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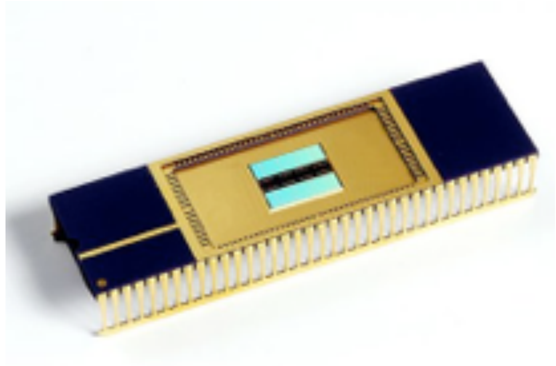
Potential avenues for THz-induced control of order parameters in solids

Steve Johnson

ETH Zurich

Solid state physics: why ultrafast?

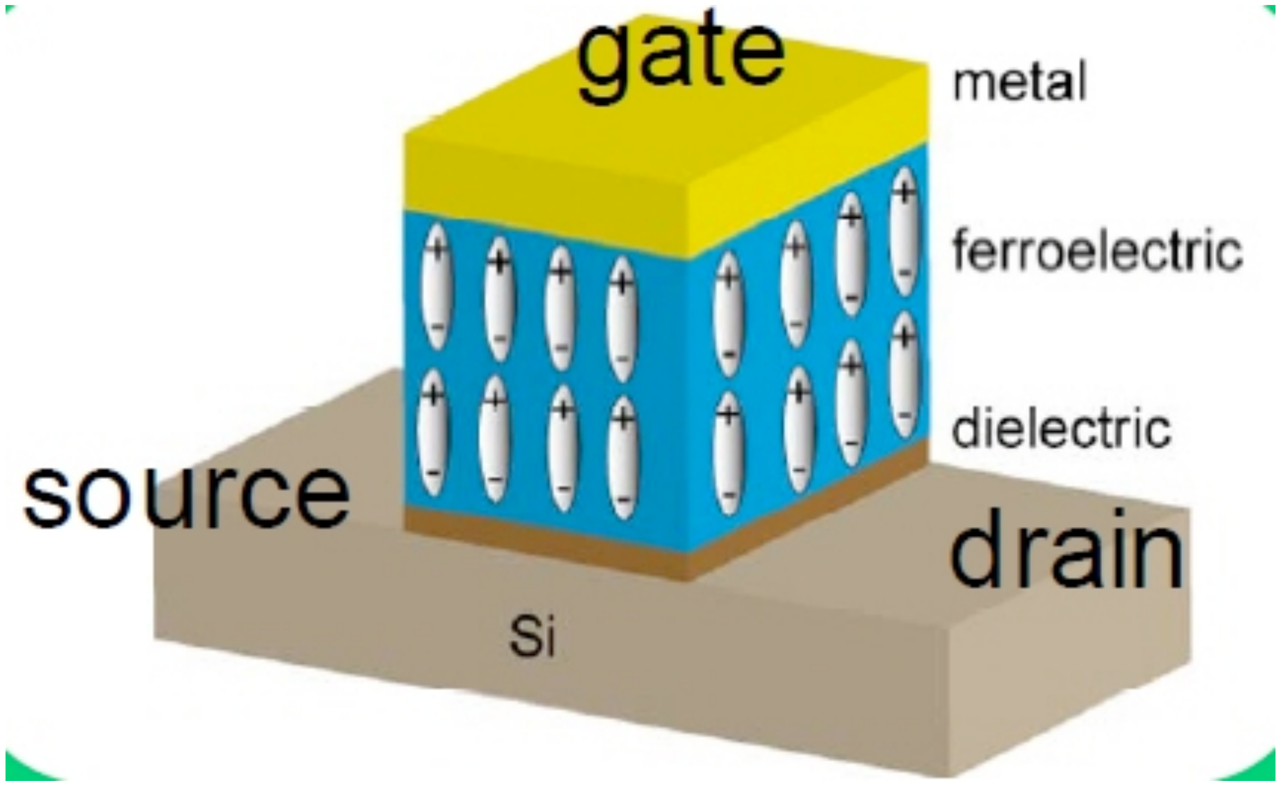


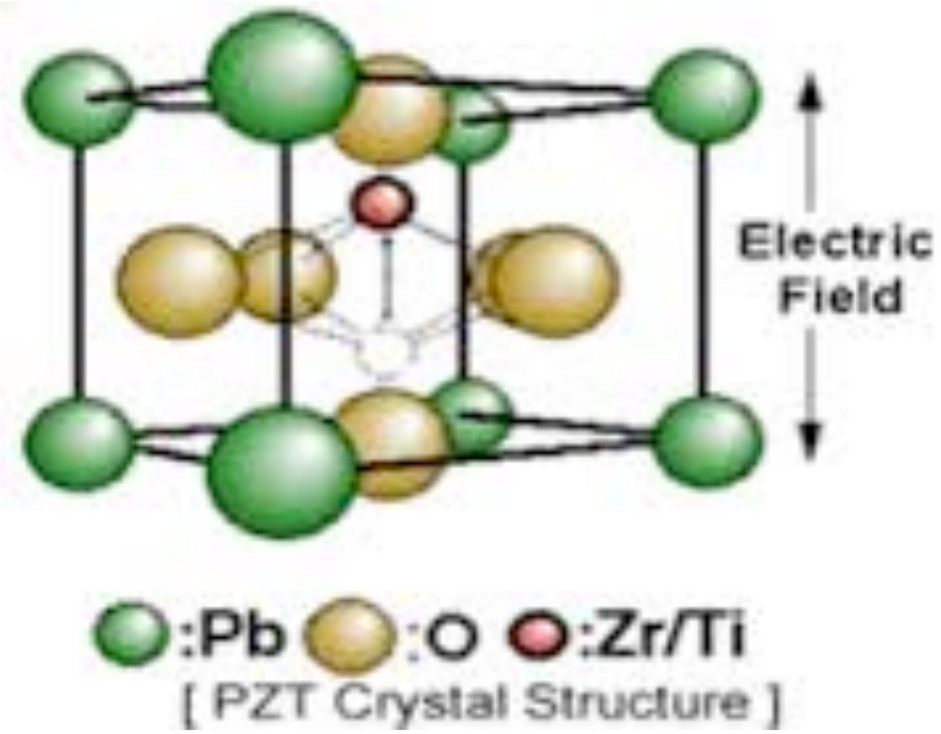
Phase-change RAM

Compact flash (CF) & secure digital (SD) cards, a Sony memory stick, and a USB memory key.

