High-field THz generation and nonlinear THz spectroscopy

Keith A. Nelson
Department of Chemistry
MIT
Collective dynamics
Crystalline phase transitions
Accelerating carriers
Driving polar lattice vibrations
Driving low-frequency electronic resonances

Ionic & polar liquids, glasses, polymers...
Driving orientation, local modes, liquid rearrangements
Driving motions of charges & chemical change

Atomic & molecular transitions, charge transfer
Rydberg transitions, ionization
Driving molecular rotations & alignment

Broad objectives
Controlling charges and dipoles (electric & magnetic)
Measuring dynamical events involving their motions
Coherent control over collective & local structure & dynamics

Ferroelectric KNbO$_3$ crystal

K$^+$
O$_2^-$
Nb$_5^+$
Theoretical calculations: Domain switching
MD simulations of THz-driven FE domain switching: PbTiO$_3$
with Andrew Rappe & Tingting Qi, U Penn

THz fields drive increasing amplitudes until switching occurs!

Collective coherent control
Outline

Sources for high THz pulse energy

Common tabletop methods
- Nonlinear optical crystals, optical rectification
- THz “polaritonics”
- Tilted optical pulse front pumping
- Plasmas, THz-IR generation

Non-tabletop methods
- Synchrotron sources
- E-beam fringe fields at LCLS

Nonlinear spectroscopy
- Nonlinear vibrational & electronic responses in solids
  - Driving phonons & electrons
- Nonlinear responses in liquids & gases
  - Driving molecular orientation through polarizabilities & dipoles

Prospects
## Credits

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<tr>
<th>Thomas Feurer</th>
<th>Janos Hebling (Pecs U)</th>
<th>Christopher Werley</th>
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<td>Joshua Vaughan</td>
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<td>Bradford Perkins</td>
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<td></td>
<td>Yan Zhou</td>
<td>Christopher Tait</td>
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<td>Stephanie Teo</td>
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**Thanks for slides from:**

Andrei Tokmakoff, MIT
Gwyn Williams, Jefferson Lab
Antoinette Taylor, LANL
X.-C. Zhang, RPI

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Gwyn Williams, Jefferson Lab
Aaron Lindenber, Stanford
Antoinette Taylor, LANL
Christoph Hauri, Paul Scherer Inst
X.-C. Zhang, RPI
Tabletop THz generation

From photoconductive antennae

\[ E_{THz} \sim \frac{dJ}{dt} = \frac{d^2P}{dt^2} \]

in the far field

Electron acceleration produces THz emission

\[ \omega > E_g \text{(GaAs)} \]

From nonlinear crystals

Difference-frequency mixing produces THz emission

Collinear or non-collinear velocity matching...

100 fs laser pulse
Optical rectification in organic crystals

Recent results from Hauri Group (EPFL/PSI, Switzerland)

- e.g. DAST (4-N,N-dimethylamino-4’-N’methyl stilbazolium tosylate), OH1, DSTMS,…
- strong optical $\chi^{(2)}$ nonlinearity
- low (IR, THz) absorption
- high damage threshold (100 GW/cm$^2$)
- good phase matching
- pump wavelength: 1.2-1.5 µm
- high conversion efficiencies ($\approx$2 %)
- good focusability
Optical rectification in organic crystals

- up to 1.6 MV/cm (0.5 Tesla)
- up to 20 µJ pulse energy
- single-cycle pulses
- CEP stabilized
- excellently suited for high-field laser-matter interaction, like THz-induced magnetic switching (paper submitted)
All-Air THz Photonics

Laser pulse 100 fs, 0.8 mJ, 800 nm, 1 kHz

Detector

BBO

Plasma

Si filter

THz


http://www.rpi.edu/~zhangxc
THz generation mechanism:

Current surge → THz generation

BBO crystal


Cross-correlation with 45 fs 3 µm in InSb

Pulse duration < 100 fs
Reflection directs THz into far field

From electron beam near field

Electron acceleration produces THz emission

Gwyn Williams, Jefferson Lab

Reflection directs THz into far field

From electron beam near field

Aaron Lindenber, Stanford/LCLS
Conventional NLO crystal
Collinear velocity matching

Optical pump
THz

ZnTe
TOP VIEW

\[ n_{vis}^{gr} = n_{THz} \]
\[ V_{vis}^{gr} = V_{THz}^{ph} \]

Equal refractive index values
⇒ Equal velocities
Optical & THz pulses copropagate through crystal

ZnTe, GaP, GaSe, etc.

