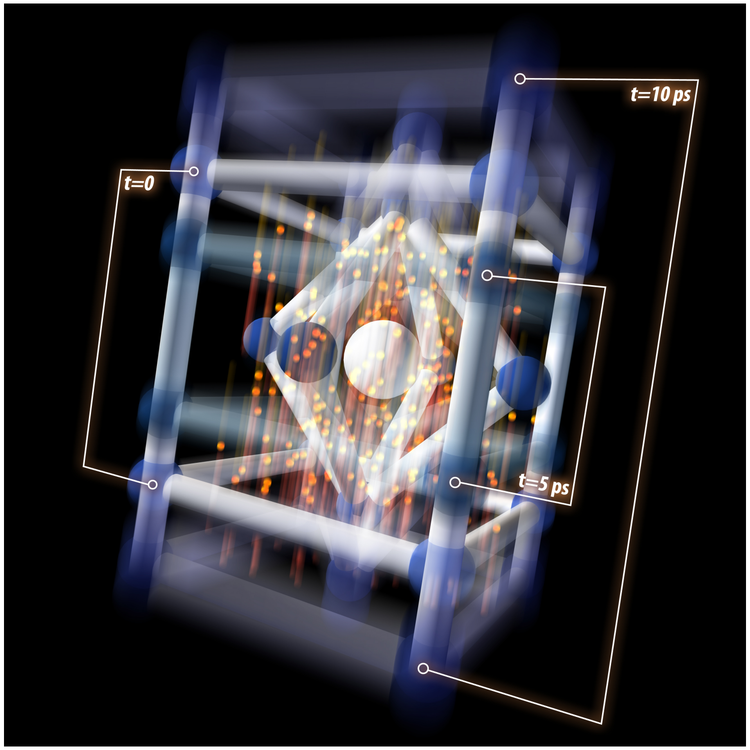
Femtosecond terahertz/x-ray studies of solids

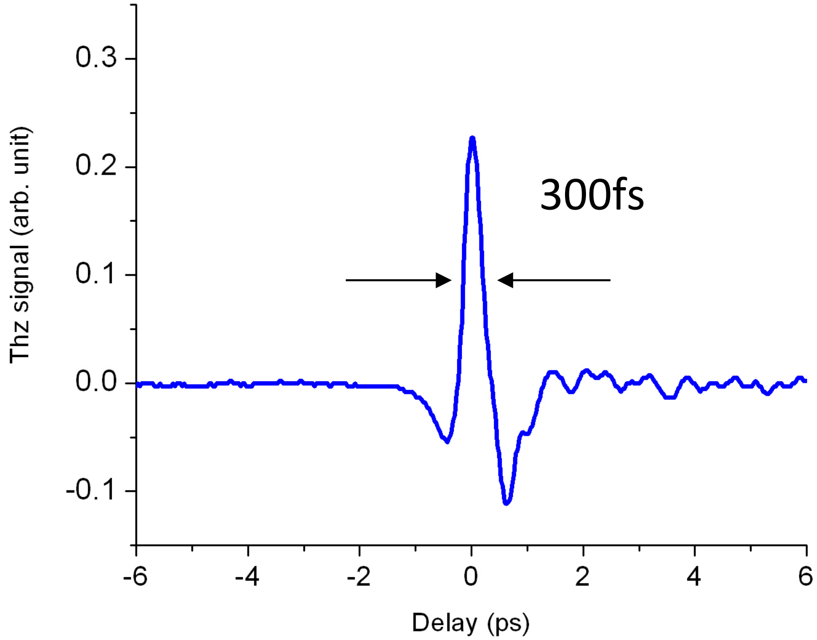
Aaron M. Lindenberg
Dept. of Materials Science and Engineering, Stanford University
SLAC National Accelerator Laboratory



2012 PULSE Ultrafast X-ray Summer School
June 19th, 2012

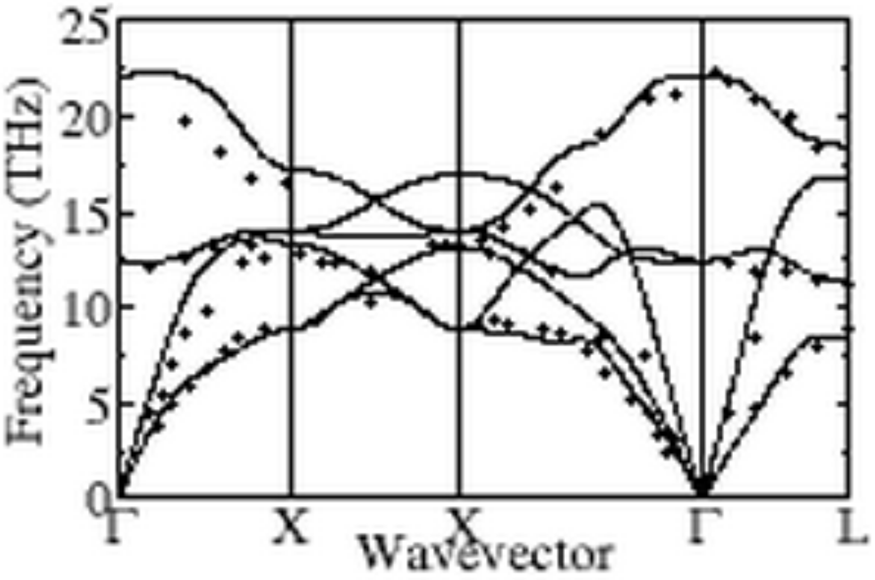
X-ray UV visible Infrared THz

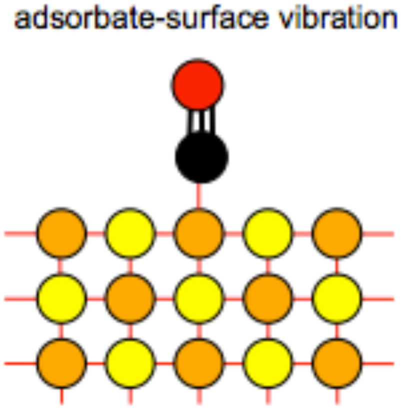
~10000eV

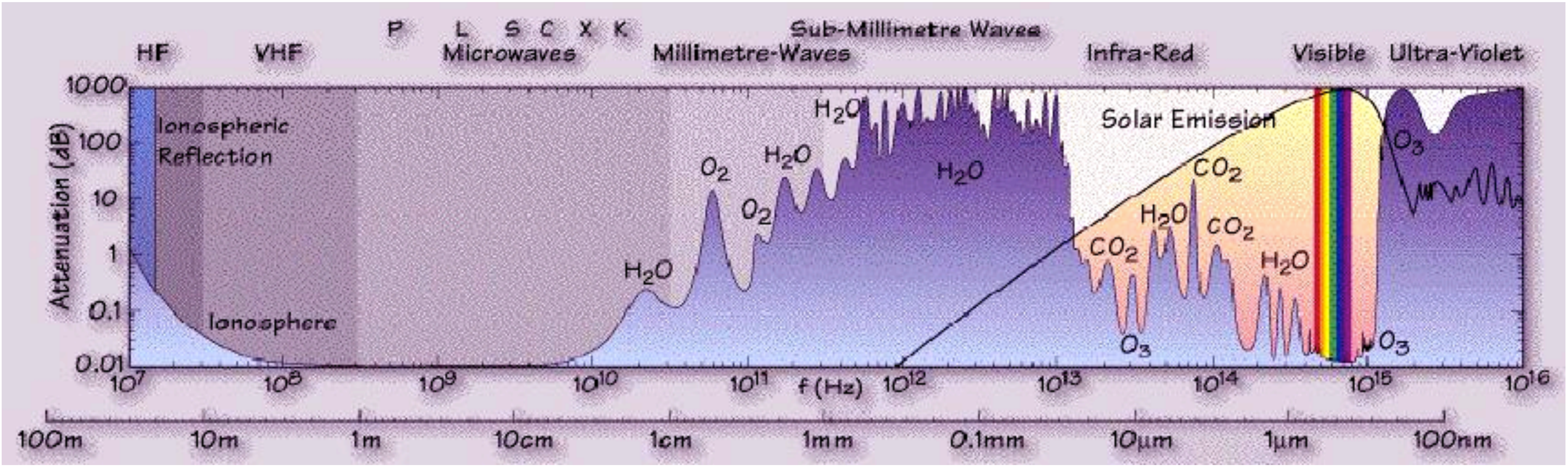


~0.004eV

Access to vibrational modes





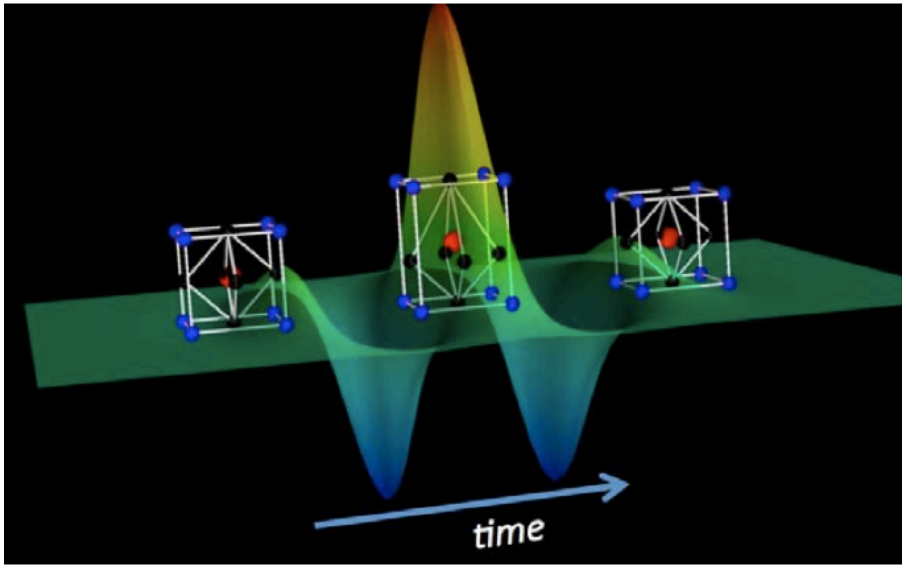


Motivating Questions

-Can we visualize at the atomic-scale and in real-time what atoms and electrons are doing in condensed matter? Key is to disentangle different degrees of freedom

-Can we use light in order to engineer and enhance the functional properties of materials and devices?

-What are the fundamental speed limits that govern how materials transform?



Beyond indiscriminate blasting of materials…

Materials under Extreme Electromagnetic Fields

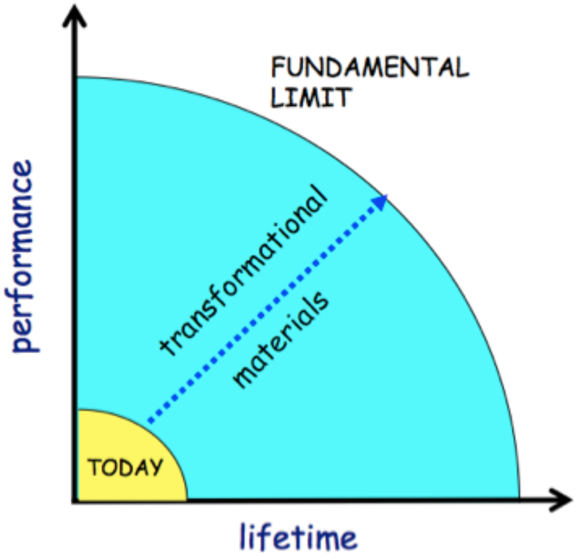
-Extreme electric fields: First steps in dielectric breakdown. What is the maximum field strength a material can withstand?

-Long-distance transmission lines

-Extreme fields in nanoscale devices, integrated circuitry

-Magnetic fields: Highest peak fields generated in destructive explosive devices (~1000 T)

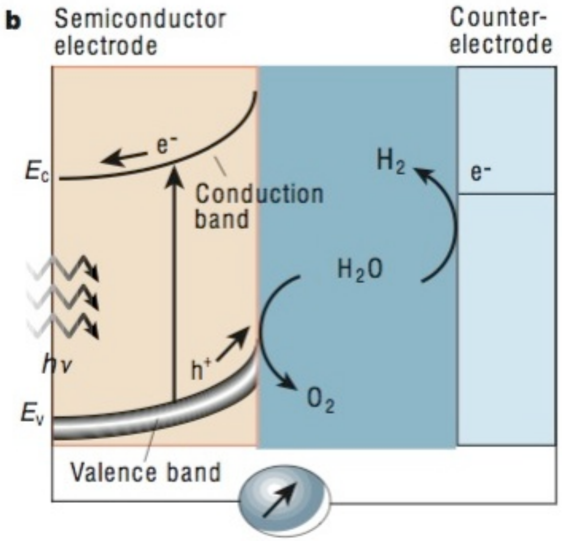
-Superconducting materials: Critical fields and currents.



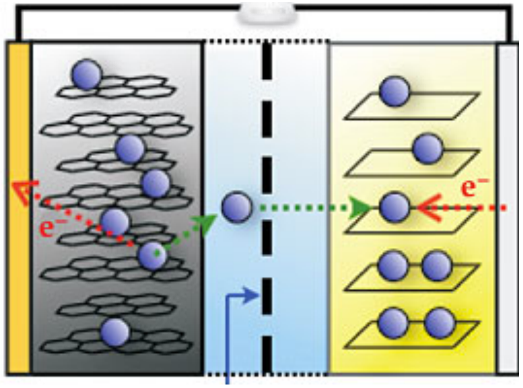
G. Crabtree et al.

Ultrafast ionic processes in real devices for energy applications

Photoelectrochemical cell

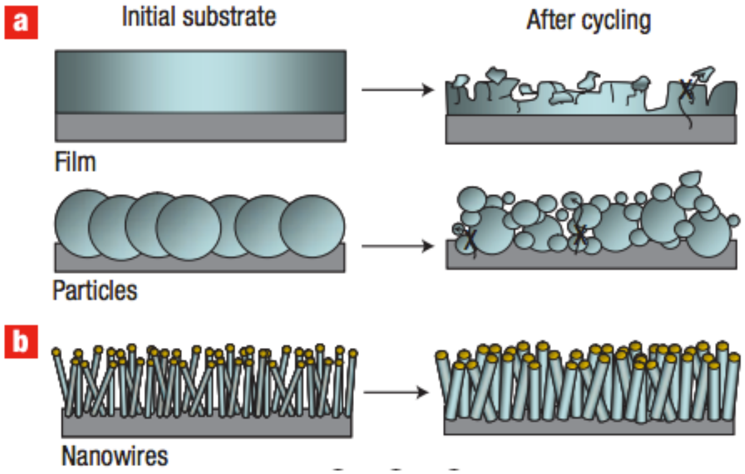


Gratzel et al.



Abruna et al. Phys. Today, Dec, 2008

Lithium ion battery



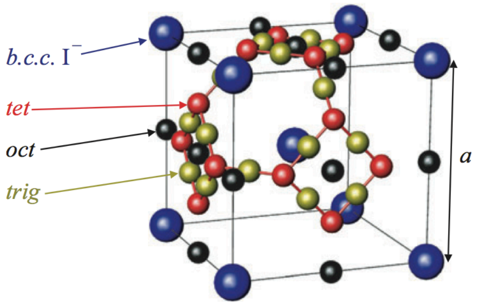
Chan et al. Nature Nano (2007)

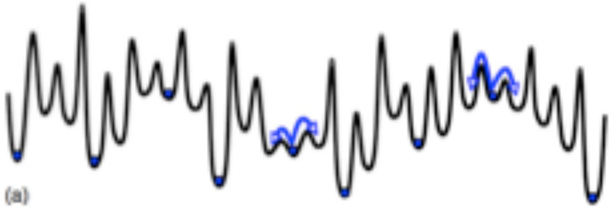
$$LiCoO_2 \rightarrow 1/2\ Li^+ + 1/2\ e^- + Li_{0.5}CoO_2$$

$$C_6 + Li^+ + e^- \rightarrow LiC_6$$

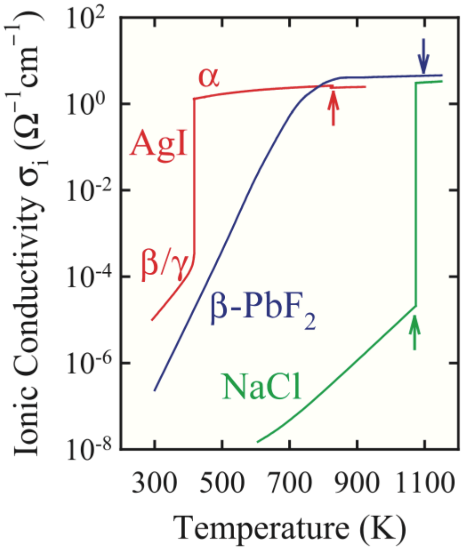
Ion-induced structural phase transformations

Superionic conductors





ionic plasma frequency ~ 1 THz

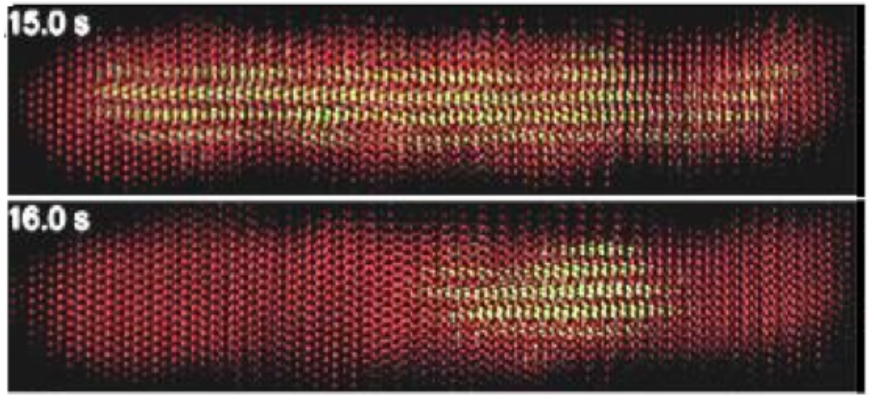


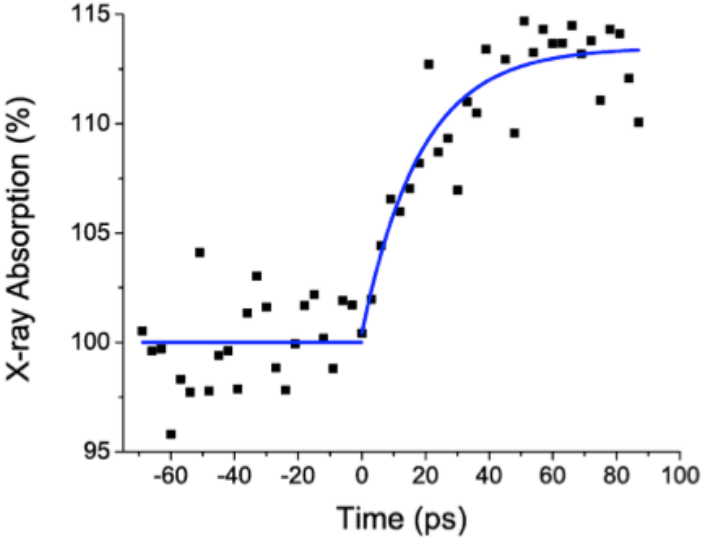
Hull et al.

$$\sigma = \sigma_o \exp\left(-\frac{E_a}{k_B T}\right)$$

-Ionic conductivities ~ 1 Ω^{-1} cm^{-1}

-Hopping time-scales of order a few picoseconds…



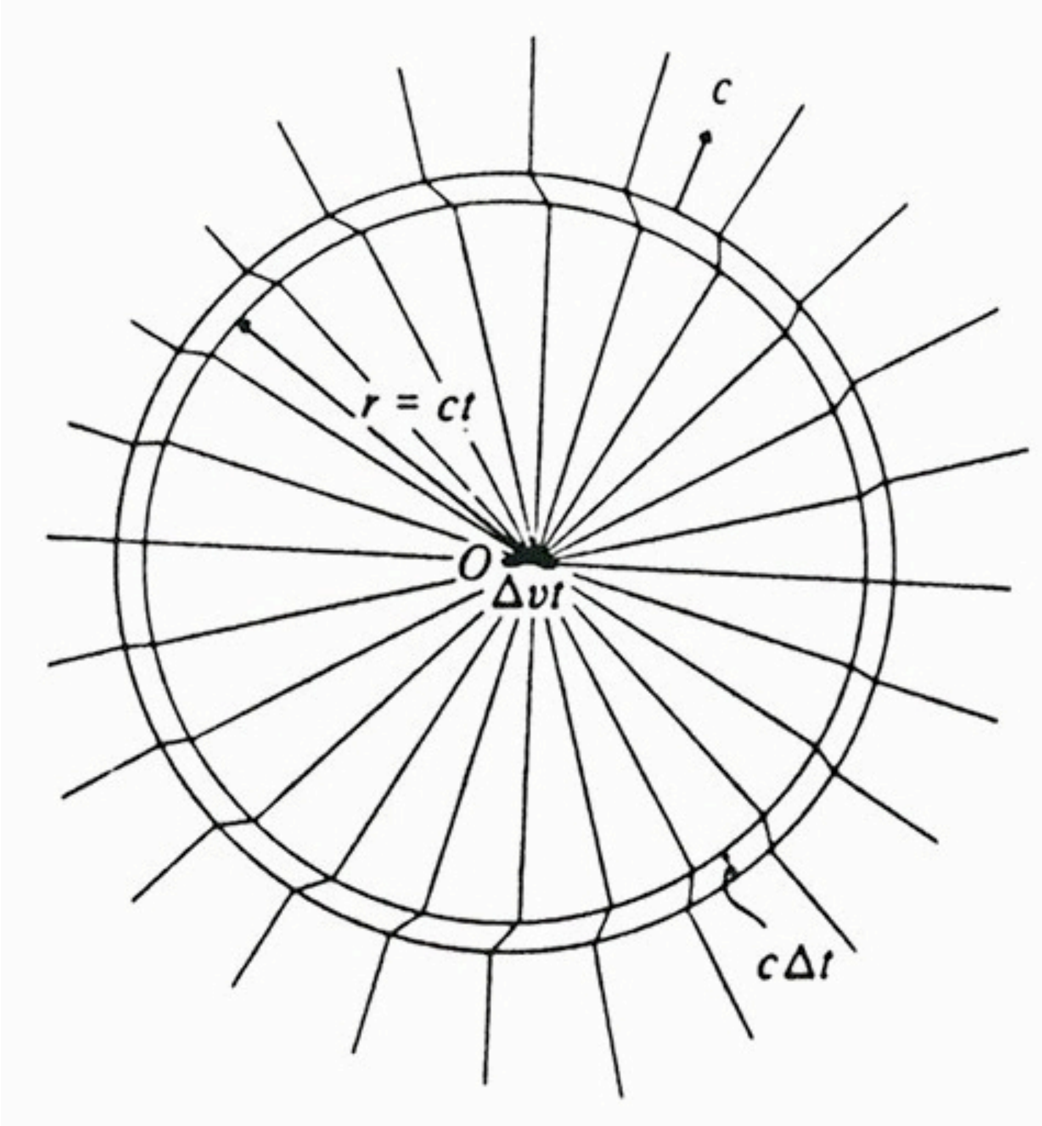


T. Miller et al (submitted)

H. Zheng et al. Science (2012)

Sources – How do you generate fields like this?