- Solid state memories: speed as key performance criterion
- Switching time scales ~ ns
- What are fundamental limits on a microscopic scale?

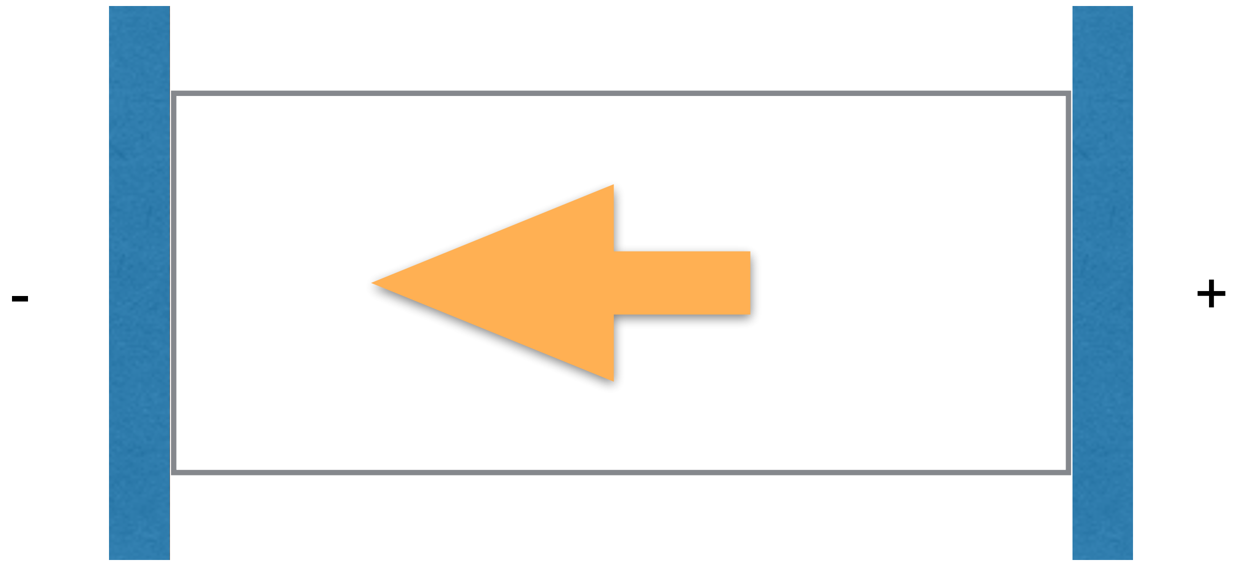
Ferroelectric memory





- Information encoded by electrical polarization, induced by structure change

Ferroelectric memory



- Switching via domain nucleation and growth under applied field, takes ~ 50 ns in real devices
- Contrast with vibrational periods of ~ 1 ps = 0.001 ns