High-dielectric NLO crystal
Collinear velocity matching is not possible

THz velocity << optical velocity
THz pulses propagate mostly laterally through crystal

Optical pump
THz
THz

LiNbO₃
TOP VIEW

\[ n_{THz} \gg n_{vis}^{gr}, V_{THz}^{ph} \ll V_{vis}^{gr} \]

Cerenkov condition:
\[ \cos \gamma = n_{vis}^{gr} / n_{THz} \]
\[ V_{vis}^{gr} \cos \gamma = V_{THz}^{ph} \]

So LiNbO₃ can’t velocity match collinearly
But it has a very high figure of merit!
THz phonon-polariton generation in LiNbO$_3$ slabs

- The large index mismatch between THz and optical light leads to Cherenkov radiation: the THz propagates mostly perpendicular to optical pulse

- Optical Intensity envelope $\rightarrow$ THz E-field waveform
High EO constants in ferroelectric crystals

It’s the ions!!

Polar lattice vibrational mode

“Soft” mode in FE phase transitions

⇒ high $\varepsilon$

Ferroelectric KNbO$_3$ crystal

$\varepsilon_K + O_{2z}^{5+}$

$\varepsilon_{\infty}$

$\mu_0 b_{21} Q$

$F_{ISRS}$

$\alpha$

$\frac{N}{M} (\frac{\partial \alpha}{\partial \omega}) |\bar{E}(t)|^2$

$T_H$ phonon coordinate

THz Field

$\ddot{Q} + \Gamma \dot{Q} + w_{TO}^2 Q = b_{12} \bar{E}$

Laser Field

$\frac{1}{2} \varepsilon_0 \sqrt{N M (\frac{\partial \alpha}{\partial \omega}) |\bar{E}(t)|^2}$

electronic nonlinearity – neglected!
THz optic phonon-polariton modes in FE crystals

Coupled lattice vibrational/electromagnetic modes
~ 0.1-10 THz frequencies, 5-500 µm wavelengths in FE crystals
Polaritons move through host crystal at light-like speeds

Polaritons can be used as THz signals
Terahertz “Polaritonics” platform possible
Direct x-ray probing of polariton lattice displacements

Fs x-ray pulse used for time-resolved diffraction
Derived from LBL synchrotron source


Fs x-rays achievable through tabletop laser system
Polaritons & polariton-induced structural change may be monitored
THz polariton imaging

Lateral propagation makes THz wave accessible to further optical inputs

Enables real-space polariton imaging

Complete temporal and spatial evolution monitored
Spatiotemporal polariton imaging & control

*The movies*

Samples: LiTaO$_3$, LiNbO$_3$ crystals  Polariton speed $\approx c/6 = 50 \, \mu$m/ps
Length scale $\sim$ 1-2 mm, Temporal range $\sim$ 20-40 ps
Spatiotemporal polariton imaging & control

*The movies*

Samples: LiTaO$_3$, LiNbO$_3$ crystals  
Polariton speed $\approx c/6 = 50 \ \mu$m/ps  
Length scale $\sim 1$-$2$ mm,  Temporal range $\sim 20$-$40$ ps
Phase-contrast THz polariton imaging

