\tau} \]

\[ \omega \gg \frac{1}{\tau} \]

Transmission vs. Frequency [THz]

- NbTi thin film on sapphire
  - Red: Normal state
  - Blue: Superconducting
THz Optics for Low-Temperature Studies (Superconductors)

THz extraction, electro-optic and small signal detection setup (motors, motors & motors)
NbN: Transmitted Spectral Intensity versus E-Field Strength

Incident THz ($\omega < \omega_g$)

Low-pass filter

5% ND THz Filter

Can we detect any non-linear THz upconversion?

Broadband THz Detector

Brookhaven Science Associates
Summary & Outlook

• Linac-based sources of coherent THz can serve as unique tools for probing ultra-fast behavior in materials subjected to strong transient fields.
  – MV/cm E-field and nearly 1T magnetic field, ps or faster pulse.

• Pulse repetition rate > 1kHz desirable for S/N, high charge and flexible bunch length.
  – Advantage for Energy Recover Linac (ERL).
  – 2σ bunch lengths from ~3ps down to 30 fs would be good. (0.1 THz to 10 THz)
  – 100 pC or higher, especially for longer bunches (need more charge with long bunches for same E).

• Dipole bend and Transition radiation sources result in similar basic power and energy, but source geometry and extraction may favor one over the other.
  – Simpler polarization and source size for dipole bend.
  – Large apparent source size for transition, but interesting radial polarization.
  – Note: expect to be able to use conventional optics to control waveform shape and polarization.

• A number of interesting science experiments can be envisioned using the source as a pump
  – Combined with UV, soft and hard x-rays for unique THz pump, spectroscopic or structural probe.
  – Shown that the phase of a thin film superconductor can be completely disrupted within a full-cycle pulse.

• Stability is Important for both the THz and for supporting optical systems (e.g., ultra-fast laser used in electro-optic sampling).
  – Laser research community expects stability comparable to that in their own Lab.
Thank you for your attention