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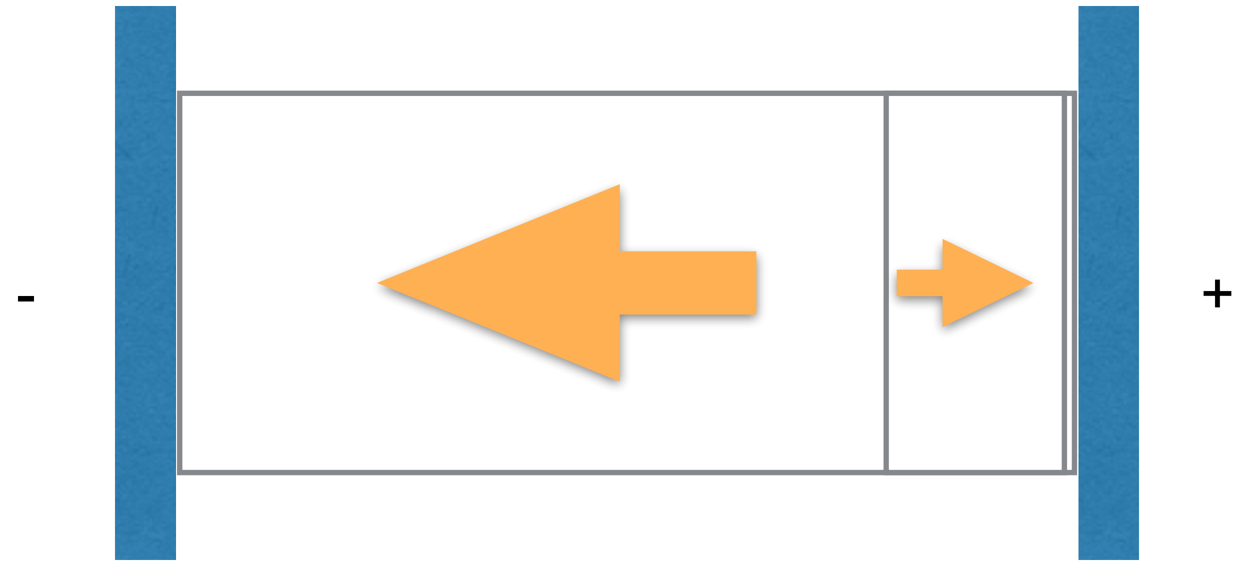
Ferroelectric memory



- Switching via domain nucleation and growth under applied field, takes ~ 50 ns in real devices