Why does an accelerated charge radiate?



Radiation from a dipole

$$E_{em} = \frac{1}{4\pi\varepsilon\varepsilon_0}\left[-\frac{d''}{r} + \frac{(d''\cdot r)\cdot r}{r^3} - \frac{d'}{r^2} + \frac{3(d'\cdot r)\cdot r}{r^4} - \frac{d}{r^3} + \frac{3(d\cdot r)\cdot r}{r^5} \right]$$

In the far field,

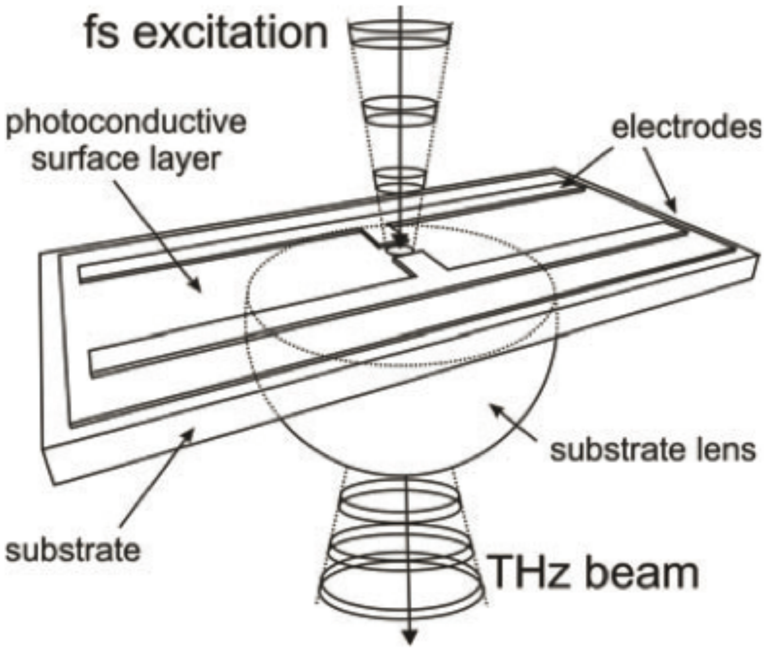
$$E_{em\perp} = -\frac{1}{4\pi\varepsilon\varepsilon_0}\frac{d''}{r}$$

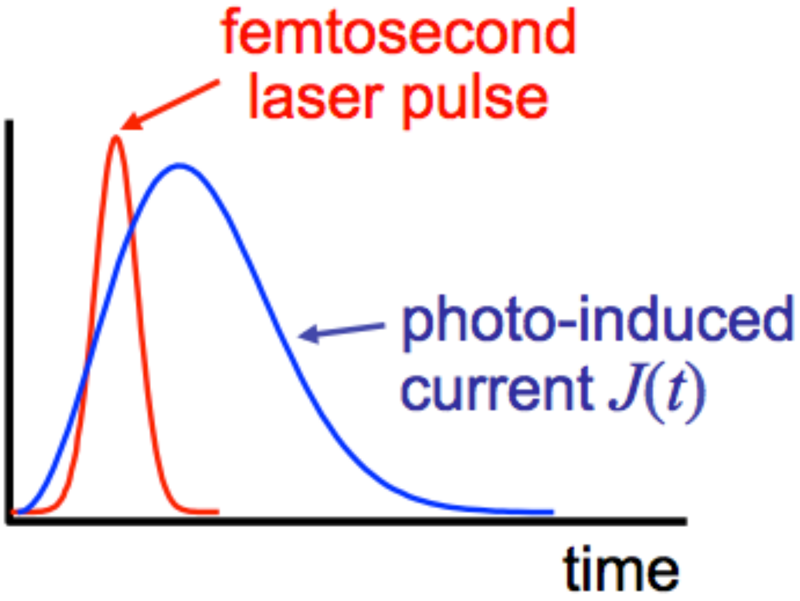
Now associated with a time-dependent polarization is a current:

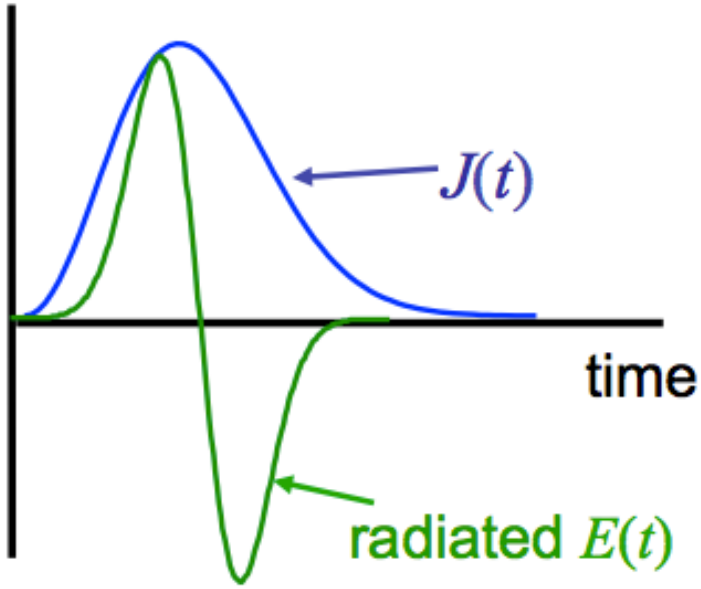
$$J(t) = \frac{\partial P}{\partial t}$$

So that:

$$E(t) = \frac{\partial J}{\partial t}$$







Quiz: How can an emitter like this be used as a detector?

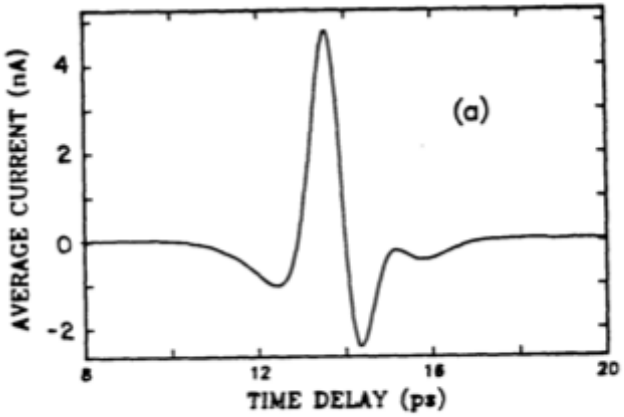
1642 J. Opt. Soc. Am. B/Vol. 8, No. 8/August 1991 H. Harde and D. Grischkowsky

Coherent transients excited by subpicosecond pulses of terahertz radiation

H. Harde* and D. Grischkowsky

IBM Watson Research Center, P.O. Box 218, Yorktown Heights, New York 10598

Received November 26, 1990; revised manuscript received March 1, 1991



OPTICS LETTERS / Vol. 18, No. 4 / February 15, 1993

Generation of high-power sub-single-cycle 500-fs electromagnetic pulses

D. You, R. R. Jones, and P. H. Bucksbaum

Department of Physics, University of Michigan, Ann Arbor, Michigan 48109-1120

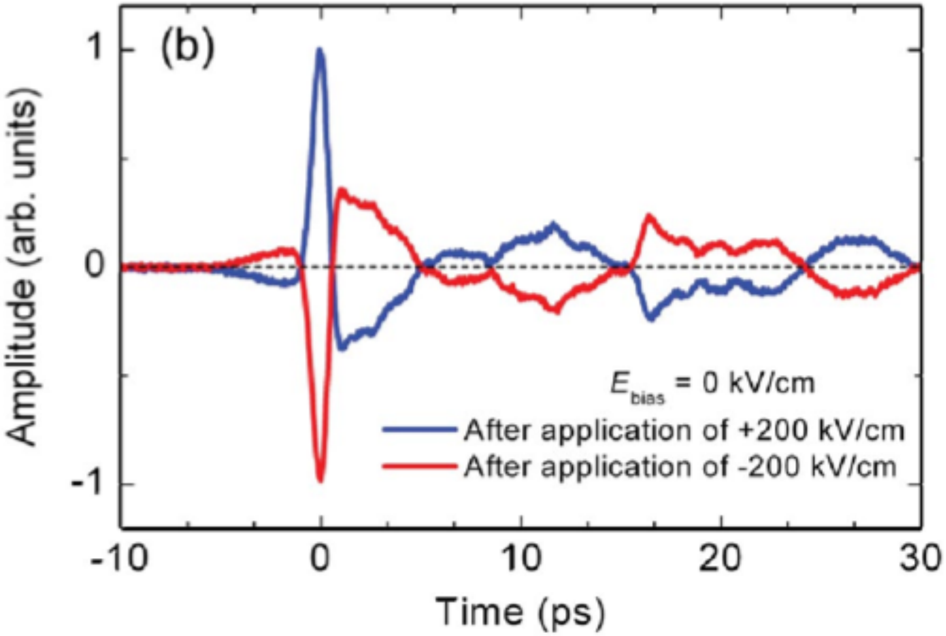
D. R. Dykaar

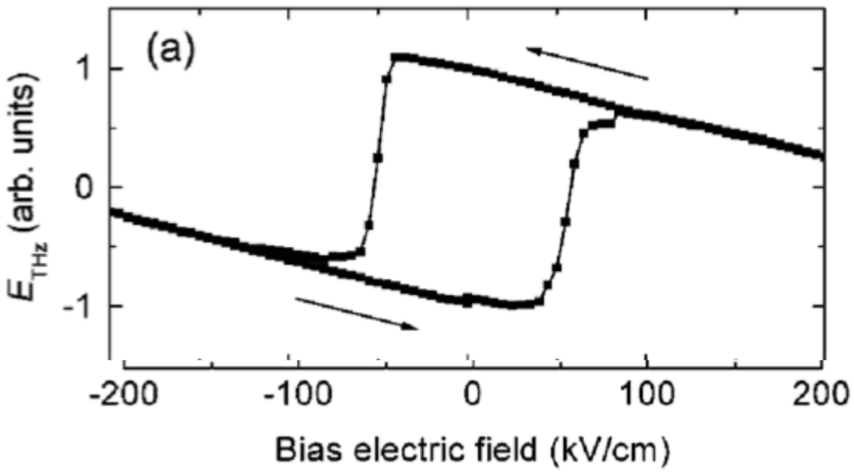
AT&T Bell Laboratories, Murray Hill, New Jersey 07974-2070

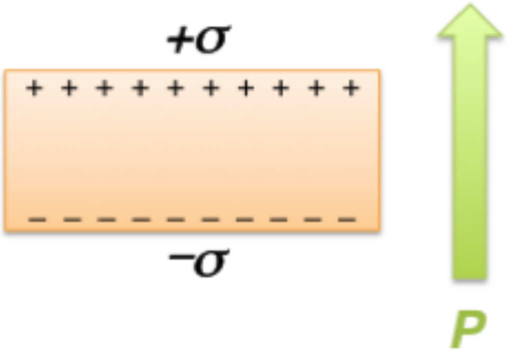
Received September 21, 1992

PRL **96,** 117402 (2006) PHYSICAL REVIEW LETTERS week ending 24 MARCH 2006

Terahertz Radiation by an Ultrafast Spontaneous Polarization Modulation of Multiferroic $BiFeO_3$ Thin Films







Takahashi et al, PRL(2006)

Nonlinear optical techniques

$$P_k = \varepsilon_0 \left(\chi^{(1)}_{ik} E_i + \chi^{(2)}_{ijk} E_i E_j + \chi^{(3)}_{ijlk} E_i E_j E_l + \ldots \right)$$

For a material with inversion symmetry, can send **r -> -r**

then: **E -> -E ; P -> -P**

which requires:

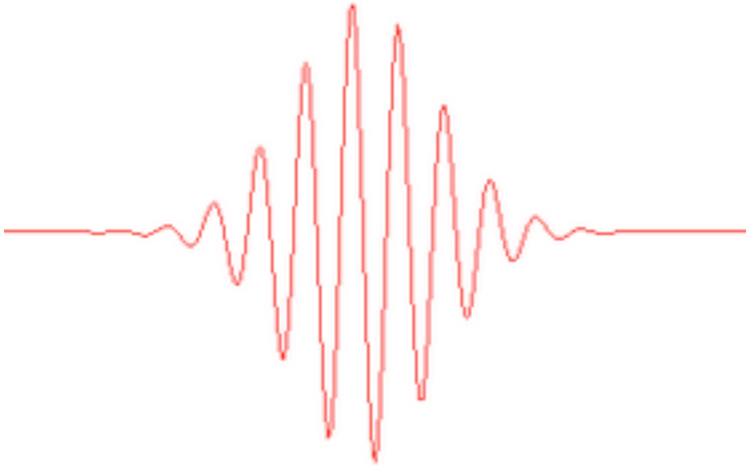
$$\chi^{(2)} = 0$$

For a material with non-zero second order nonlinear susceptibility, there are a few different possible mixing processes:

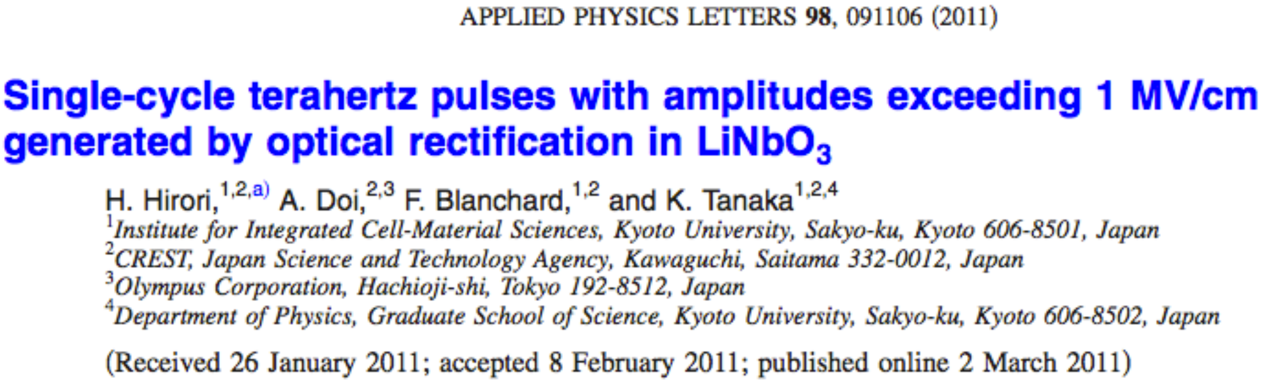
$$P_{2\omega} = \chi^{(2)}(\omega, \omega; \omega + \omega) E_\omega^2$$

$$P_0 = \chi^{(2)}(\omega, \omega; \omega - \omega) E_\omega^2$$

Quiz: For short pulses, how do you get THz pulses through this mechanism?