C.Werley et al., JOSA B (in press)
Polaritonic structures & devices

Integrated THz functionalities

Polaritonic waveguide splitter, interferometer, resonator, 90° bend

Air

LiNbO$_3$

Air

LiTaO$_3$

Air

All LiNbO$_3$ channels are 200-300 $\mu$m wide

5 mm x 4 mm crystal, 250 $\mu$m thick

Polaritonic grating

_Nature Materials_ 1, 95 (2002)
Polaritonic structures & devices

Waveguide

Focusing reflector
**Spatiotemporal polariton imaging**

THz “movies” yield complete spatial & temporal evolution

**Spatiotemporal THz coherent control**

Input: Single beam, Single fs pulse

Programmable Spatiotemporal Fs Pulse Shaper

Output: Many beams, Many fs pulses

THz amplification, phased array generation

**Integrated THz functional elements**
fabricated by fs laser machining

Spatiotemporal polariton coherent control

*Through spatiotemporal fs pulse shaping*
Spatiotemporal polariton coherent control

Timed/phased array generation

4 spots
Tilt down

8 spots
Focus

40 spots
Tilt up

8 spots
Focus
Tilt down

(c) Nielsen Lab 2002

(c) Nielsen Lab 2002

(c) Nielsen Lab 2002
Spatiotemporal polariton coherent control

Coherent THz amplification

Horizontal array
Cylindrically focused “line” sources
Linear temporal sweep

Large THz pulse energies
Spatiotemporal control over THz field

With reconfigurable spatiotemporal fs pulse shaping
With non-reconfigurable echelon structure

THz wave coherent amplification

*Tilted pulse front:* Simple, compact setup

Grating introduces tilt
Lens adjusts tile angle
Crystal in image plane

Works well at Hz-KHz-MHz rep rates

*J. Hebling et al., Opt. Exp. 10, 1161 (2002)*
Higher THz pulse energies

**10 Hz**

THz Pulse Energy: **50 µJ**
- THz Pulse Energy: **10 nJ**
- THz Av. Power: > 2.5 mW
- THz Pulse Energy: > 2.5 nJ
- THz Pulse Energy: > 7 µJ
- THz Av. Power: > 2.5 mW
- THz Av. Power: > 2.5 mW
- THz Pulse Energy: > 2.5 nJ
- Field strength: 750 kV/cm
- Energy efficiency: 4 x 10^-4
- Energy efficiency: ~ 10^-3
- Energy efficiency: 1.8 x 10^-5
- Photon efficiency: 15 %
- Photon efficiency: ½%
- Photon efficiency: 15 %
- Suitable for practical applications

J. Hebling et al., *APL* 90, 171121 (2007)


M. Hoffmann et al., *APL* 93, 141107 (2008)
THz multiple-cycle waveform generation

Chirp-and-delay optical pump yields tunable THz frequency

Multi-μJ multiple-cycle pulse energies

THz beam profile

Beam profile with pyroelectric camera (Spiricon Pyrocam III)
Focusing with an aspheric lens
100 mm pixel size
2.5 mJ THz pulse energy, 1 kHz rep rate

Excellent for imaging, spectroscopy, other applications
Improved focusing
Cherenkov THz wave generation in LiNbO₃

Nonlinear THz spectroscopy

Microjoule level pulses ⇒ THz nonlinear spectroscopy

THz nonlinear spectroscopy measurements conducted
- Nonlinear THz transmission, self-phase modulation
- THz-induced ionization, fluorescence
- THz pump – optical probe
- THz pump – THz probe

Nonlinear vibrational & electronic responses studied
- Solid-state vibrational & electronic responses
- Phase transitions, chemical reactions
- Liquid-state molecular alignment
- Gas-phase molecular orientation, ionization
THz Transmission Spectrometer
THz Pump – THz Probe Capabilities

Also THz pump – Optical probe
Measure optical birefringence, SHG,…
THz pump-probe nonlinear spectroscopy

Collinear pump & probe pulses

EO readout of transmitted pump & probe fields
Nonlinear THz spectroscopy

Crossed THz pump beams induce anharmonic lattice vibrations

\[ \text{LiNbO}_3 \text{ crystal} \]

Anharmonic lattice vibrations

\[ \text{THz 1} \quad 0.5 \text{ THz} \]

\[ \text{THz 2} \quad 0.5 \text{ THz} \]