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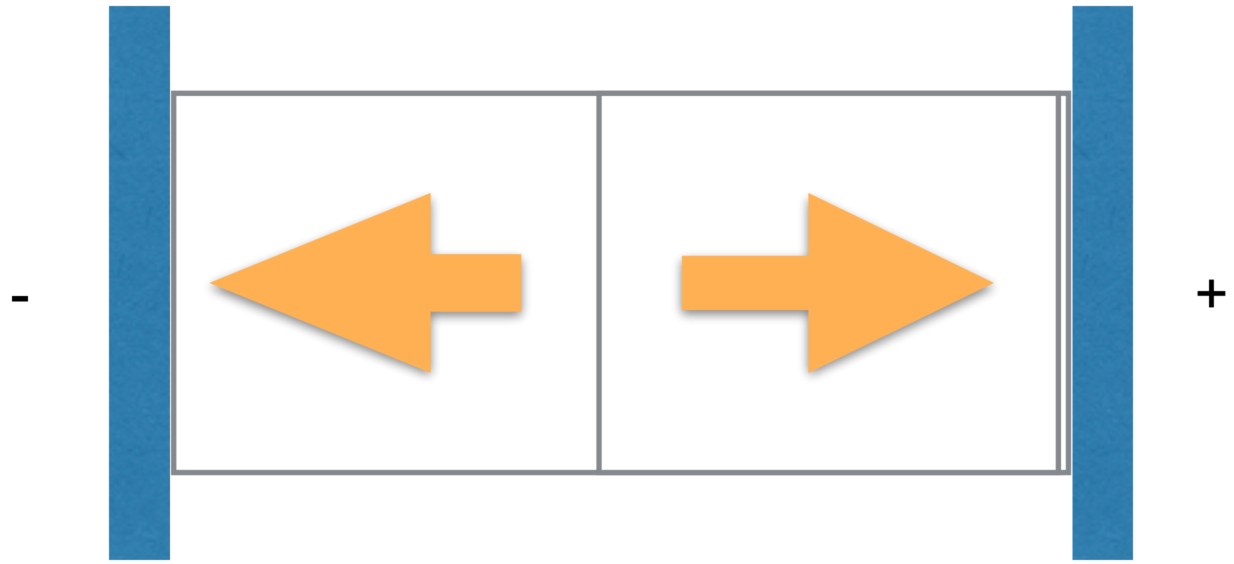
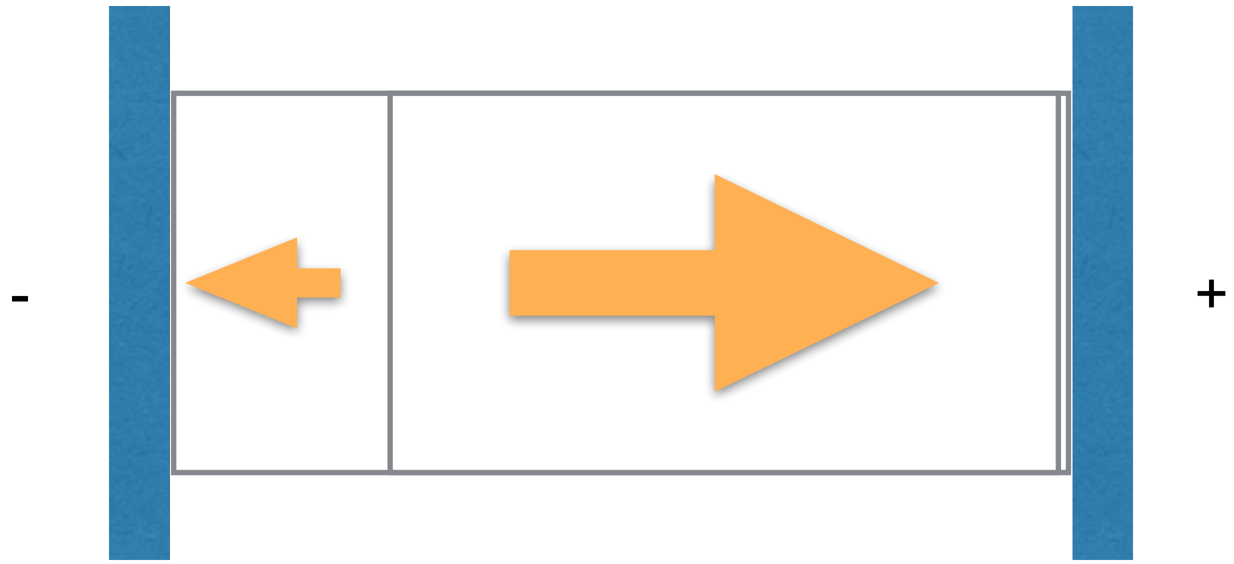
Ferroelectric memory



- Switching via domain nucleation and growth under applied field, takes ~ 50 ns in real devices

Ferroelectric memory



- Switching via domain nucleation and growth under applied field, takes ~ 50 ns in real devices