Optical rectification

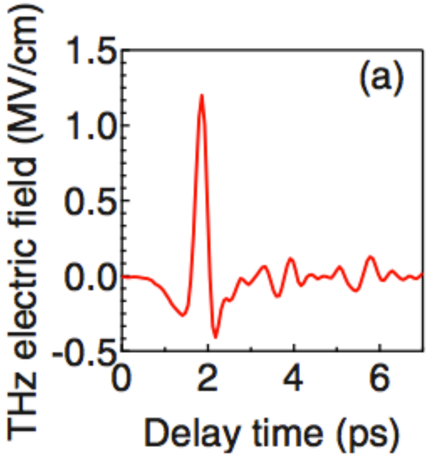


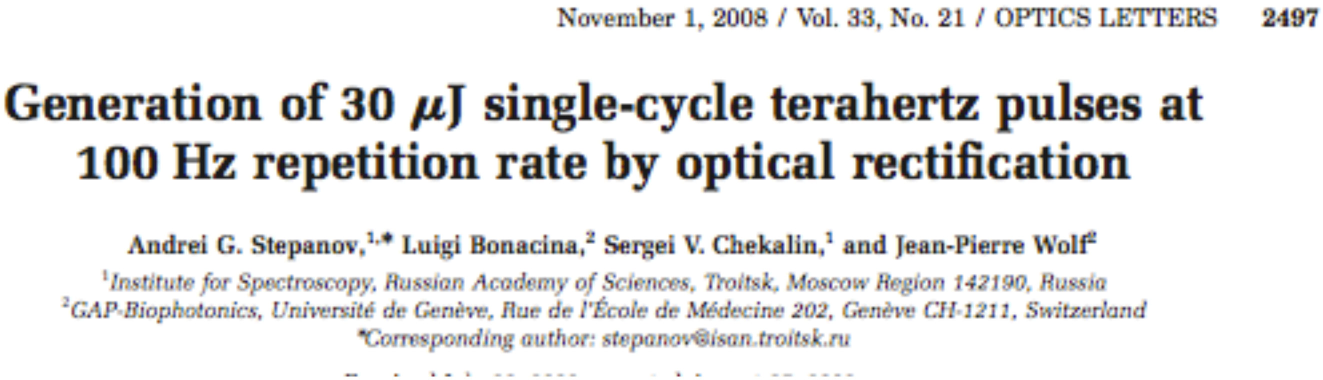
APPLIED PHYSICS LETTERS **98**, 091106 (2011)

Single-cycle terahertz pulses with amplitudes exceeding 1 MV/cm generated by optical rectification in $LiNbO_3$

H. Hirori,[1,2,a)] A. Doi,[2,3] F. Blanchard,[1,2] and K. Tanaka[1,2,4]
[1]*Institute for Integrated Cell-Material Sciences, Kyoto University, Sakyo-ku, Kyoto 606-8501, Japan*
[2]*CREST, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan*
[3]*Olympus Corporation, Hachioji-shi, Tokyo 192-8512, Japan*
[4]*Department of Physics, Graduate School of Science, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan*

(Received 26 January 2011; accepted 8 February 2011; published online 2 March 2011)

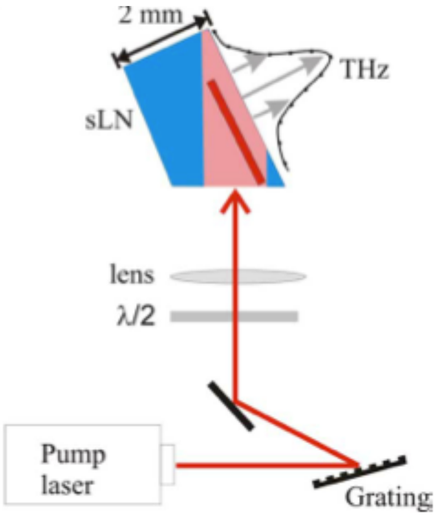




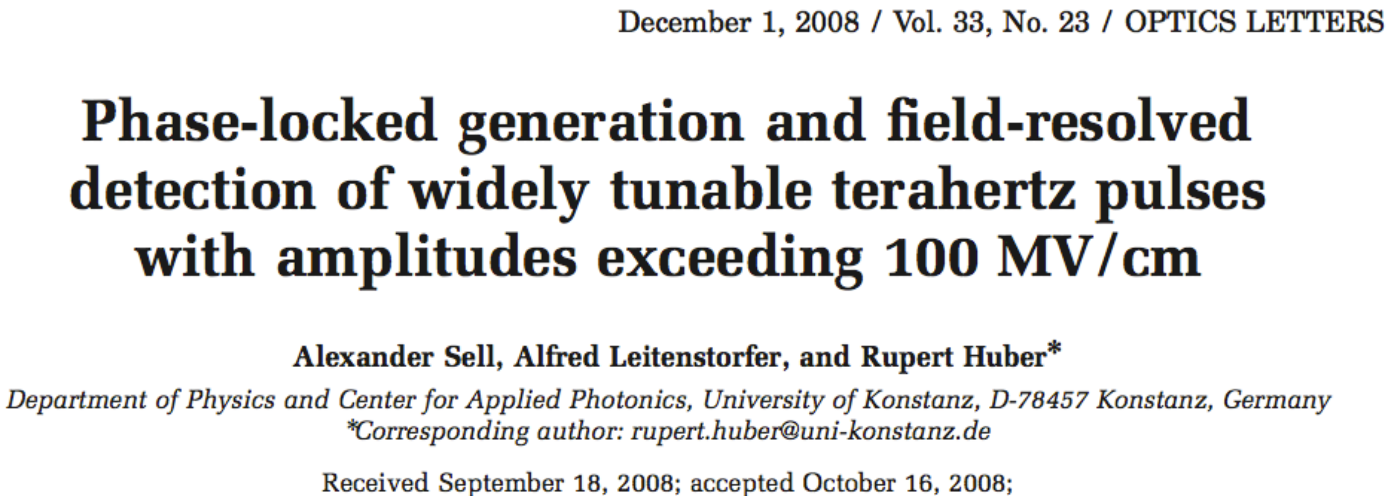
November 1, 2008 / Vol. 33, No. 21 / OPTICS LETTERS 2497

Generation of 30 μJ single-cycle terahertz pulses at 100 Hz repetition rate by optical rectification

Andrei G. Stepanov,[1,*] Luigi Bonacina,[2] Sergei V. Chekalin,[1] and Jean-Pierre Wolf[2]
[1]*Institute for Spectroscopy, Russian Academy of Sciences, Troitsk, Moscow Region 142190, Russia*
[2]*GAP-Biophotonics, Université de Genève, Rue de l'École de Médecine 202, Genève CH-1211, Switzerland*
Corresponding author: stepanov@isan.troitsk.ru



Difference frequency generation

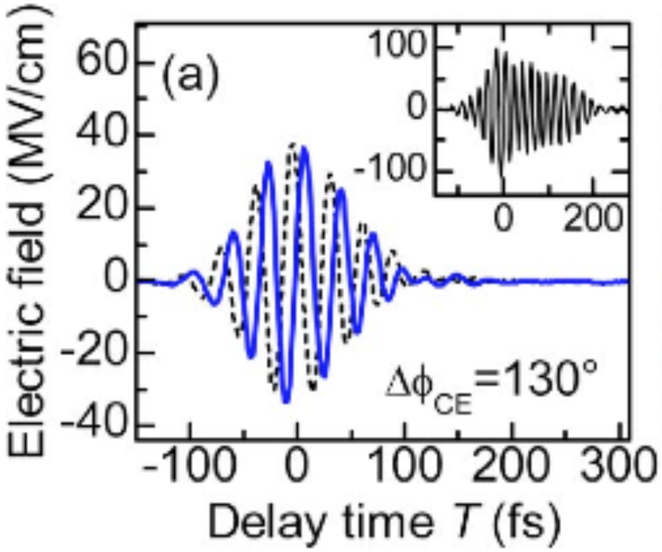


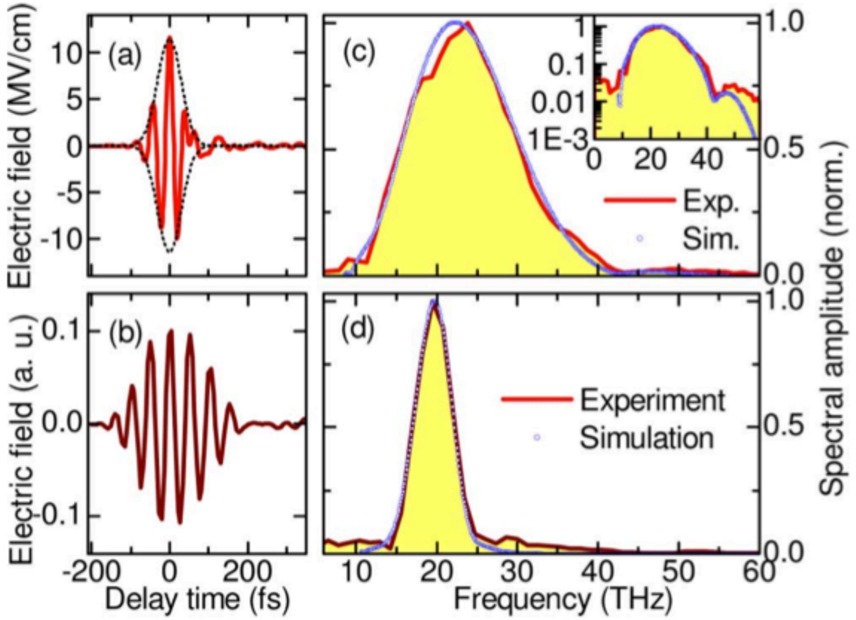
December 1, 2008 / Vol. 33, No. 23 / OPTICS LETTERS

Phase-locked generation and field-resolved detection of widely tunable terahertz pulses with amplitudes exceeding 100 MV/cm

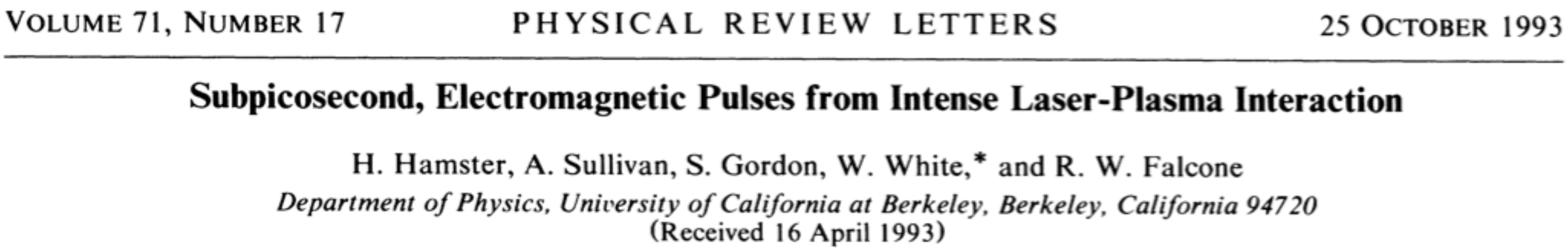
Alexander Sell, Alfred Leitenstorfer, and Rupert Huber*
Department of Physics and Center for Applied Photonics, University of Konstanz, D-78457 Konstanz, Germany
**Corresponding author: rupert.huber@uni-konstanz.de*

Received September 18, 2008; accepted October 16, 2008;



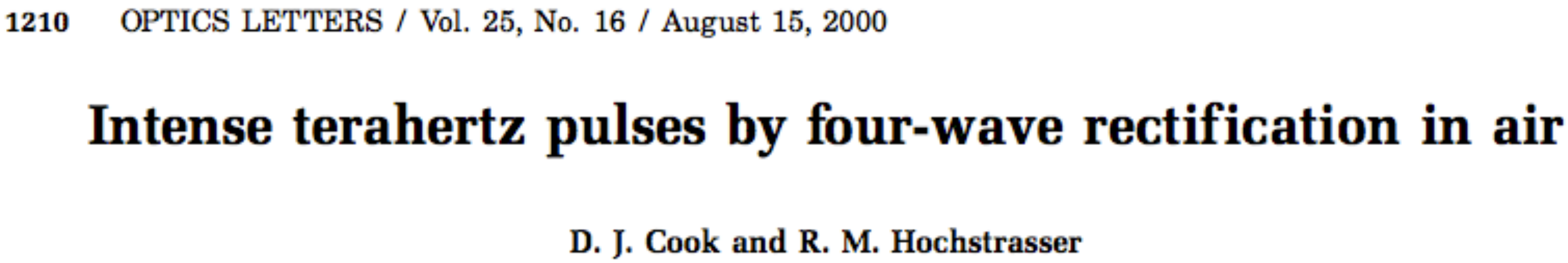


Overview of sources: Laser-produced plasmas


VOLUME 71, NUMBER 17 PHYSICAL REVIEW LETTERS 25 OCTOBER 1993

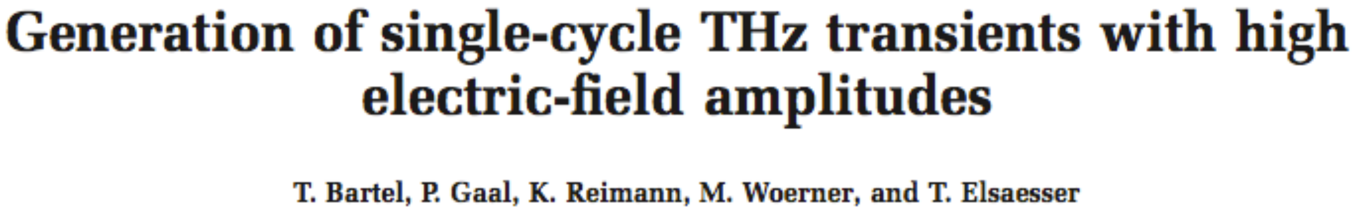
Subpicosecond, Electromagnetic Pulses from Intense Laser-Plasma Interaction

H. Hamster, A. Sullivan, S. Gordon, W. White,* and R. W. Falcone
Department of Physics, University of California at Berkeley, Berkeley, California 94720
(Received 16 April 1993)

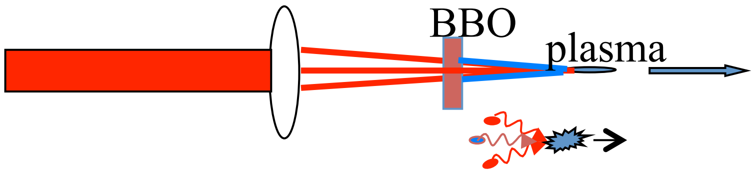

1210 OPTICS LETTERS / Vol. 25, No. 16 / August 15, 2000

Intense terahertz pulses by four-wave rectification in air

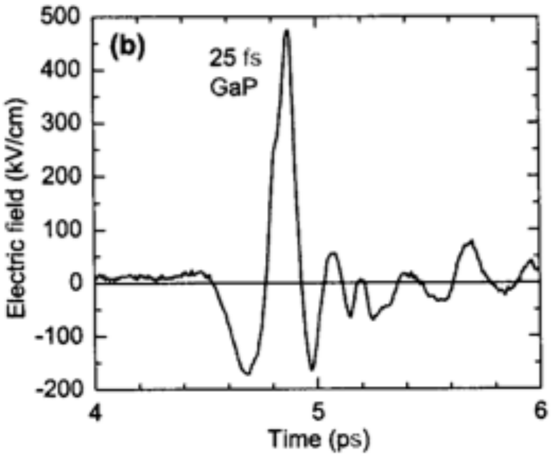
D. J. Cook and R. M. Hochstrasser


Generation of single-cycle THz transients with high electric-field amplitudes

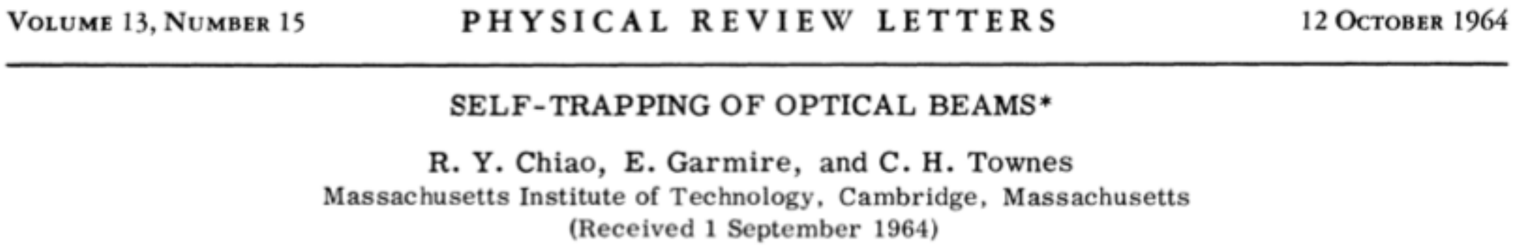
T. Bartel, P. Gaal, K. Reimann, M. Woerner, and T. Elsaesser



$$E_{THz} \propto \chi^{(3)} E_{2\omega} E_\omega E_\omega$$



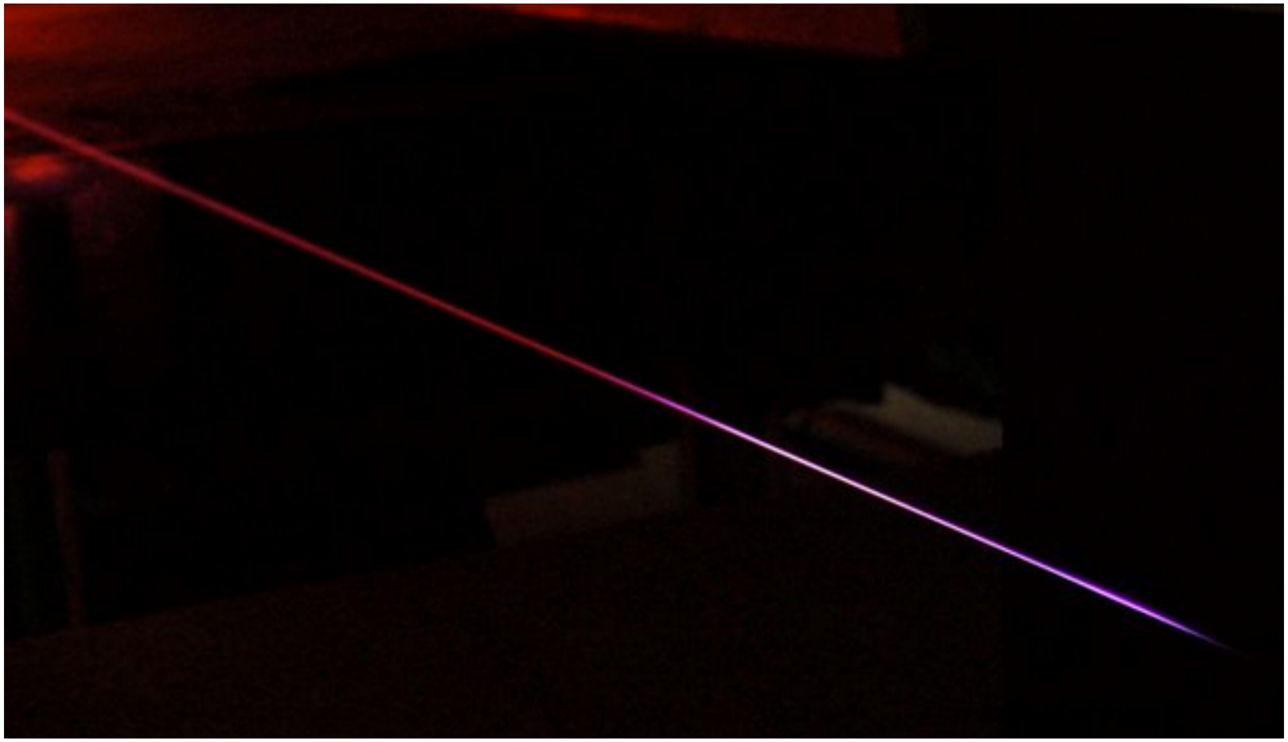
Femtosecond optical filaments



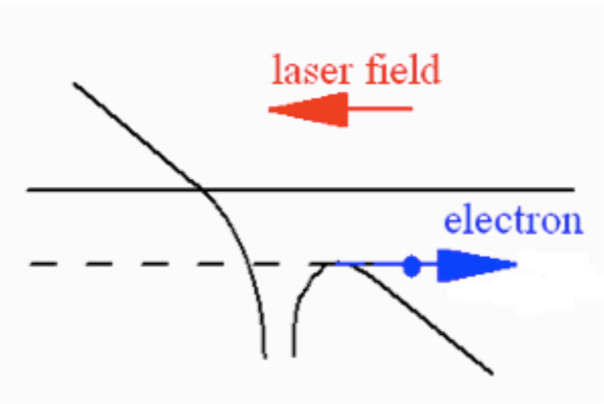
VOLUME 13, NUMBER 15 PHYSICAL REVIEW LETTERS 12 OCTOBER 1964

SELF-TRAPPING OF OPTICAL BEAMS*

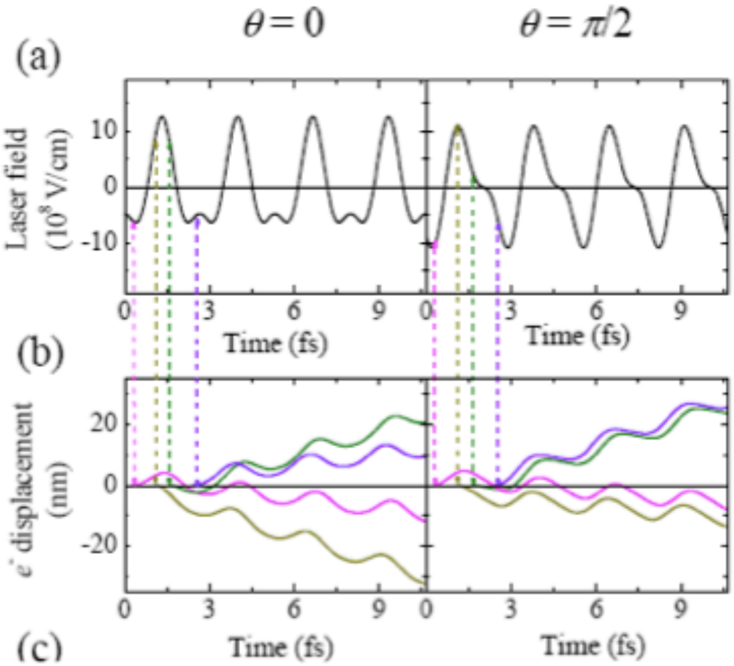
R. Y. Chiao, E. Garmire, and C. H. Townes
Massachusetts Institute of Technology, Cambridge, Massachusetts
(Received 1 September 1964)