Start toward collective coherent control
Excitation of the soft-mode in SrTiO$_3$


\[
\frac{d^2 q}{dt^2} + \Gamma \frac{dq}{dt} + \omega_0^2 q + aq^3 = AE(t)
\]
SrTiO$_3$ TFISH detects symmetry loss

- **High $T$:** instantaneous response
- **Lower $T$:** Soft mode $2\times \omega$
- **Still lower $T$:** Rise then decay
- **Lowest $T$:** Slowest decay
Oscillating & non-oscillating components

Moderate $T$: Mode softening

Soft mode & *polar nanoregion response*
Spectrally integrated THz pump-probe results
Overall hot electron relaxation dynamics
Bulk Ge & GaAs crystals

THz-induced carriers in InSb

THz pump – THz probe electron dynamics

InSb impact ionization & THz-induced tunneling

THz fields pull weakly bound electrons out of conduction band

Accelerated carriers ionize additional electrons

THz fields can release weakly bound electrons generally
THz-induced carriers in InSb
THz pump – THz probe electron & lattice dynamics

Temporal and spectral resolution reveal buildup of carriers, relaxation into phonon manifold.

Difference phonon absorption at 1.2 THz due to carrier equilibration with lattice phonons

*JOSA B 26*, A29-A34 (2009)
*PRB 79*, 161201 (R) (2009)
Exciton ionization in ZnSe MQWs


- Field ionization of excitons (instantaneous)
- Dynamical Franz-Keldysh effect in band to band transition
Carrier Multiplication in GaAs MQWs


Impact ionization
Graphene

Exfoliated Graphene
- Linear electronic dispersion
- Massless (or very low mass) carriers
- High mobility ($>10^4 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$)
- Small size (due to difficulties in exfoliation technique)
- Low intrinsic carrier concentration

CVD graphene
- Linear electronic dispersion
- Massless (or very low mass) carriers
- Limited mobility ($\sim2500 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$)
- Large sizes (macroscopic meter size sheets!)
- Fairly high doping from etching and possibly impurities

$E_f = -270 \text{ meV (Hole Doped CVD)}$
Intraband Drude conductivity

\[
\frac{\sigma_1(\omega)}{\sigma_Q} = \frac{8k_B T}{\pi \hbar} \ln \left( e^{-E_F/2k_B T} + e^{E_F/2k_B T} \right) \frac{1}{\omega^2 \tau + 1/\tau}
\]

- When Temp increases, weak effects on conductivity (increase) and transmission (decrease)
- When \( \tau \) decreases, strong effects on conductivity (decreases) and transmission (increases) – Suggested by theoretical results from Bao

\[\text{Induced transparency as fluence increases}\]

**Nonlinear Transmission**

```

<table>
<thead>
<tr>
<th>Frequency (THz)</th>
<th>Transmission (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>70</td>
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<tr>
<td>0.7</td>
<td>80</td>
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<tr>
<td>0.9</td>
<td>90</td>
</tr>
<tr>
<td>1.1</td>
<td>100</td>
</tr>
<tr>
<td>1.3</td>
<td>100</td>
</tr>
<tr>
<td>1.5</td>
<td>100</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fluence (µJ/cm²)</th>
<th>Transmission (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.4</td>
<td>70</td>
</tr>
<tr>
<td>25</td>
<td>80</td>
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<tr>
<td>135</td>
<td>90</td>
</tr>
<tr>
<td>220</td>
<td>100</td>
</tr>
<tr>
<td>390</td>
<td>100</td>
</tr>
</tbody>
</table>

\[\Delta T/T = (T_{\text{high field}} - T_{\text{low field}})/T_{\text{low field}}\]
```
Nonlinear THz Responses in CVD Graphene

Hole-doped graphene

Saturation of THz absorption

Time-dependent absorption recovery

THz field drives carriers & impact ionization

arXiv:1101.4985v1 {cond-mat.mtrl.-sci} 26 Jan 2011
THz dipole antenna

Collaboration w/ R. Averitt group, Boston U

530 GHz, on resonance
THz dipole antenna
Low-order mode
Field enhancement

C.A. Werley et al, Optics Express 20, 8551-8567 (2012)