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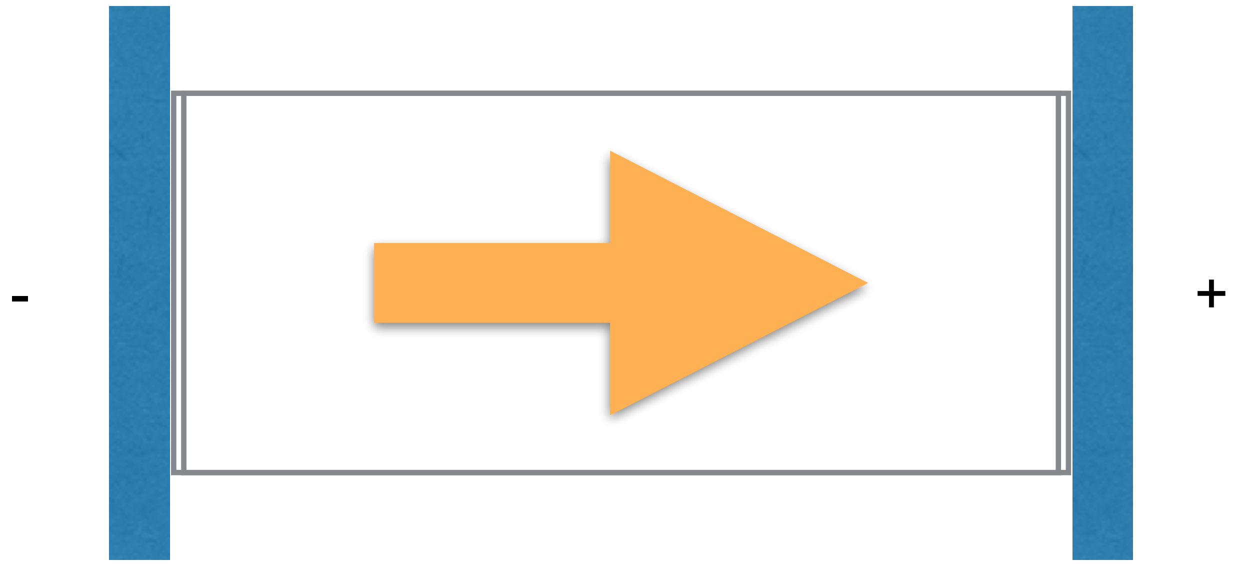
Ferroelectric memory