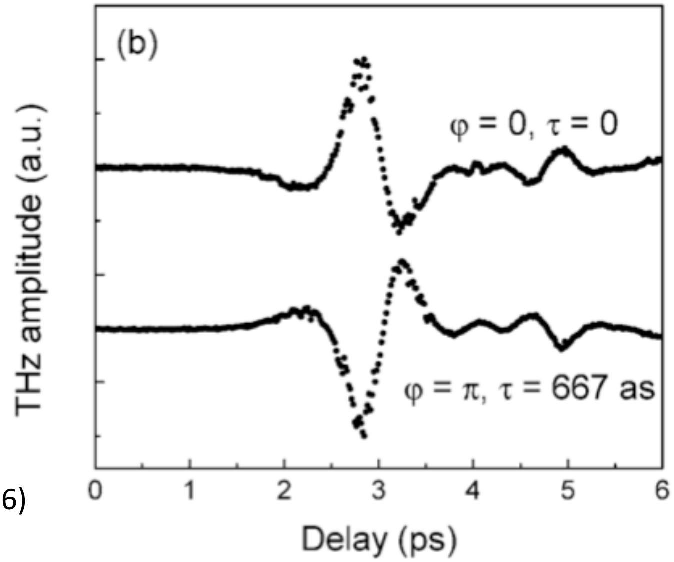
1D model of electron in asymmetric field



Optics Express, Kim et. al. **15**, 4577, (2007)

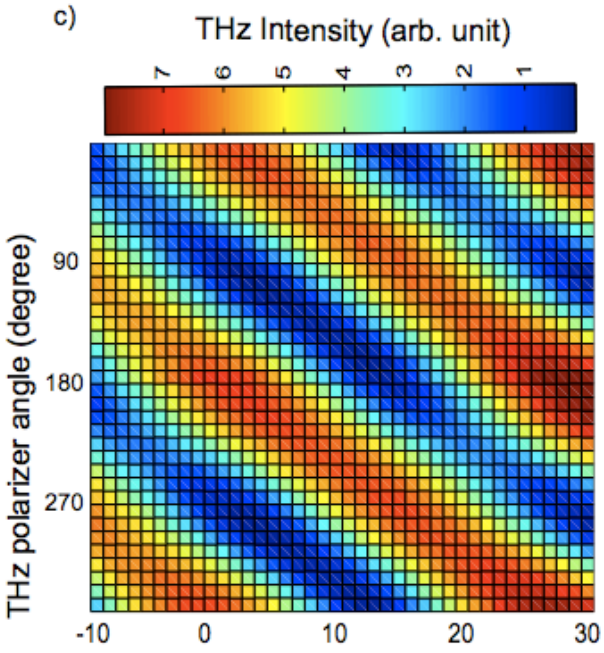


Phase control of THz polarity:

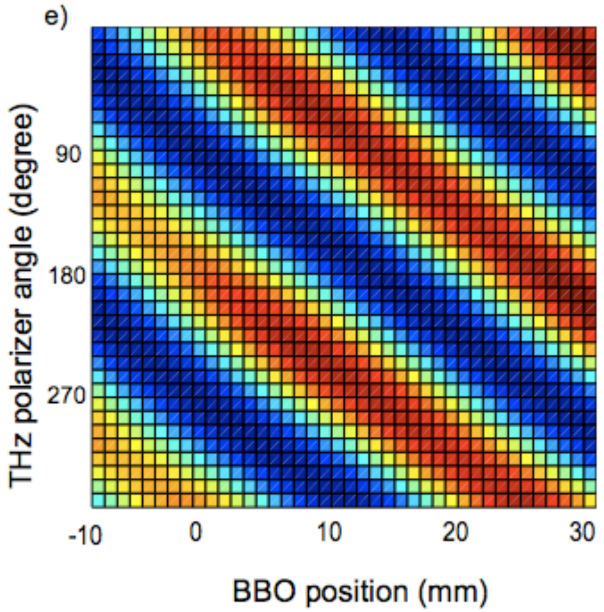


Xie et al., PRL (2006)

Electron trajectories in transverse plane



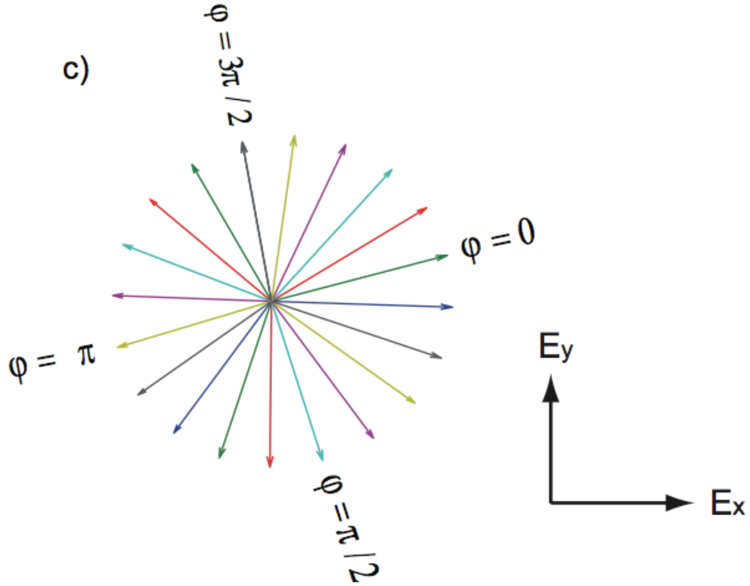
Experiment



Theory

$$\mathbf{J}(t) \propto \int_{-\infty}^{t} \dot{N}(t')\mathbf{v}(t,t')dt'$$

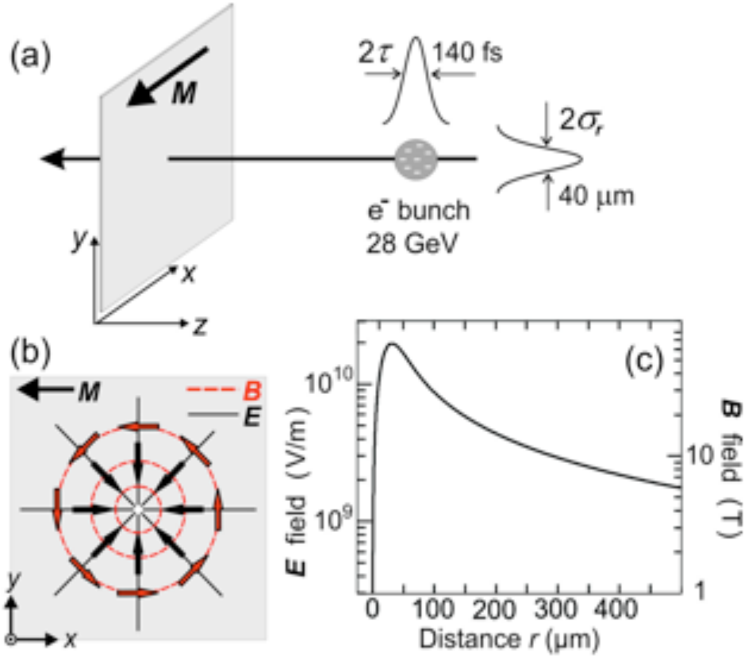
(sum over electron birth times)



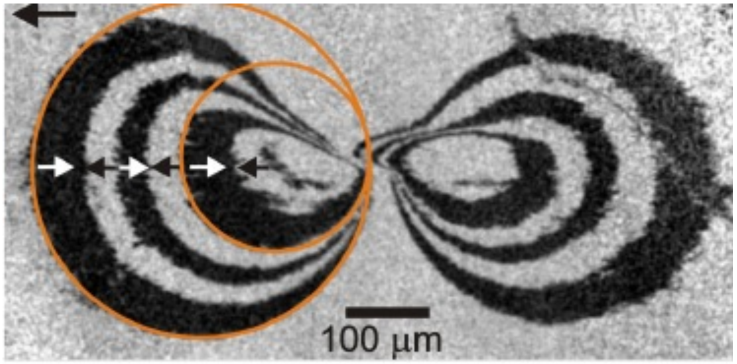
H. Wen et al. PRL (2009)
H. Wen et al. APL (2010)

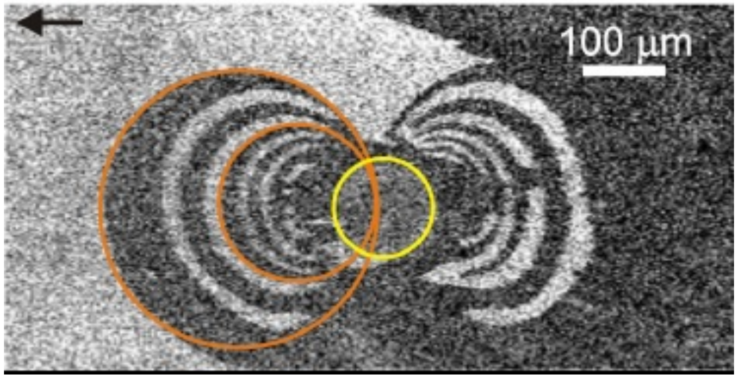
What can you do with strong THz fields?

Ultrafast magnetization switching by THz fields



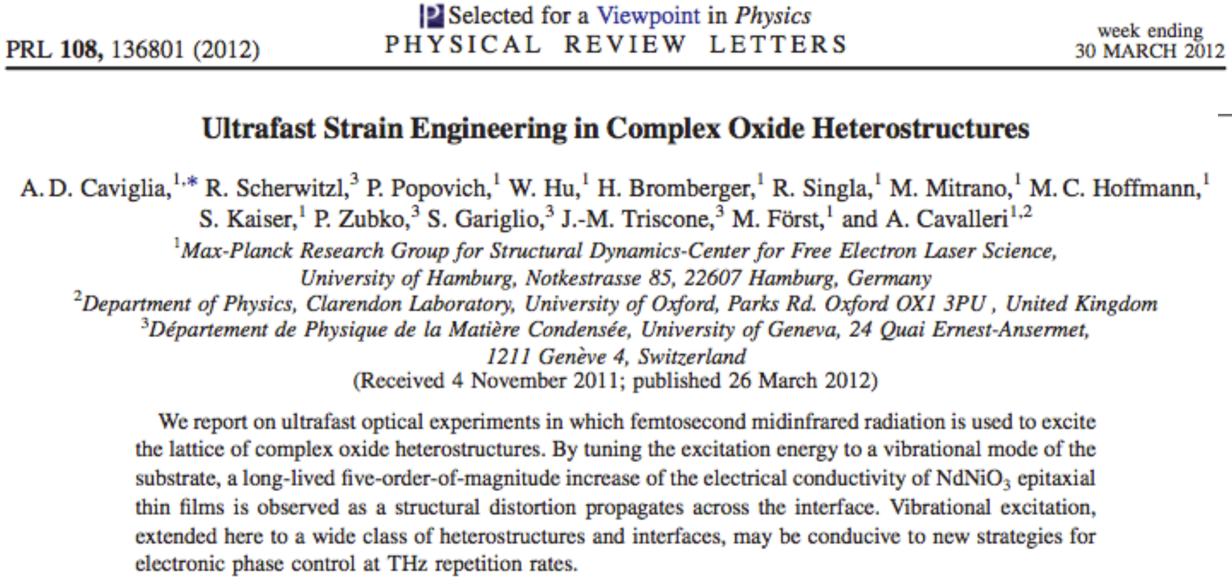
Compressed bunch (140 fs)





Uncompressed bunch (5 ps)

S. Gamble et al. Phys. Rev. Lett. (2009)
J. Stohr et al., Appl. Phys. Lett. (2009)
I. Tudosa et al., Nature (2004)



Selected for a Viewpoint in *Physics*
PRL **108,** 136801 (2012) PHYSICAL REVIEW LETTERS week ending 30 MARCH 2012

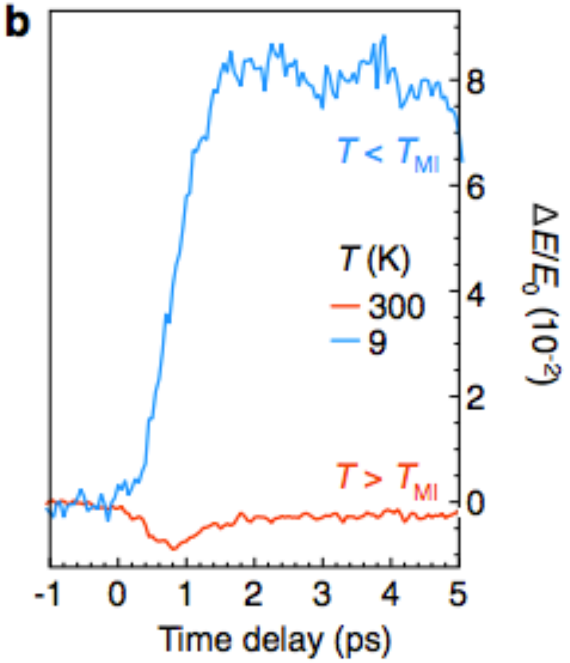
Ultrafast Strain Engineering in Complex Oxide Heterostructures

A. D. Caviglia,[1,*] R. Scherwitzl,[3] P. Popovich,[1] W. Hu,[1] H. Bromberger,[1] R. Singla,[1] M. Mitrano,[1] M. C. Hoffmann,[1] S. Kaiser,[1] P. Zubko,[3] S. Gariglio,[3] J.-M. Triscone,[3] M. Först,[1] and A. Cavalleri[1,2]
[1]*Max-Planck Research Group for Structural Dynamics-Center for Free Electron Laser Science, University of Hamburg, Notkestrasse 85, 22607 Hamburg, Germany*
[2]*Department of Physics, Clarendon Laboratory, University of Oxford, Parks Rd. Oxford OX1 3PU , United Kingdom*
[3]*Département de Physique de la Matière Condensée, University of Geneva, 24 Quai Ernest-Ansermet, 1211 Genève 4, Switzerland*

(Received 4 November 2011; published 26 March 2012)

We report on ultrafast optical experiments in which femtosecond midinfrared radiation is used to excite the lattice of complex oxide heterostructures. By tuning the excitation energy to a vibrational mode of the substrate, a long-lived five-order-of-magnitude increase of the electrical conductivity of $NdNiO_3$ epitaxial thin films is observed as a structural distortion propagates across the interface. Vibrational excitation, extended here to a wide class of heterostructures and interfaces, may be conducive to new strategies for electronic phase control at THz repetition rates.

$NdNiO_3$



LETTERS
PUBLISHED ONLINE: 7 AUGUST 2011 | DOI: 10.1038/NPHYS2055
nature physics

Nonlinear phononics as an ultrafast route to lattice control

M. Först[1]★, C. Manzoni[1†], S. Kaiser[1], Y. Tomioka[2], Y. Tokura[3], R. Merlin[4] and A. Cavalleri[1]★

Ultrafast large-amplitude relocation of electronic charge in ionic crystals

Flavio Zamponi, Philip Rothhardt, Johannes Stingl, Michael Woerner[1], and Thomas Elsaesser

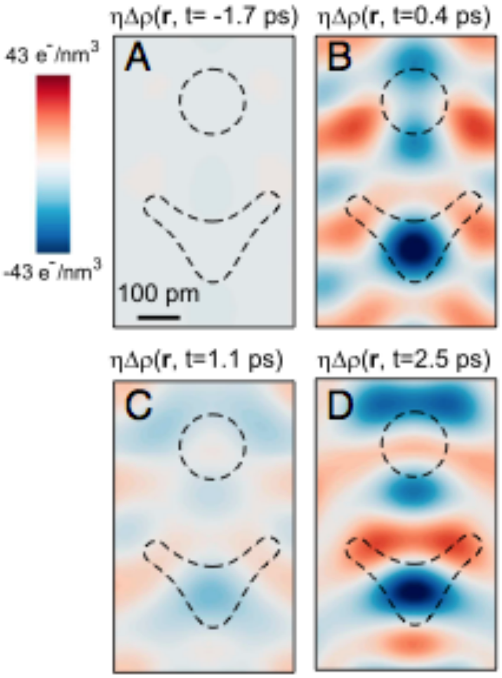
Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, 12489 Berlin, Germany

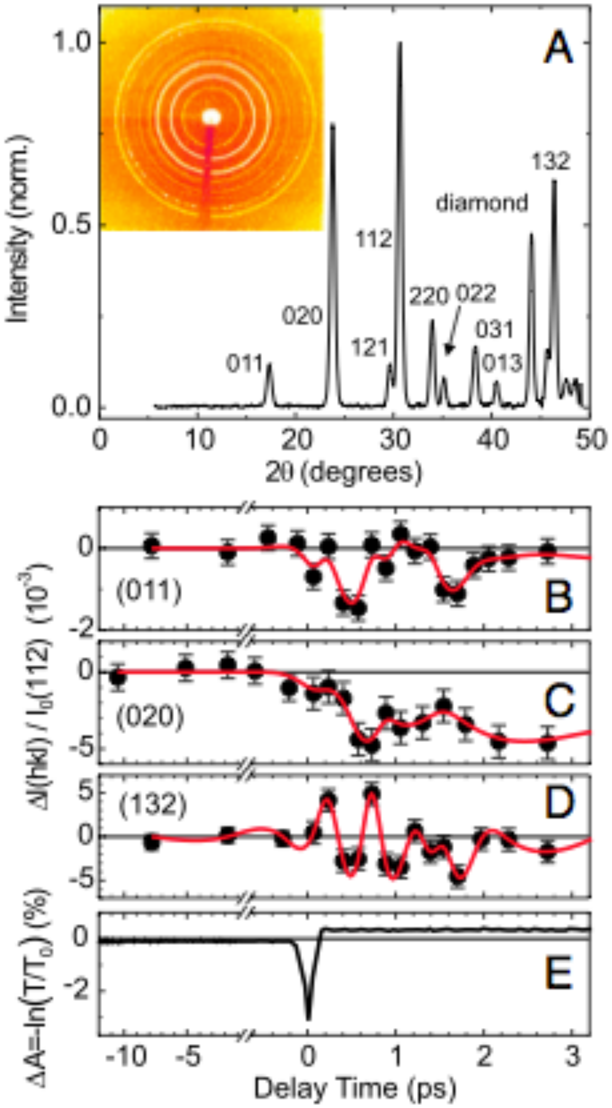
Edited by Franz Kaertner, Center for Free-Electron Laser Science, Germany, and accepted by the Editorial Board February 7, 2012 (received for review May 23, 2011)

The interplay of vibrational motion and electronic charge relocation in an ionic hydrogen-bonded crystal is mapped by X-ray powder diffraction with a 100 fs time resolution. Photoexcitation of the prototype material KH_2PO_4 induces coherent low-frequency motions of the PO_4 tetrahedra in the electronically excited state of the crystal while the average atomic positions remain unchanged. Time-dependent maps of electron density derived from the diffraction data demonstrate an oscillatory relocation of electronic charge with a spatial amplitude two orders of magnitude larger than the underlying vibrational lattice motions. Coherent longitudinal optical and tranverse optical phonon motions that dephase on a time scale of several picoseconds, drive the charge relocation, similar to a soft (transverse optical) mode driven phase transition between the ferro- and paraelectric phase of KH_2PO_4.

of 100 fs duration (15, 16) from the excited powder sample. The pattern of diffracted X-rays consists of Debye–Scherrer diffraction rings that are recorded with a large-area CCD detector. A series of such patterns measured at different delay times after excitation allows for reconstructing the momentary nuclear positions and charge distributions. Further details of the experiment and the methods applied for data analysis are discussed in *Materials and Methods*.