530 GHz, on resonance

Nano-gap THz field enhancement: work by Dai-Sik Kim and by Thomas Feurer
Metamaterials: Tailored electromagnetic responses

• Engineered resonant structures
• Specific EM responses
• Depends strongly on materials used

Enhancing field strength with metamaterials

Metamaterial responses are highly sensitive to the substrate
Nonlinear metamaterial responses in n-doped GaAs:
Electron acceleration at moderate fields

$n_e = 1 \times 10^{16} \text{ cm}^{-3}$

No metamaterial response in unexcited system due to substrate conductivity $\sigma$

THz excitation reduces $\sigma$
MM resonance appears!
Nonlinear metamaterial responses in n-doped GaAs:
Impact ionization at high fields

Strong THz field is enhanced at induced MM resonance!
MV/cm fields $\Rightarrow$ impact ionization $\Rightarrow$ increases $\sigma$
MM resonance is suppressed!

Metamaterial response enhances THz field and sensitizes THz measurement

$E_g = 1.4$ eV
1 THz = 4 meV
Nonlinear metamaterial responses in SI GaAs: Tunneling & impact ionization increase $\sigma$

$n_e = 2 \times 10^6 \text{ cm}^{-3}$
Metamaterial response in unexcited system due to low substrate conductivity $\sigma$

Strong THz field is enhanced MV/cm fields $\Rightarrow$ impact ionization $\Rightarrow$ increases $\sigma$ by $10^8$!
MM resonance is suppressed!

Huge conductivity change suggests THz sensing applications!
THz-induced damage in GaAs

Damage along field lines
Pattern resembles dielectric breakdown
Multielement Detector on GaAs

Pads for electrical contact

Active area
Multielement GaAs Detector Data
(for a single element)
Multielement GaAs Detector Data
(for a single element)
Multielement GaAs Detector Data (for a single element)

single element of array detector
pulse energy $\sim 0.3$ microjoules
field strength $\sim 65$ kV/cm
Multielement GaAs Detector Data
(for a single element)

red: Gaussian
FWHM ~2.3 mm
Vanadium Dioxide: Insulator to metal phase transition

\[ T_c = 340 \text{K} \]

w/ Mengkun Liu & Rick Averitt, Boston University

*Nature* 487, 345–348 (11 July 2012)

<table>
<thead>
<tr>
<th>Low temperature</th>
<th>High temperature</th>
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<tbody>
<tr>
<td>Monoclinic structure</td>
<td>Rutile structure</td>
</tr>
<tr>
<td>Insulating: ( \sigma &lt; 10^{-2} (\Omega \text{cm})^{-1} )</td>
<td>Metallic: ( \sigma &gt; 10^3 (\Omega \text{cm})^{-1} )</td>
</tr>
</tbody>
</table>

M. M. Qazilbash et al., Phys. Rev. B 77, 115121 (2008),
VO₂ THz metamaterials
T-dependent insulator-metal transition

0.4 THz metamaterial resonance
Metallic VO₂ shorts gaps
THz resonance disappears at high $T$ in metallic
VO₂ THz metamaterials

THz fluence dependence

THz resonance disappears at high THz fluence
$\text{VO}_2$ THz metamaterials

THz fluence dependence

Measurement $T = 325 \text{ K}$ Simulation

![Graph showing transmission vs frequency for VO$_2$ THz metamaterials with measurement and simulation at $T = 325 \text{ K}$]
THz-induced IMT dynamics

~ 5 ps for transition to occur
Poole-Frenkel ionization and carrier heating in VO$_2$

Two-temperature model

\[ \sigma = \sigma_0 \exp \left( \frac{\sqrt{e^3 |E(t)|}}{r k_B T} \right) \]

\[ C_e \frac{dT_e}{dt} = -G(T_e - T_i) + \sigma(t)E^2(t) \]

\[ C_i \frac{dT_i}{dt} = +G(T_e - T_i) \]

- \( \sigma \) - conductivity
- \( E \) - THz electric field
- \( C_e, C_i \) – electron/lattice specific heat
- \( T_e, T_i \) – electron/lattice temperature
- \( G \) - electron-phonon coupling coefficient