Results of the femtosecond experiments are summarized in Fig. 2. Fig. 2*A*, *Inset* shows a diffraction pattern of the unexcited KDP powder sample as recorded with the femtosecond X-ray pulses. For an integration time as short as 7 min, one clearly distinguishes 11 different Debye–Scherrer rings. In Fig. 2*A*, the diffracted intensity integrated over the individual rings is plotted as a function of 2*θ*, *θ* being the diffraction angle. For an assignment





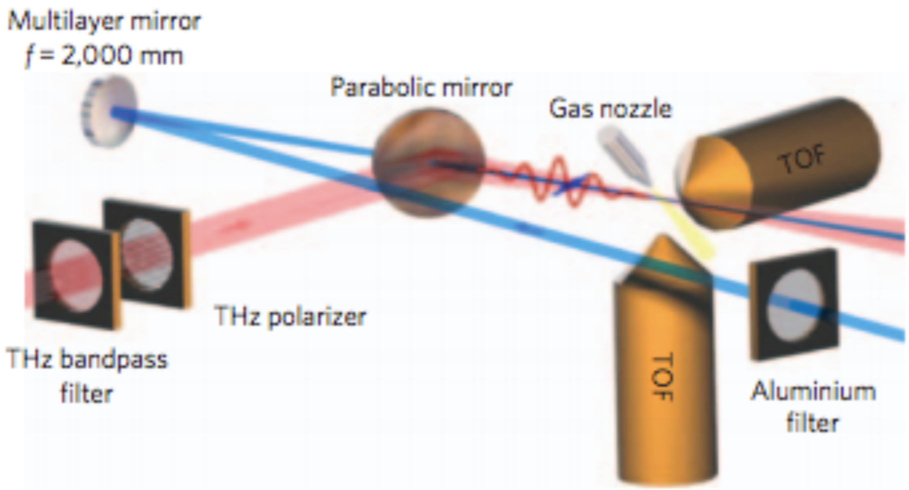
nature photonics

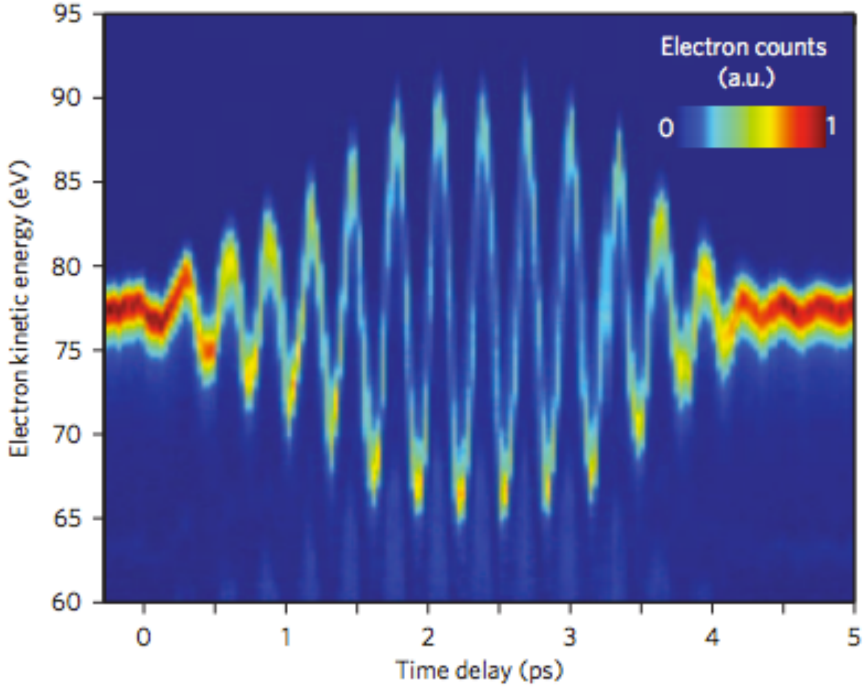
ARTICLES

PUBLISHED ONLINE: 23 AUGUST 2009 | DOI: 10.1038/NPHOTON.2009.160

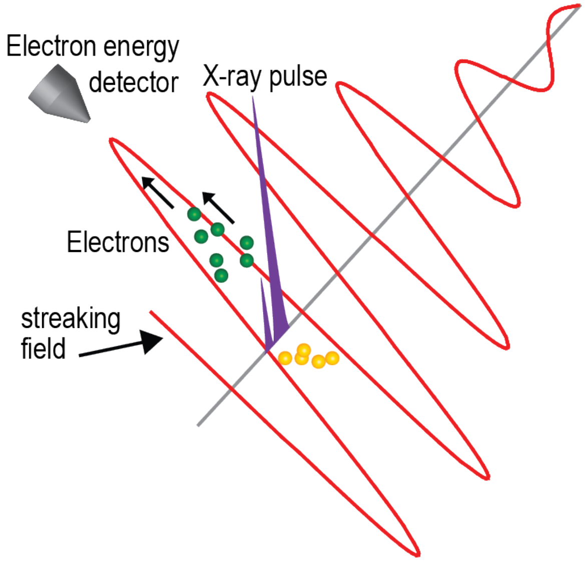
Single-shot terahertz-field-driven X-ray streak camera

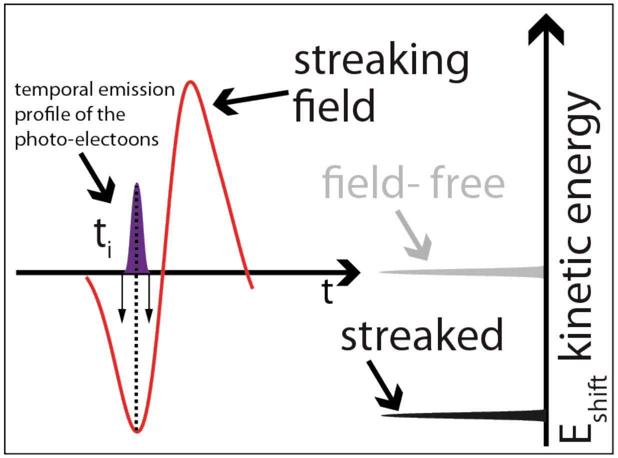
Ulrike Frühling[1], Marek Wieland[2], Michael Gensch[1,3], Thomas Gebert[2], Bernd Schütte[2], Maria Krikunova[2], Roland Kalms[2], Filip Budzyn[2], Oliver Grimm[2,4], Jörg Rossbach[2], Elke Plönjes[1] and Markus Drescher[2]*





FEL pulse characterization with THz streaking





- X-ray induced photoemission in a **gas target**
- **Photoemission spectrum** is replica of the temporal structure of the ionizing pulse
- Momentum/kinetic energy distribution of photoelectrons is affected by the overlapping **streaking field**.
- width of observed spectrum depends on the **duration of FEL pulse**/photoemission and strength of field
- If the field is characterized, photoemission spectrum can be **directly mapped to the FEL pulse profile**

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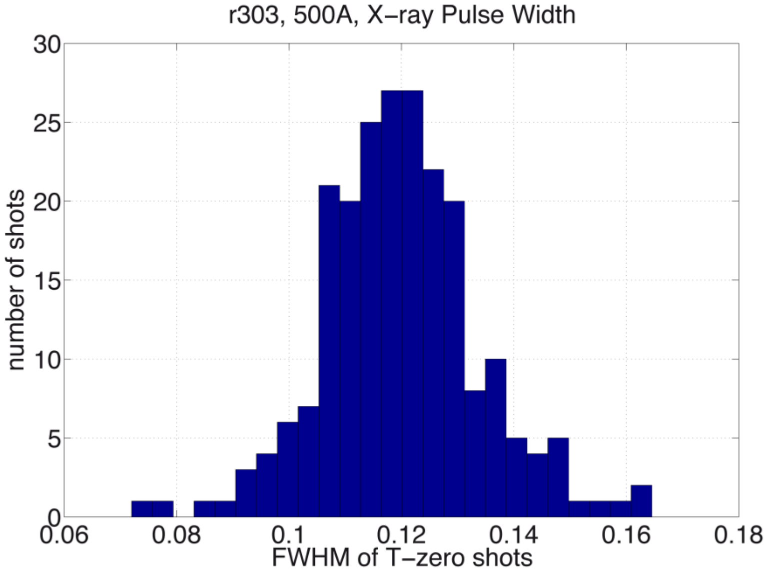
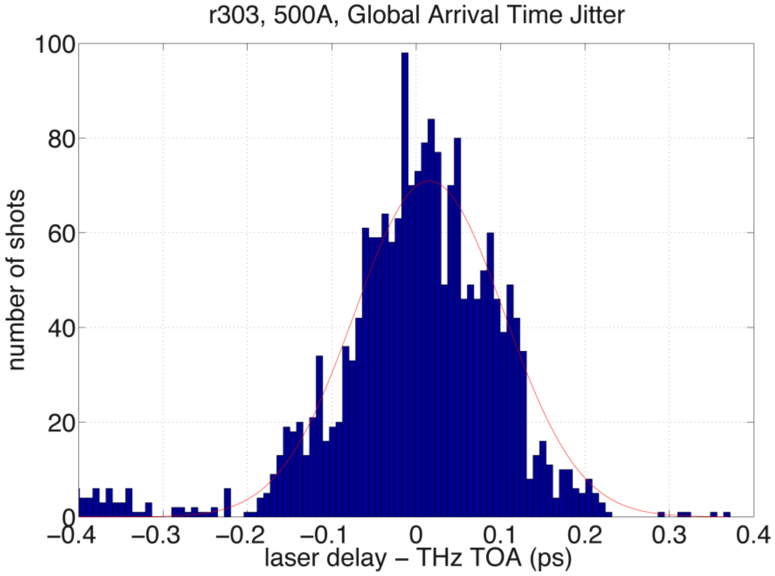
Results: X-ray Pulse Width and Time-of-Arrival



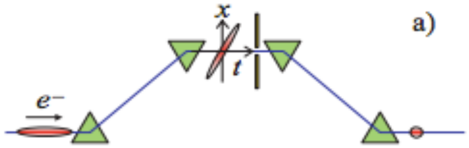


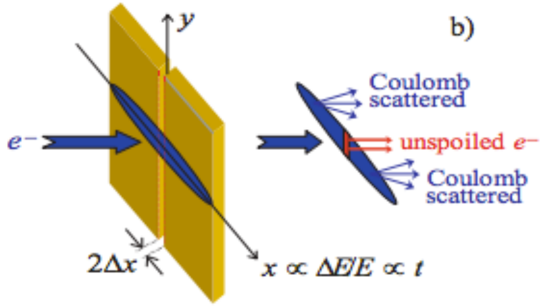
- Full charge (~ 150pC, undercompressed (~ 500A))
- Time-of-Arrival jitter ~ 90fs rms
- X-ray pulse feature ~ 120 fs FWHM (without deconvolution of instrument resolution!)

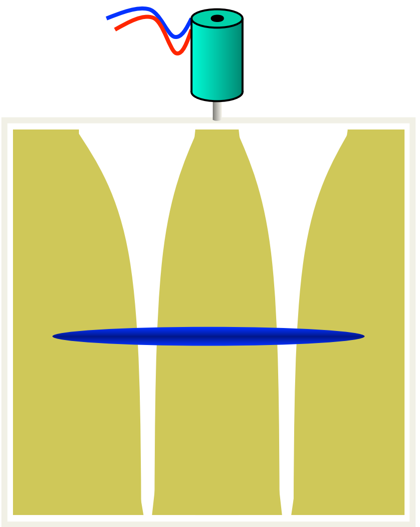
A. Cavalieri, H. Schlarb, M. Hoffmann

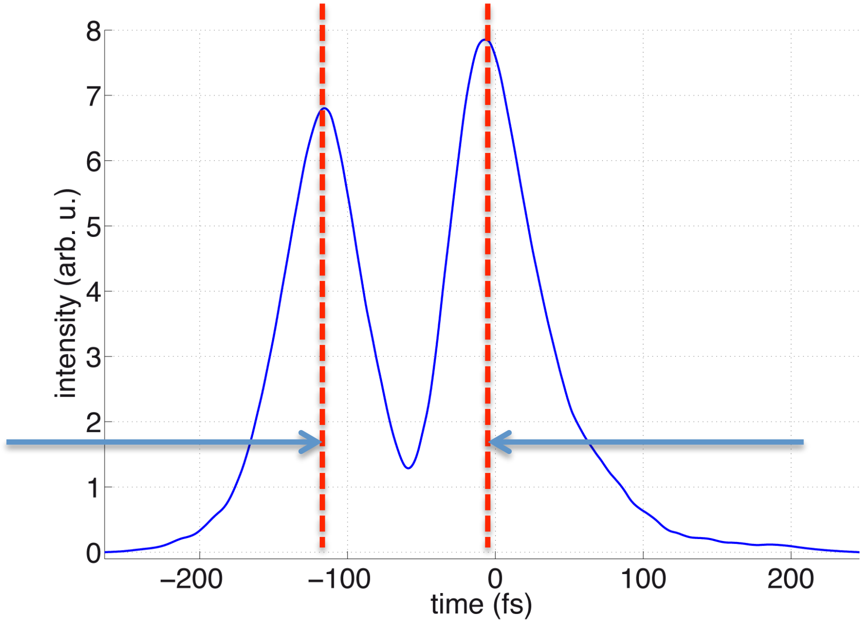
Temporal profile of X-ray double pulses

- “slotted spoiler” produces X-ray double pulses with variable separation
- THz-streaking reveals FEL sub-structure









Peak Separation : 105 ± 9 fs

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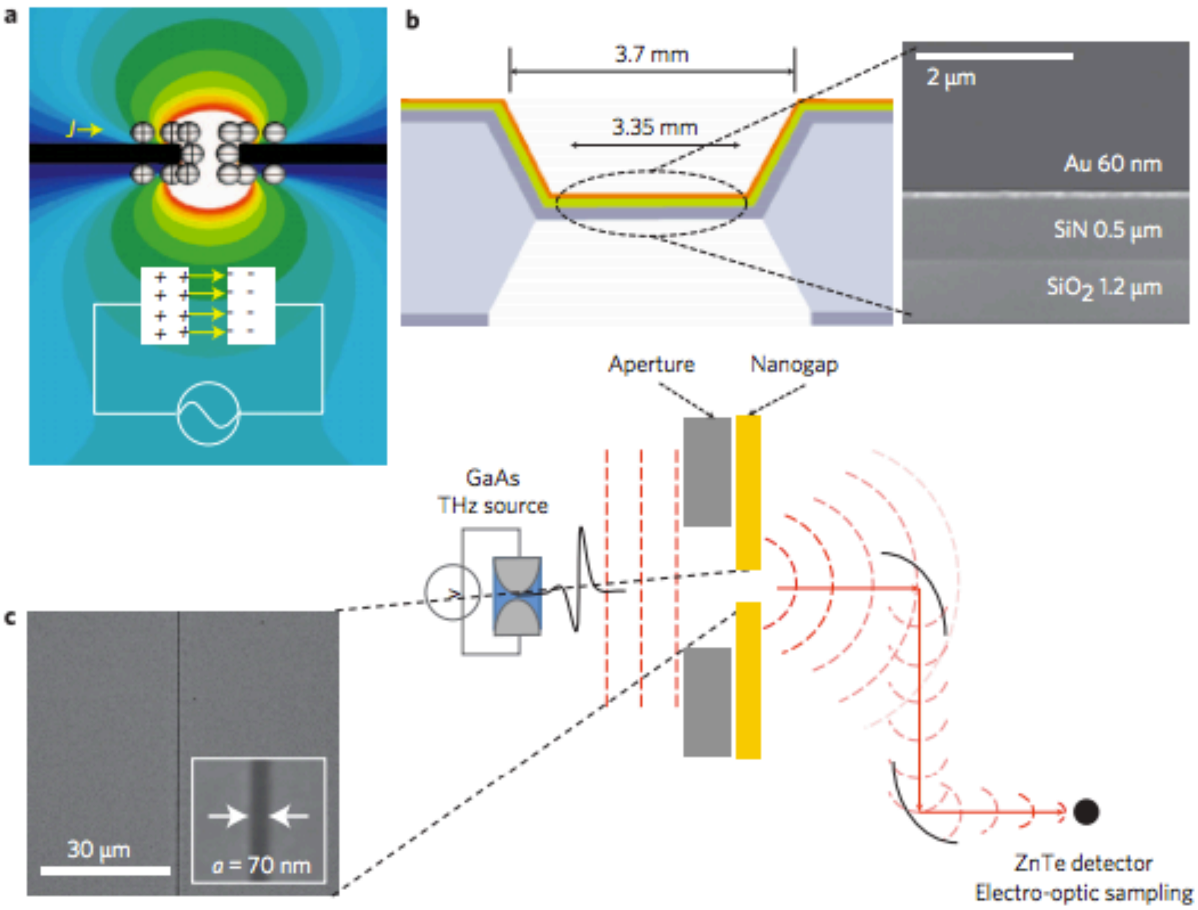
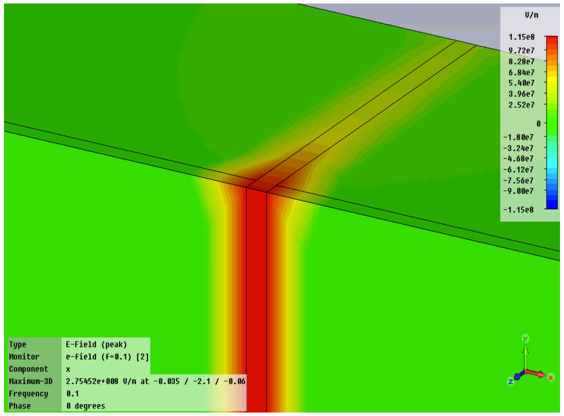
nature photonics
LETTERS
PUBLISHED ONLINE: 22 FEBRUARY 2009 | DOI: 10.1038/NPHOTON.2009.22

Terahertz field enhancement by a metallic nano slit operating beyond the skin-depth limit

M. A. Seo[1], H. R. Park[1], S. M. Koo[2], D. J. Park[1], J. H. Kang[3], O. K. Suwal[4], S. S. Choi[4], P. C. M. Planken[5], G. S. Park[1], N. K. Park[2], Q. H. Park[3]* and D. S. Kim[1]*



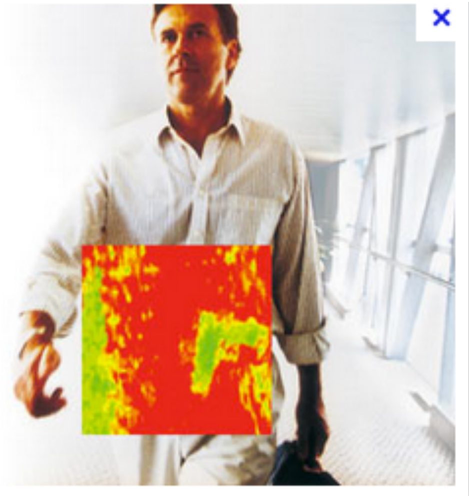


Type	E-Field (peak)
Monitor	e-field (f=0.1) [2]
Component	x
Maximum-3D	2.75452e+008 V/m at -0.035 / -2.1 / -0.06
Frequency	0.1
Phase	0 degrees

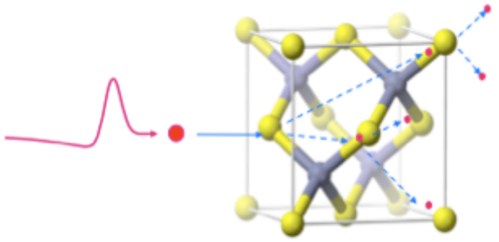
JOURNAL OF APPLIED PHYSICS **108**, 063304 (2010)

Detecting excess ionizing radiation by electromagnetic breakdown of air

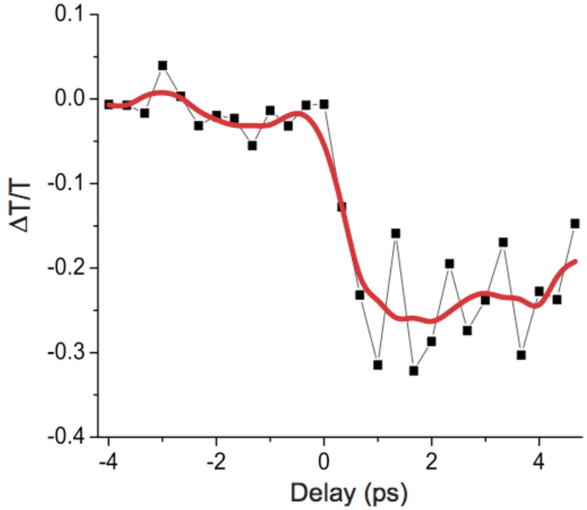
Victor L. Granatstein[a)] and Gregory S. Nusinovich
Center for Applied Electromagnetics, Institute for Research in Electronics and Applied Physics, University of Maryland, College Park, Maryland 20742, USA

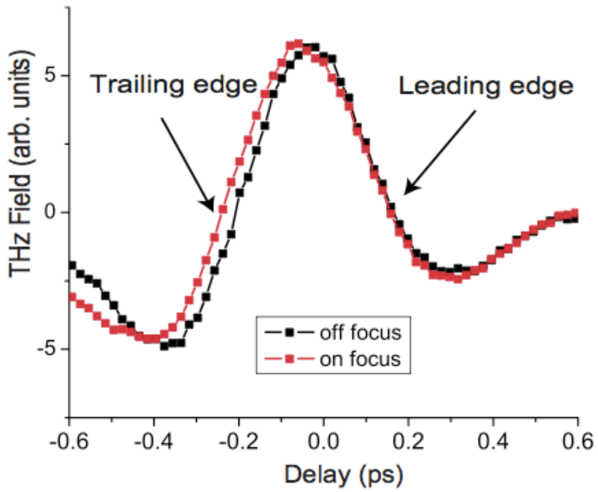


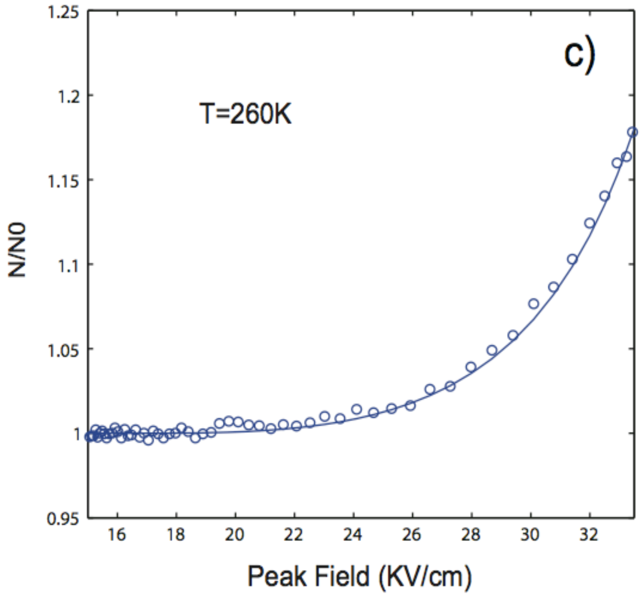
THz-induced breakdown processes and directing charges in materials



InSb







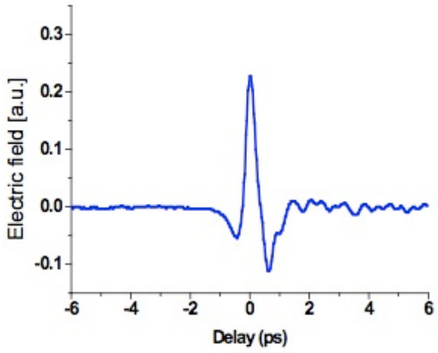
H. Wen et al. PRB (2008)

Microscopic model of avalanche processes in THz field.

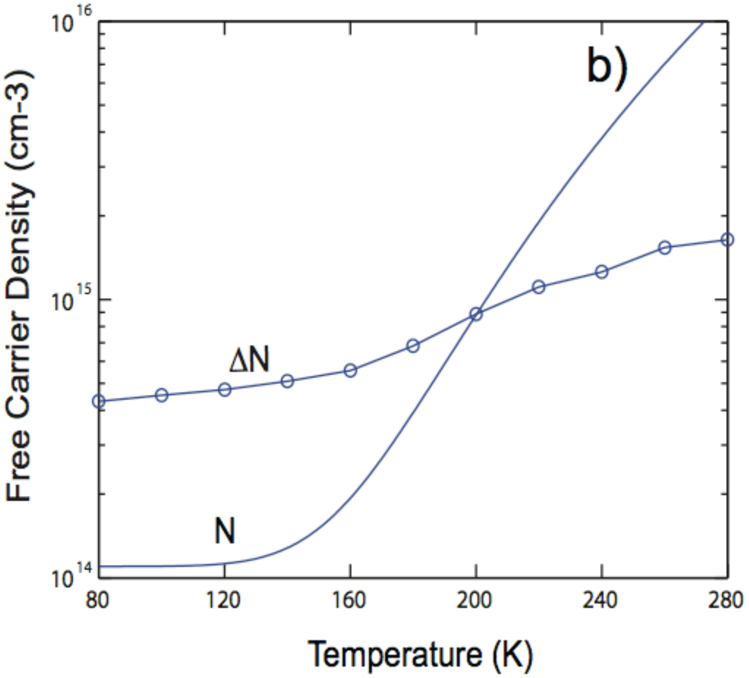
$$\frac{dN(t)}{dt} = N(t)\int_{-\infty}^{\infty} f(E,t)\varpi(E)\,dE$$

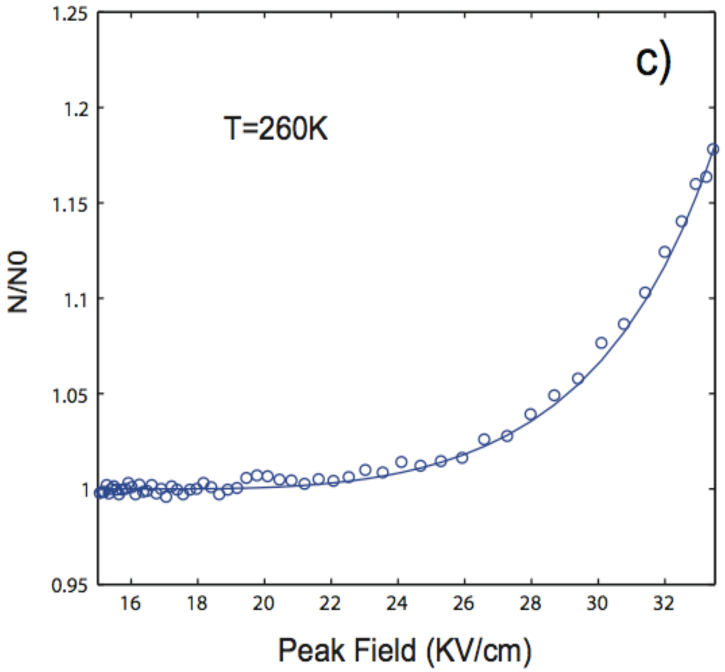
distribution function

ionization rate

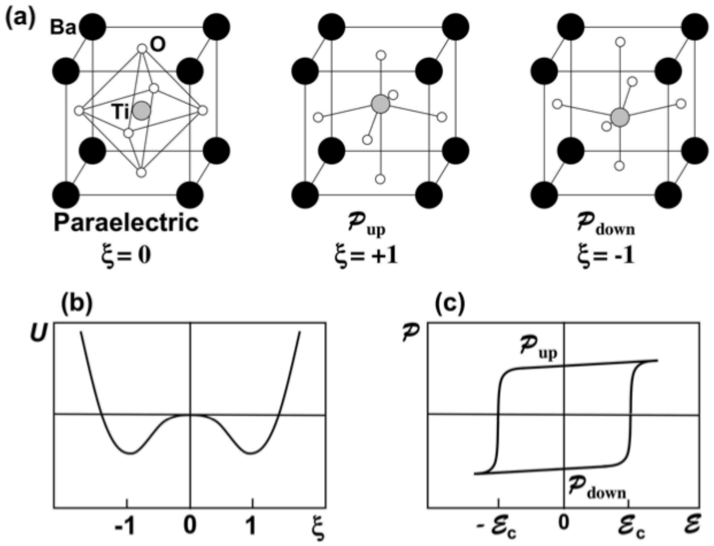


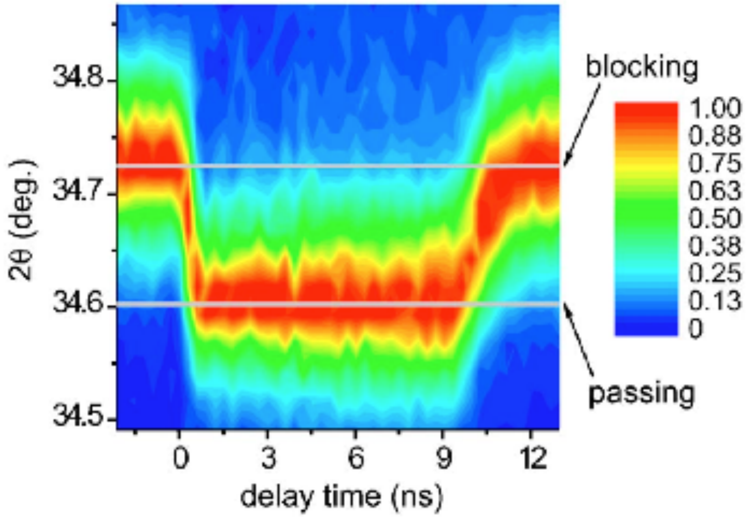
$$E(x,t) = \frac{\left(\int_0^t eF(x,t')dt'\right)^2}{2m^*} \longrightarrow N(t) = \frac{N(0)}{d}\int_0^d e^{\int_0^t \varpi(z,t')dt'}\,dz.$$



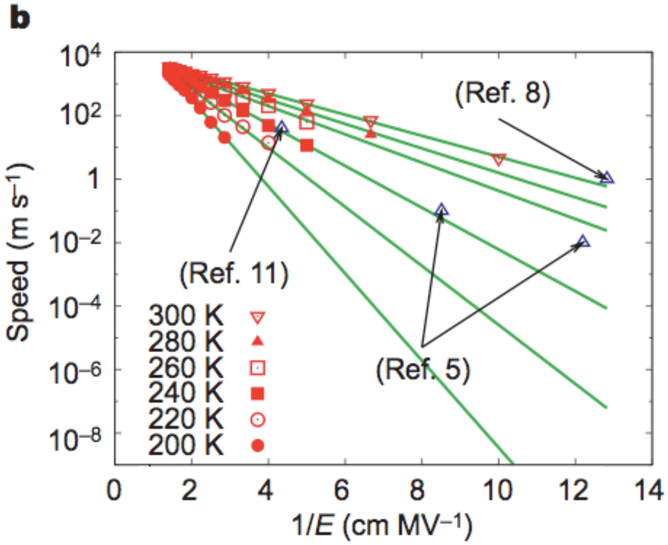


Ferroelectric Dynamics

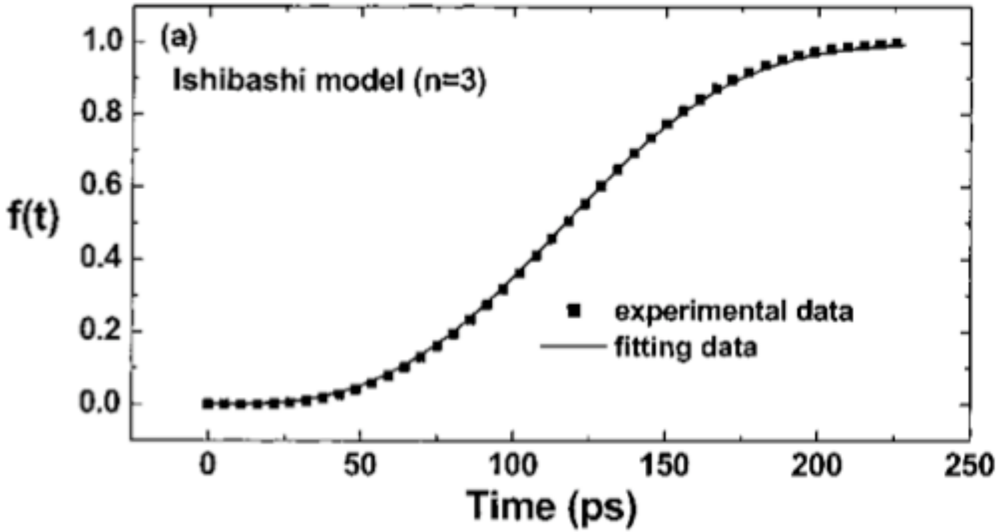




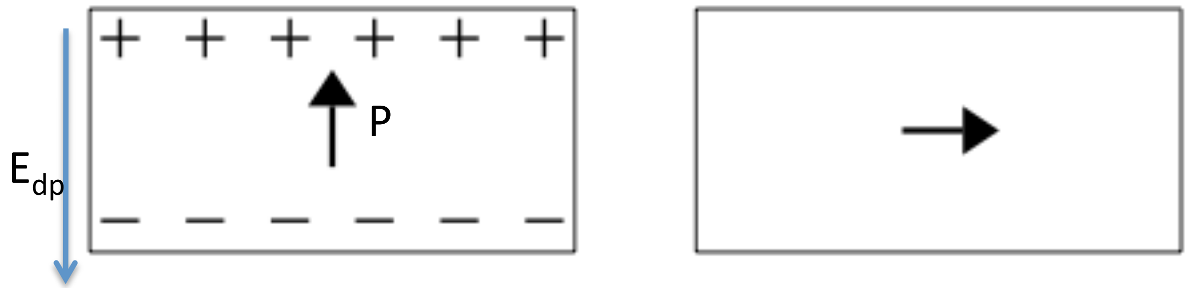
Grigoriev et al., Appl. Phys. Lett. (2006)



Y.-H. Shin et al., Nature (2007)



Li et al, Appl. Phys. Lett. (2004)



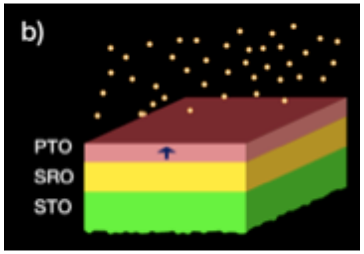
-Polarization leads to surface charge:

$$\sigma = \mathbf{P} \cdot \mathbf{n}$$

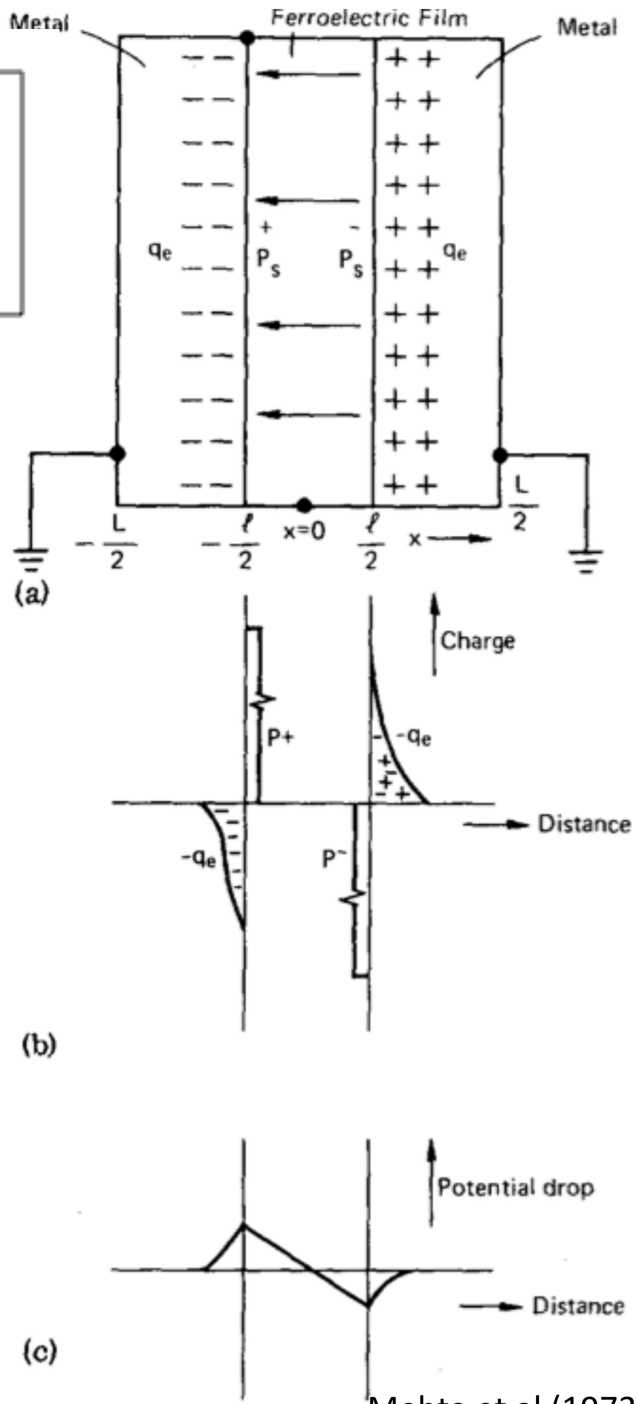
and therefore a field opposite to the polarization direction, called the depolarizing field.

-This can be compensated for by free charges as in the case of a ferroelectric capacitor, by adsorbed ions, or by domain formation to minimize the free energy.

-Nevertheless, ferroelectricity is maintained at nanolayer thicknesses approaching that of a single unit cell.

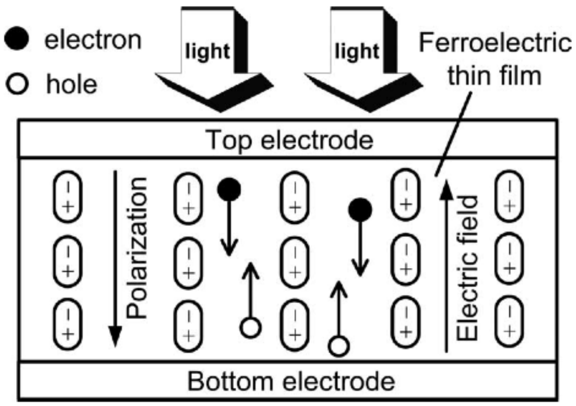


Stephenson et al.

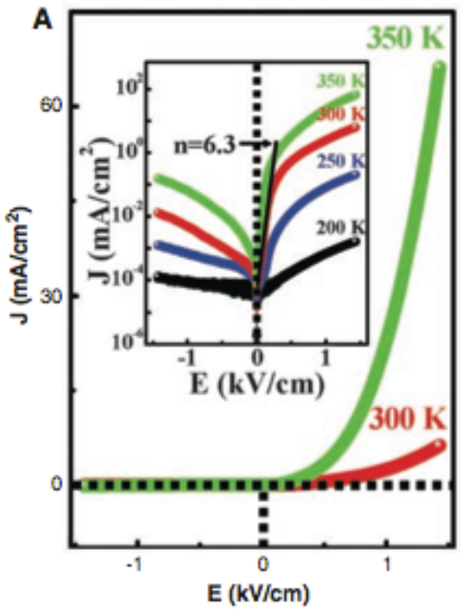


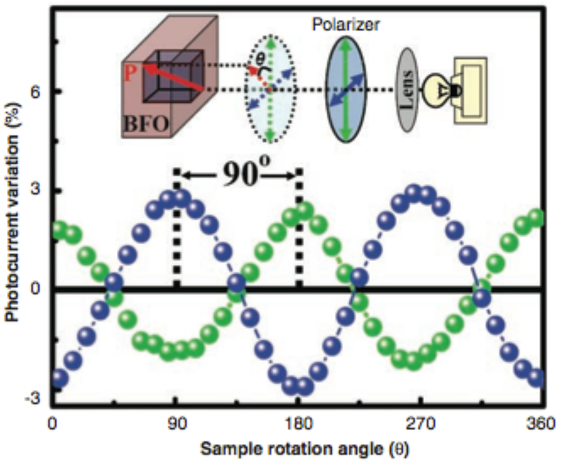
Mehta et al (1973)

Ferroelectric photovoltaic response

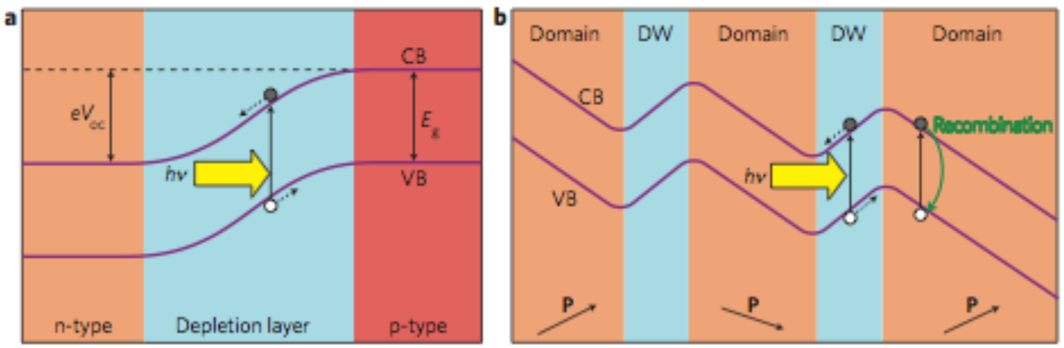


Qin et al. (2008)





T. Choi et al. (2009)



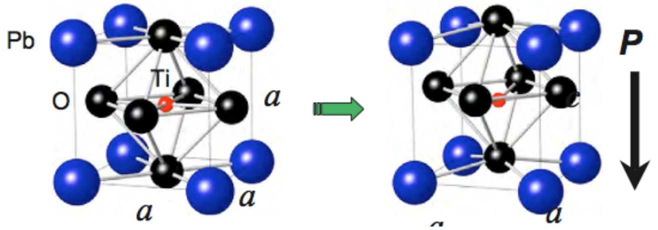
Yang et al. (2010)