- With P-F \( \sigma \), \( \Delta T \sim 20 \text{ K} \)
- With \( \sigma_0 = 10 \text{ (} \Omega \cdot \text{cm})^{-1} \), \( \Delta T < 1 \text{ K} \)
THz-induced damage in VO$_2$: Damage patterns and the Poole-Frenkel mechanism

Damage along equipotential lines
Metamaterial-enhanced chemical decomposition

Partial decomposition of TNT!
THz Kerr effect in liquids & solids

THz pulse drives polarizability anisotropies, induces birefringence

\[ n(t) = n_0 + \int dt' R(t-t') |E|^2(t') \approx n_0 + n_2 I \]

Hoffmann, et. al., APL (2009)
Nonlinear Kerr response in air!

Nonlinear signal in air!
What from??
Orientation and Alignment of Gas Phase Molecules by Single Cycle THz Pulses

Sharly Fleischer
w/ Yan Zhou & Robert W. Field
EO sampling, OCS 250torr

Macroscopic dipole

$$E \propto \frac{d \langle \cos \theta \rangle}{dt}$$


S. Fleischer et al. *PRL* 107, 163603 (2011)
Alignment of OCS, 350 torr, 300K

Measured through Kerr effect (optical birefringence)

S. Fleischer, Y. Zhou, R.W. Field, KAN, PRL 107, 163603 (2011)
Two delayed THz pulses, 350 torr OCS

Small second pulse induces large second revivals!
Two-pulse simulation

Alignment factor (a.u.)

Time (ps)

33 ps

16.5 ps

16.5 ps
Two-quantum coherences

J=4

J=3

J=2

J=1

J=0

2B

4B

6B

10B

14B
### Density matrix

\[
\begin{array}{cccccc}
\rho_{11} & \rho_{12} & \rho_{13} & \rho_{14} & \rho_{15} \\
\rho_{21} & \rho_{22} & \rho_{23} & \rho_{24} & \rho_{25} \\
\rho_{31} & \rho_{32} & \rho_{33} & \rho_{34} & \rho_{35} \\
\rho_{41} & \rho_{42} & \rho_{43} & \rho_{44} & \rho_{45} \\
\rho_{51} & \rho_{52} & \rho_{53} & \rho_{54} & \rho_{55}
\end{array}
\]

1Q coherences
Orientation \( \langle \cos \theta \rangle \)

2Q coherences
Alignment \( \langle \cos^2 \theta \rangle \)
Population transfer

Multiple-quantum coherences

\[ J \leftrightarrow J \pm 1 \leftrightarrow J \pm 2 \]
Second pulse timing varied

Fully coherent 2D 2-quantum THz spectroscopy
OCS alignment 180 torr

THz FID depletes population!
OCS alignment, no purge outside OCS cell

Water FID drives OCS 2QCs!
THz pulse can saturate rotational transitions!

Multiple-cycle waveform can be tuned to specific water lines

Waveform can be optimized for energetic material response
Can saturate water absorption lines
Simulation $\Rightarrow$ enhanced propagation in air!
Summary

**Strong THz sources**
Tabletop sources provide high THz pulse energies, high THz field amplitudes
Enable versatile nonlinear spectroscopy at low and high order

**Nonlinear THz spectroscopy**
Can be declared a subfield!
Nonlinear responses observed in solid, liquid, gas, plasma phases
Electronic, magnetic, vibrational, rotational responses
Collective structural and localized chemical rearrangements

**Prospects**
Nonlinear THz spectroscopy is just beginning
Multidimensional, high-order, multispectral spectroscopy
THz coherent control over molecular & collective responses
Credits

Thomas Feurer                Janos Hebling (Pecs U)                Christopher Werley
Joshua Vaughan               Mattias Hoffmann                    Nate Brandt
Nikolay Stoyanov            Ka-Lo Yeh                             Qiang Wu (Tianjin U)
David Ward                   Harold Hwang                          Kung-Hsuan Lin
Eric Statz                   
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