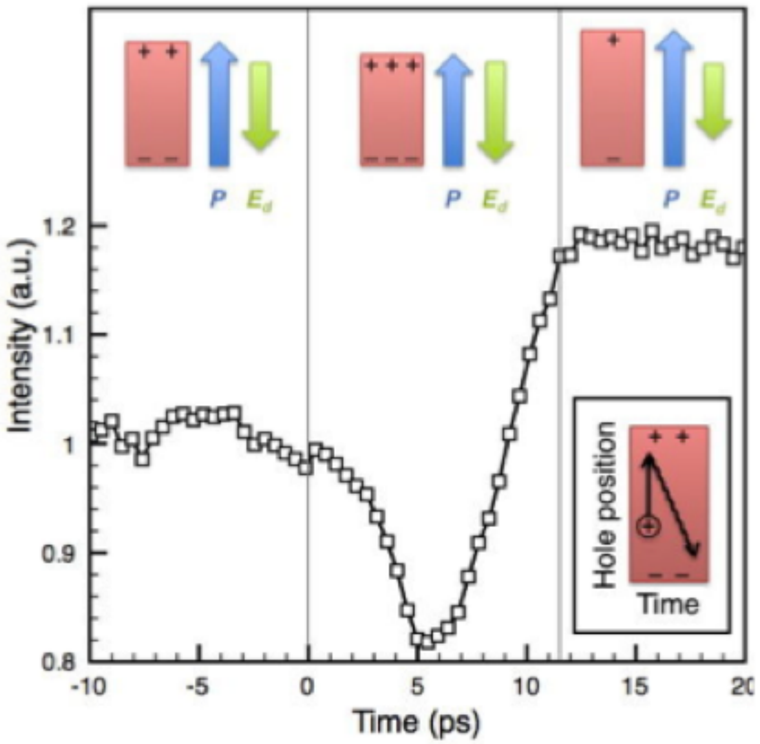
Development of electrode-less probes of intrinsic photovoltaic response…

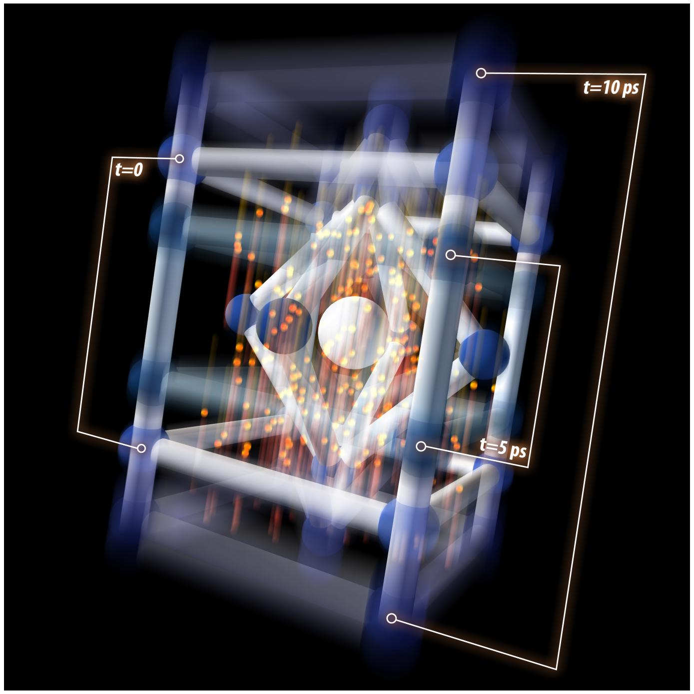
Ultrafast photovoltaic response

Time-dependent changes in the tetragonality driven by transient photocurrents

-2-step process: positive charges move first along the polarization direction, followed by displacement anti-parallel to the polarization, screening the depolarizing field.







Daranciang et al, Phys. Rev. Lett. (2012)

TFISH studies

A relevant 3rd order processes:

$$P_{2\omega} = \chi^{(3)} E_{\omega}^{2} E_{THz}$$

THz field modulates the efficiency of the second harmonic with efficiency linear in the incident THz field (TFISH).

$$I_{2\omega} = \left| \chi^{(3)} E_{\omega}^{2} E_{THz} + \chi^{(2)} E_{\omega}^{2} \right|$$

Cross term is linear in the THz field:

$$I_{2\omega} \propto \chi^{(2)} \chi^{(3)} I_{\omega}^{2} E_{THz}$$

APPLIED PHYSICS LETTERS **92**, 011131 (2008)

Coherent heterodyne time-domain spectrometry covering the entire "terahertz gap"

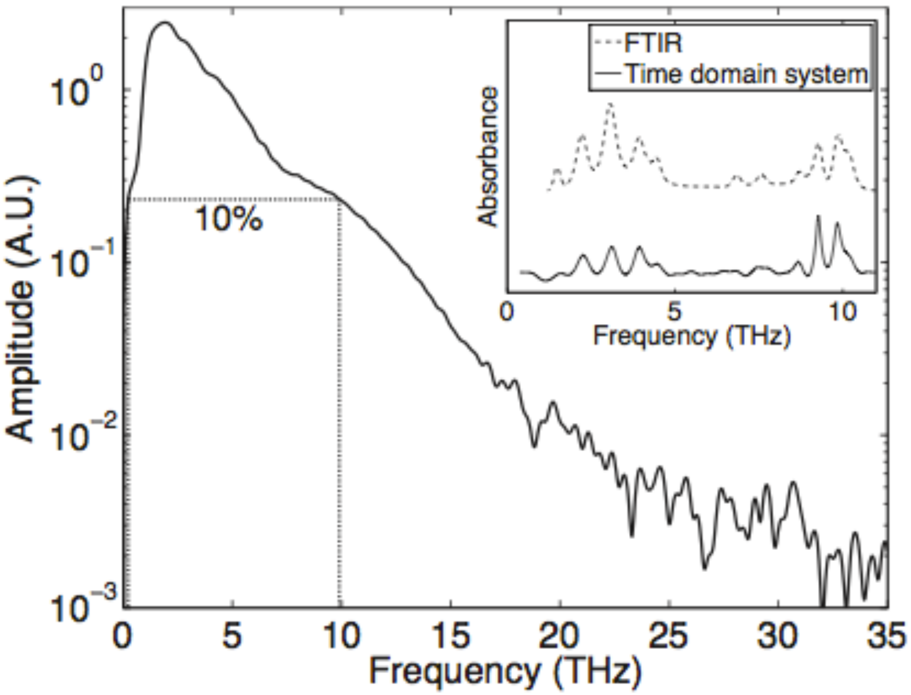
Nicholas Karpowicz, Jianming Dai, Xiaofei Lu, Yunqing Chen, Masashi Yamaguchi,
Hongwei Zhao, and X.-C. Zhang[a]
Center for Terahertz Research, Rensselaer Polytechnic Institute, Troy, New York 12180, USA

Liangliang Zhang and Cunlin Zhang
Department of Physics, Capital Normal University, Beijing 100037, China

Matthew Price-Gallagher and Clark Fletcher
HydroElectron Ventures, Inc., 1303 Greene Avenue Suite 304, Westmount, Quebec H3Z 2A, Canada

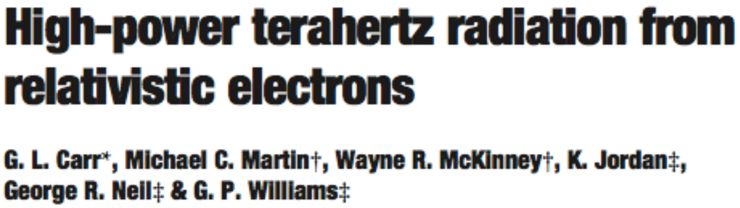
Orval Mamer and Alain Lesimple
Mass. Spec. Unit, 740 Dr. Penfield, Suite 5300, McGill University, Montreal, Quebec H3A 1A4, Canada

Keith Johnson
Massachusetts Institute of Technology, P.O. Box 380792, Cambridge, Massachusetts 02238-0792, USA



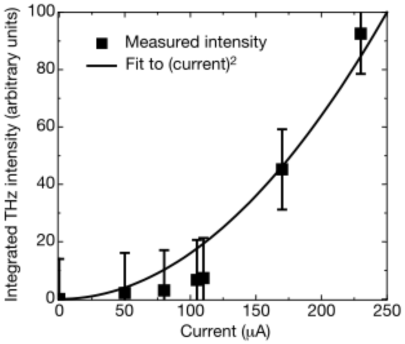
Karpowicz et al. APL (2008)

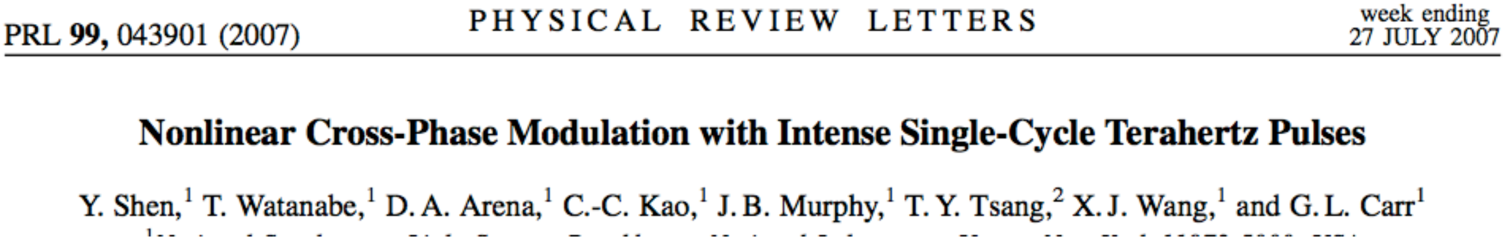
Short pulse THz sources based on coherent synchrotron radiation



High-power terahertz radiation from relativistic electrons

G. L. Carr*, Michael C. Martin†, Wayne R. McKinney†, K. Jordan‡, George R. Neil‡ & G. P. Williams‡

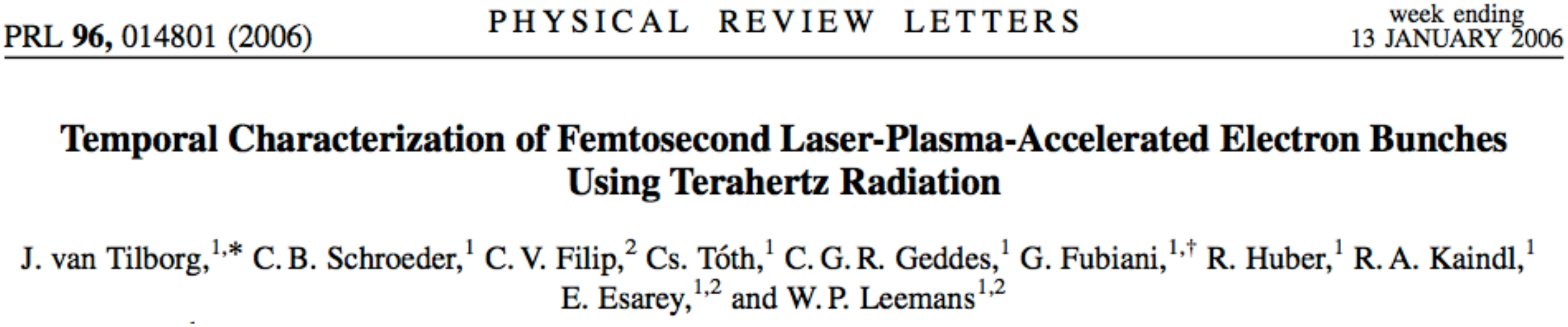




PRL **99,** 043901 (2007) PHYSICAL REVIEW LETTERS week ending 27 JULY 2007

Nonlinear Cross-Phase Modulation with Intense Single-Cycle Terahertz Pulses

Y. Shen,[1] T. Watanabe,[1] D. A. Arena,[1] C.-C. Kao,[1] J. B. Murphy,[1] T. Y. Tsang,[2] X. J. Wang,[1] and G. L. Carr[1]

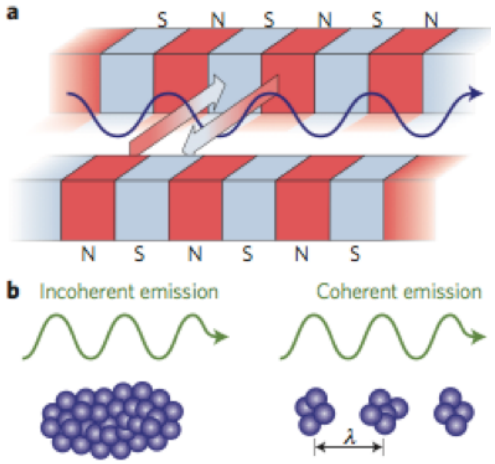


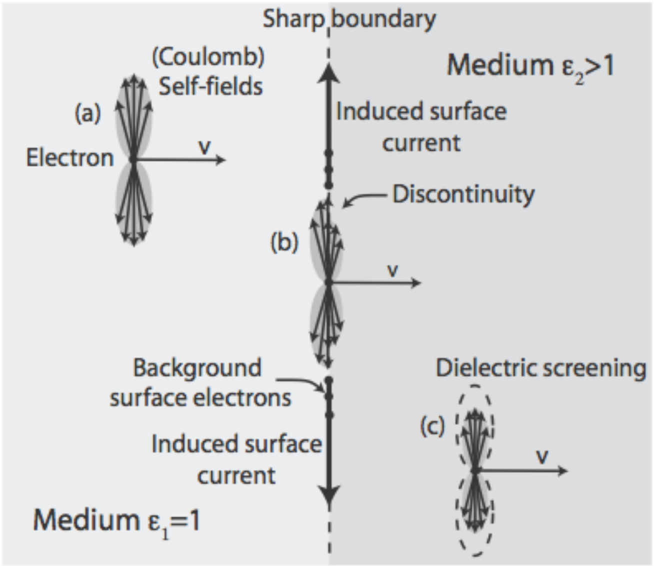
PRL **96,** 014801 (2006) PHYSICAL REVIEW LETTERS week ending 13 JANUARY 2006

Temporal Characterization of Femtosecond Laser-Plasma-Accelerated Electron Bunches Using Terahertz Radiation

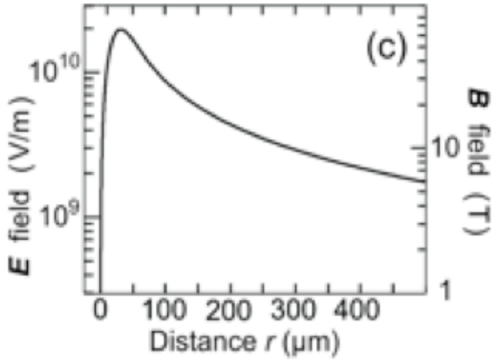
J. van Tilborg,[1,*] C. B. Schroeder,[1] C. V. Filip,[2] Cs. Tóth,[1] C. G. R. Geddes,[1] G. Fubiani,[1,†] R. Huber,[1] R. A. Kaindl,[1]
E. Esarey,[1,2] and W. P. Leemans[1,2]

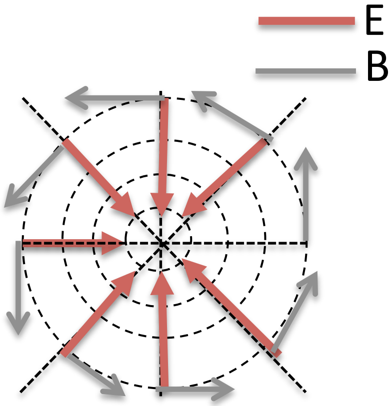
THz Coherent Transition Radiation from the LCLS

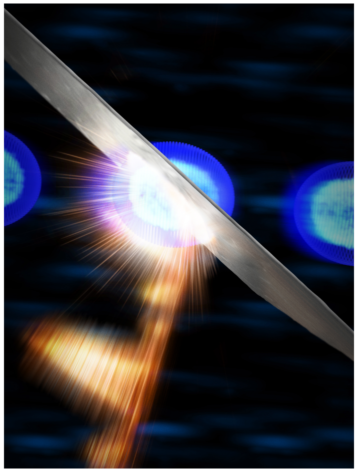




N^2 scaling for wavelengths > electron bunch length



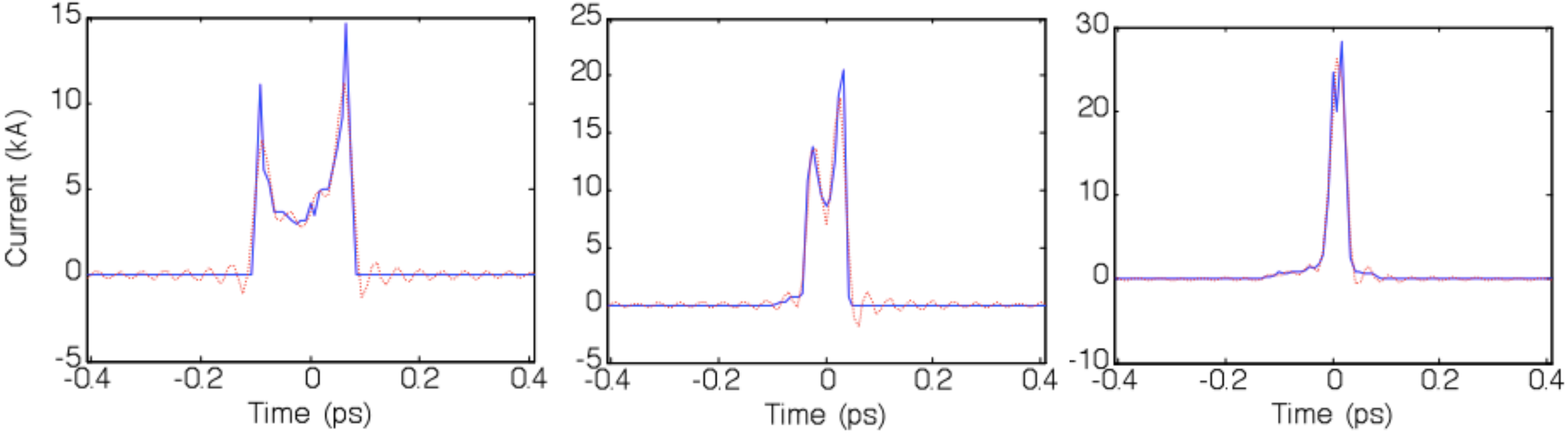




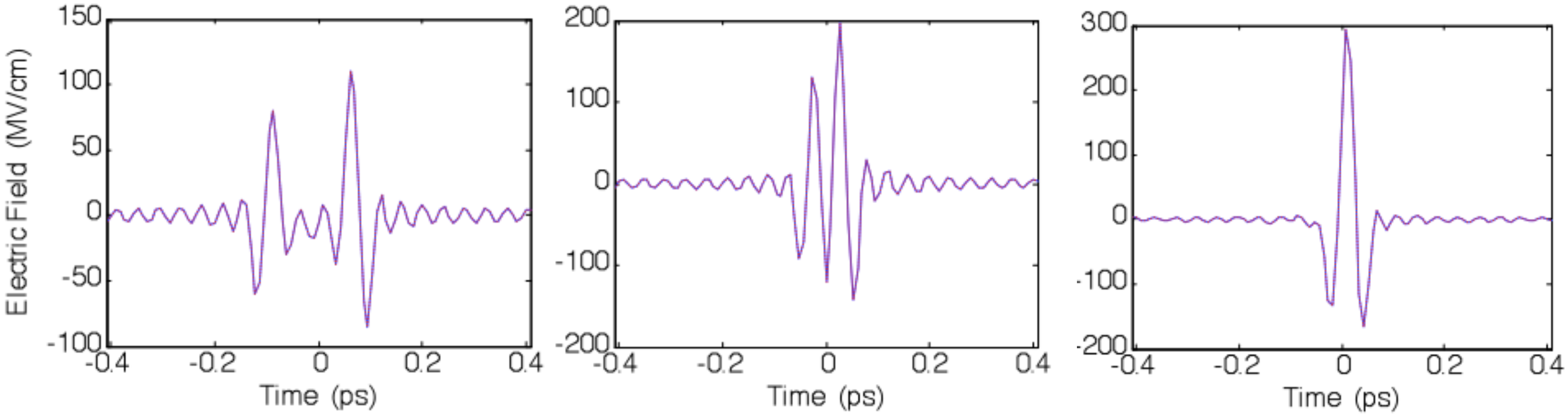
Daranciang et al., Appl. Phys. Lett. (2011)

Simulations

Time-dependent current

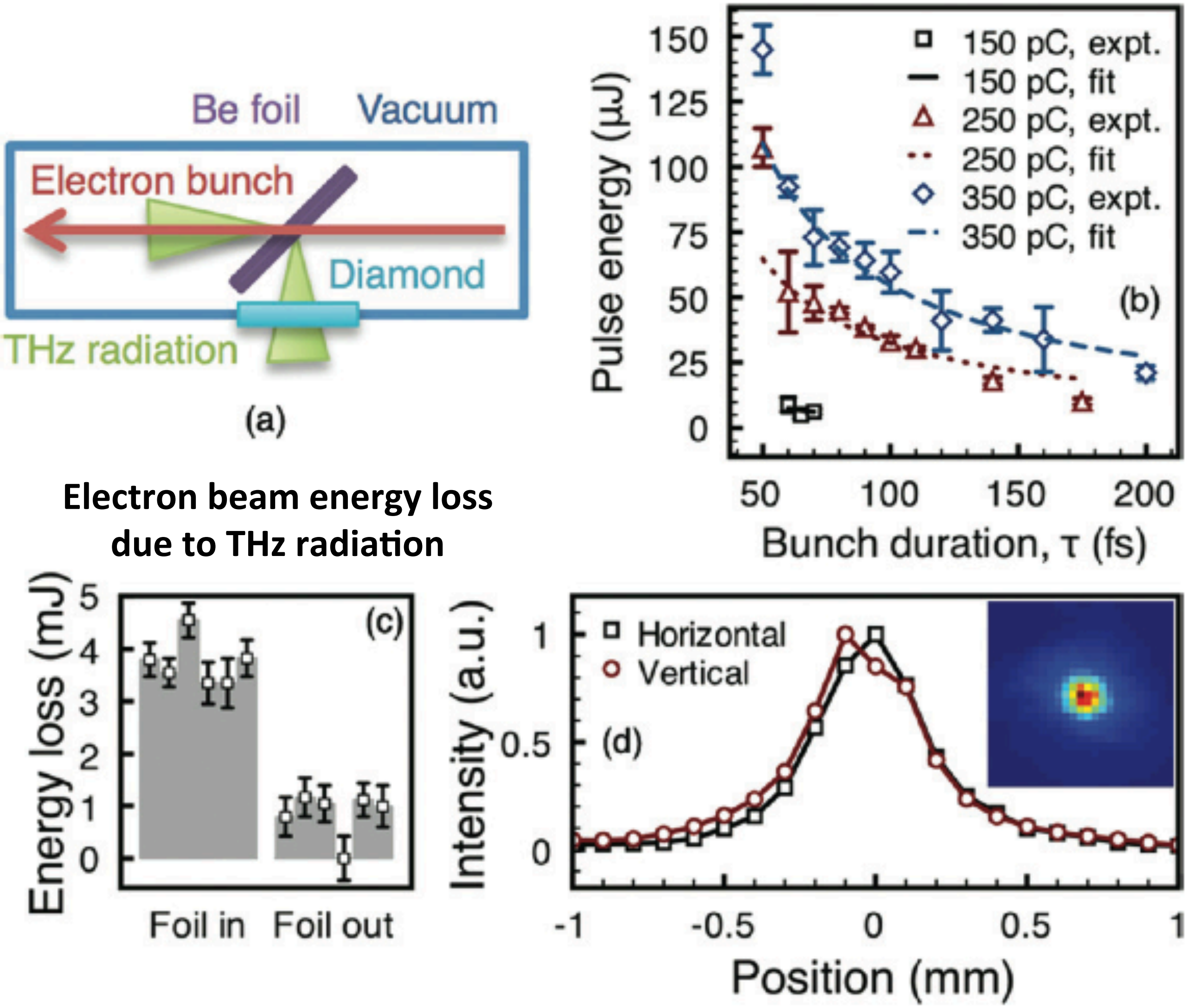


THz electric field



Loos et al.

Generation of ultrashort, ultraintense single-cycle fields

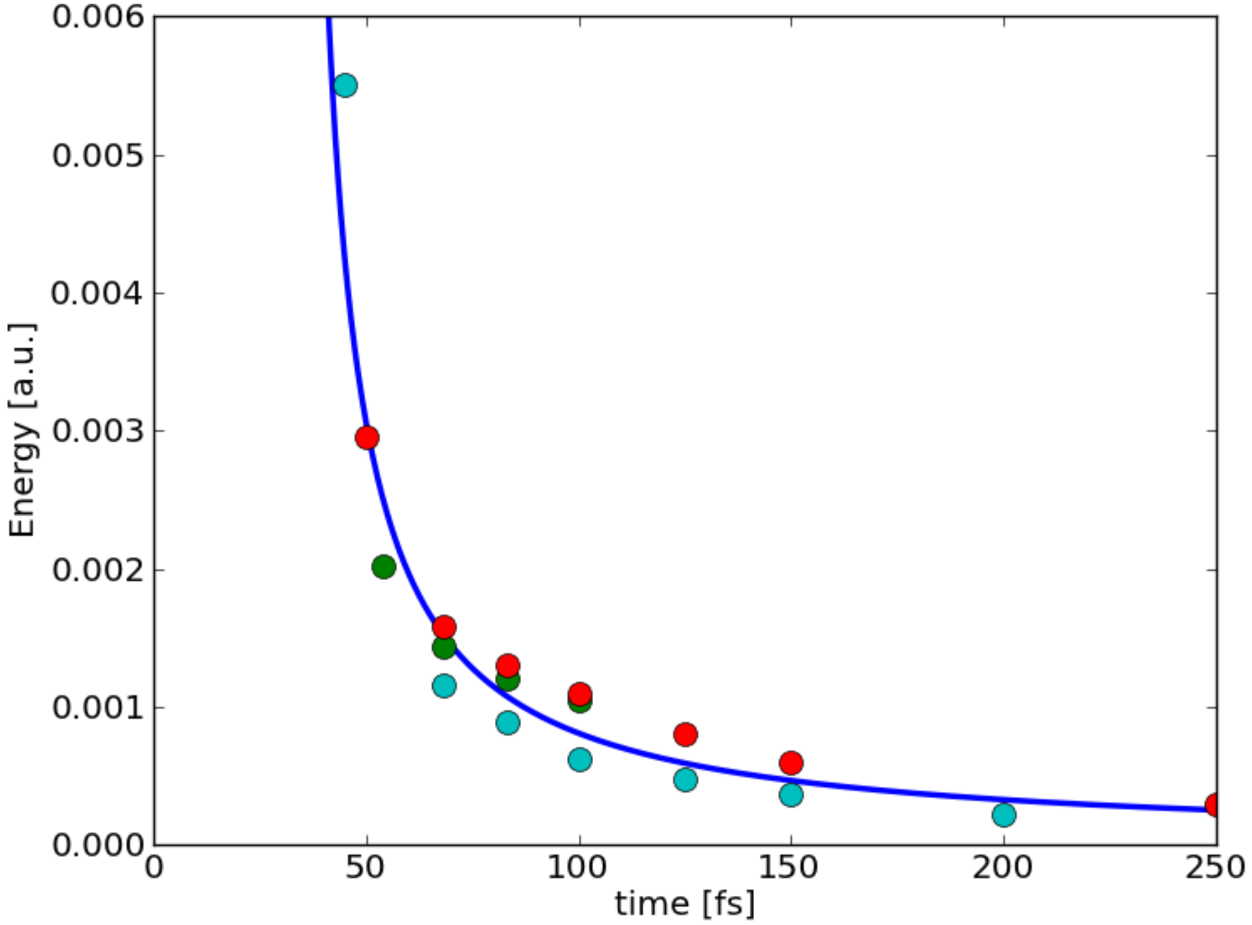


Electron beam energy loss due to THz radiation

$$U \sim \frac{Q^2}{\tau}$$

Daranciang et al.,
Appl. Phys. Lett. (2011)

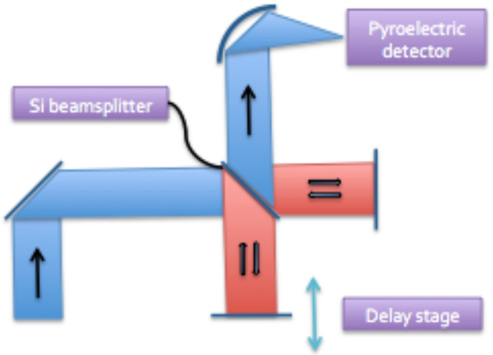
Scaling with charge and pulse duration

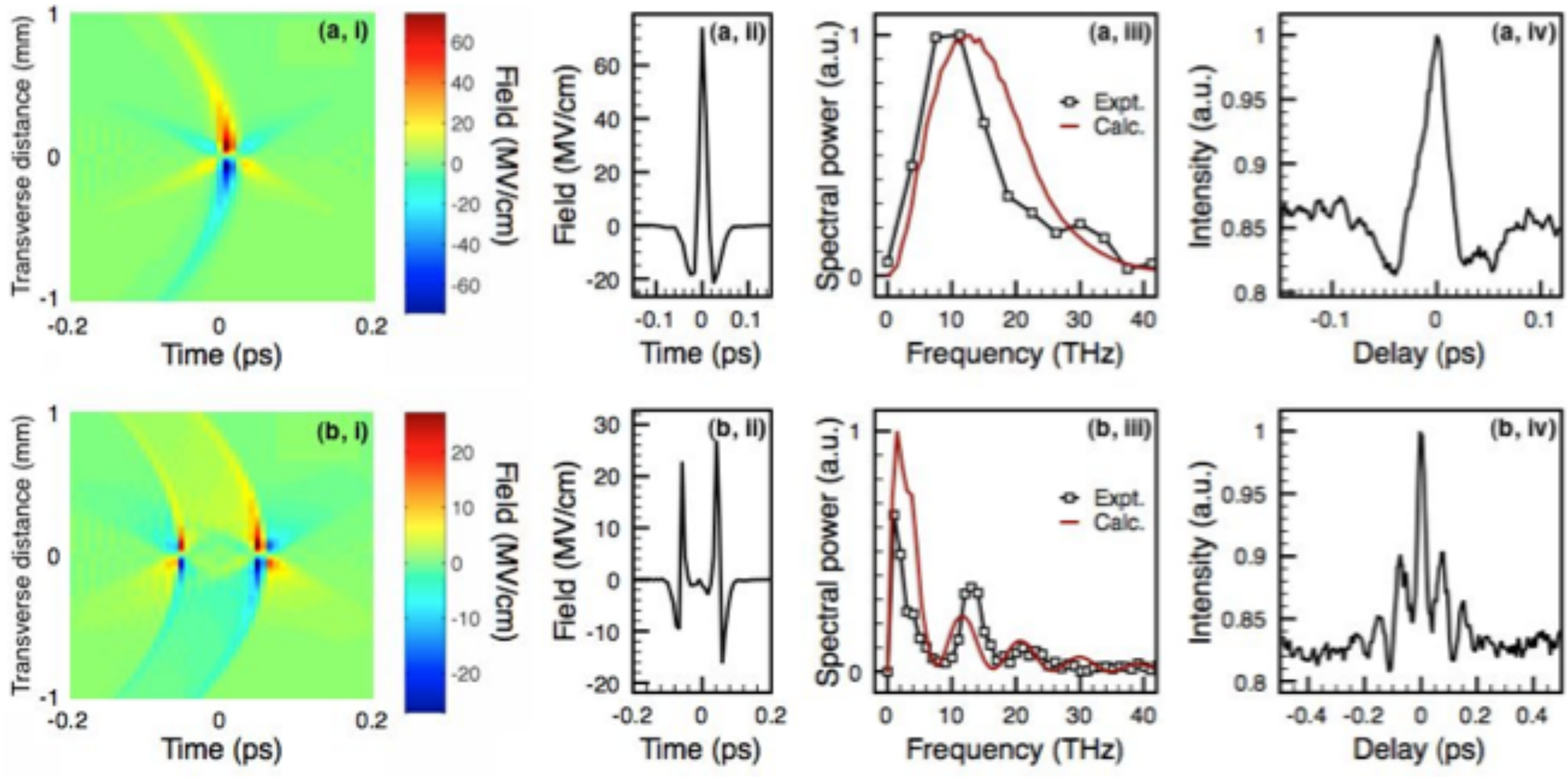


Peak energies of ~0.3 mJ

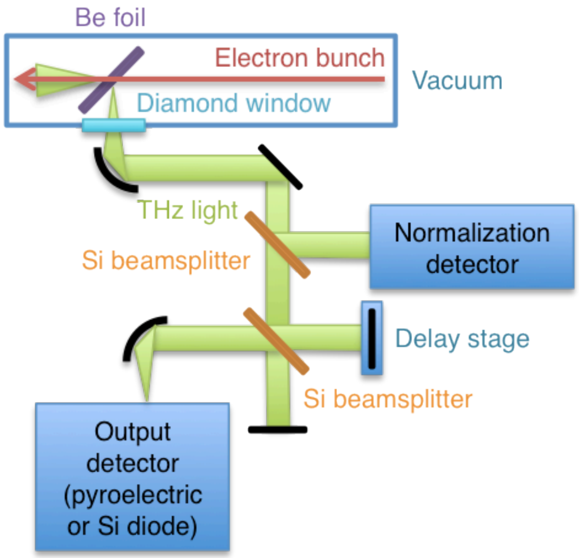
E-fields ~ 0.2 volt/angstrom; B fields ~ 10 T

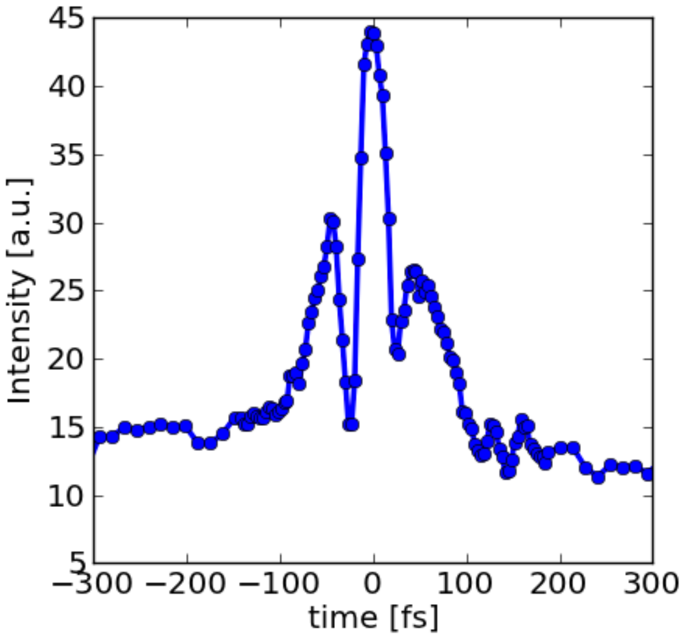
Field autocorrelations - Spectrum



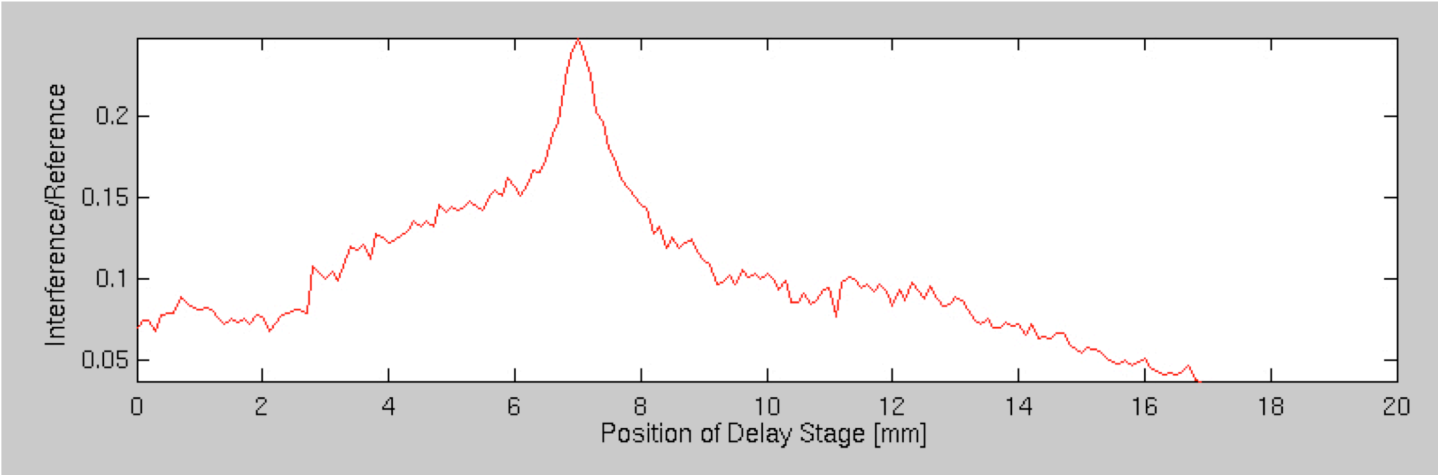


THz nonlinear response and nonlinear autocorrelations

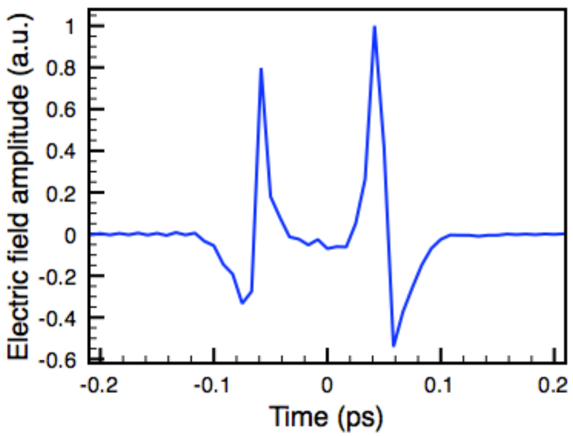

Be foil
Electron bunch
Vacuum
Diamond window
THz light
Si beamsplitter
Normalization detector
Delay stage
Si beamsplitter
Output detector (pyroelectric or Si diode)

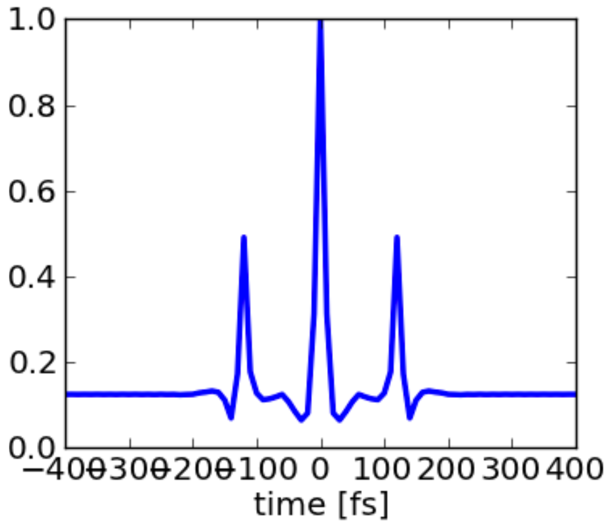


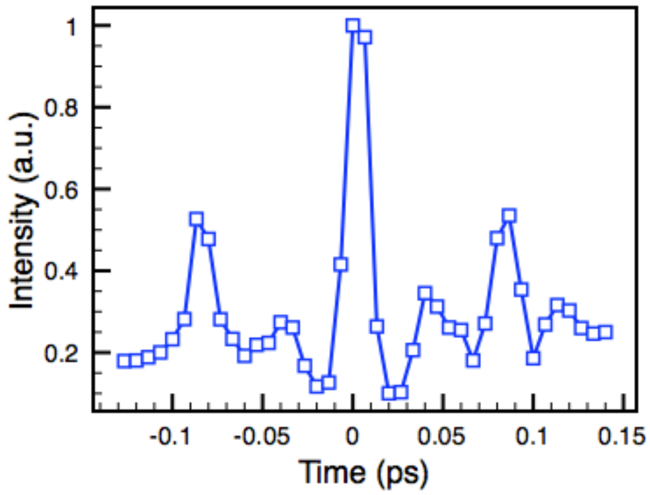
Z-scan



THz nonlinear autocorrelations







-Enables recovery of THz temporal pulse shape

-Effects indicative of high-order nonlinearity, non-perturbative response

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