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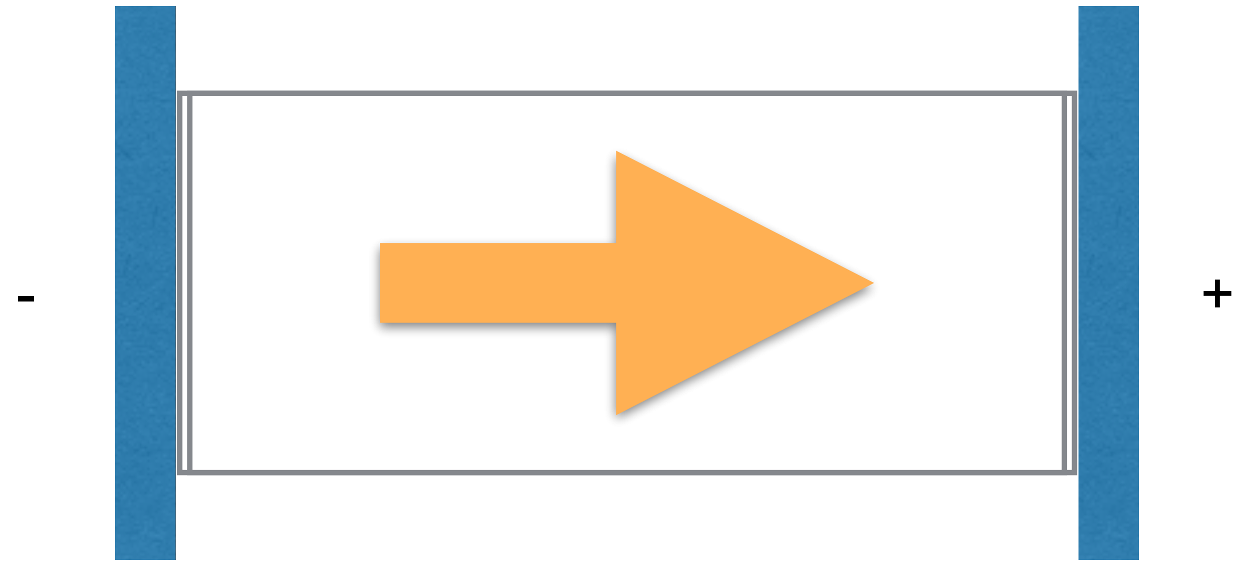
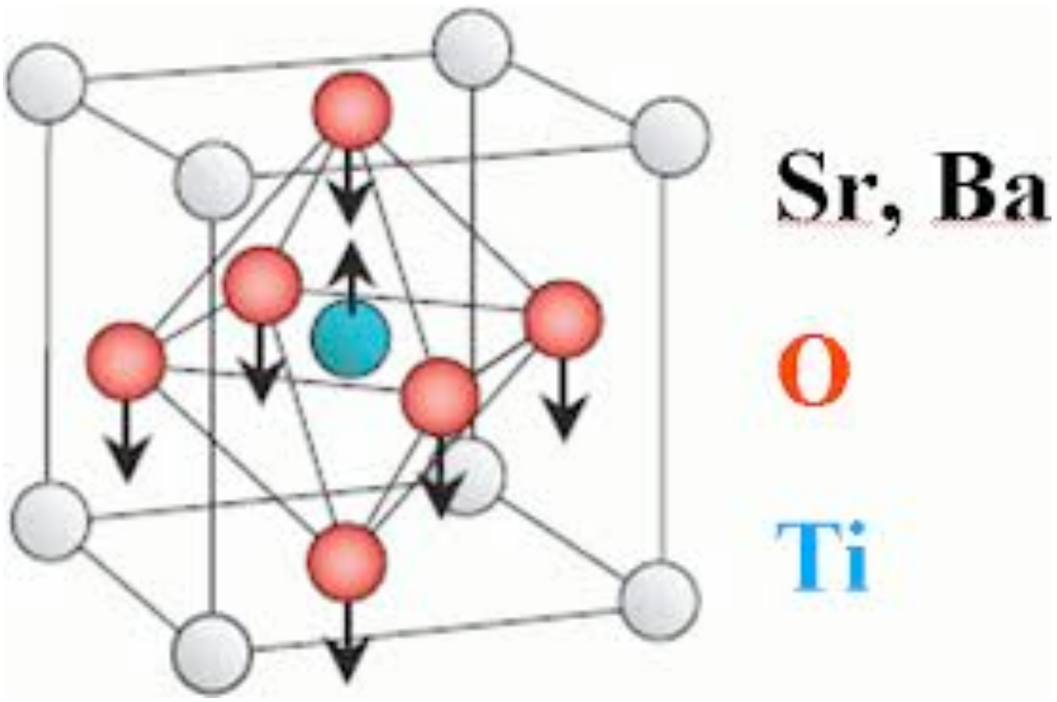
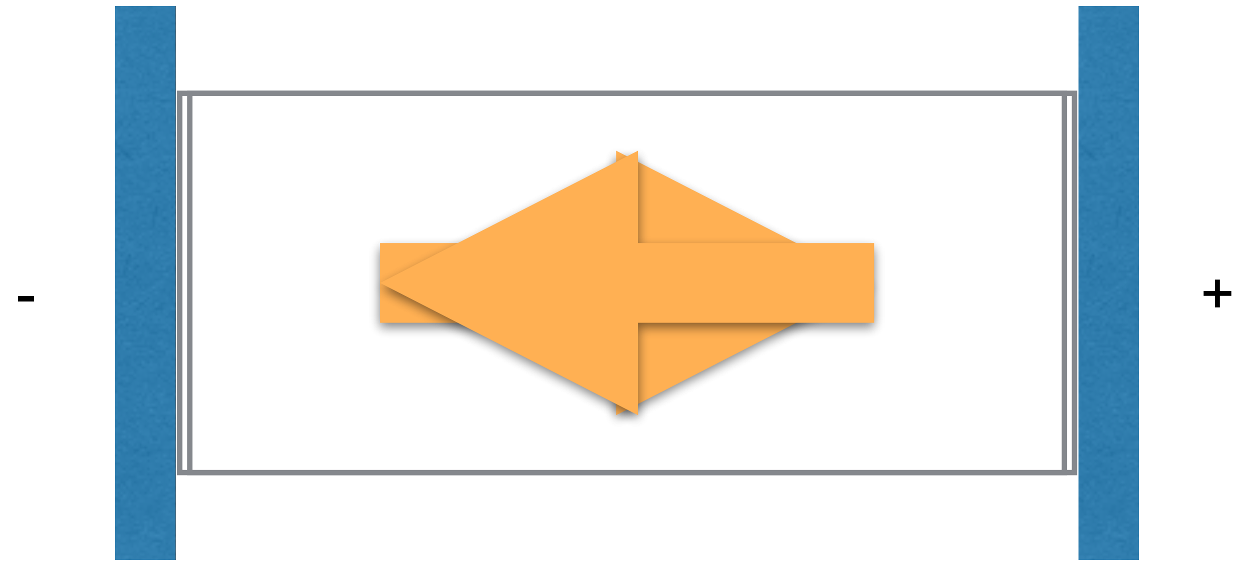
Ferroelectric memory

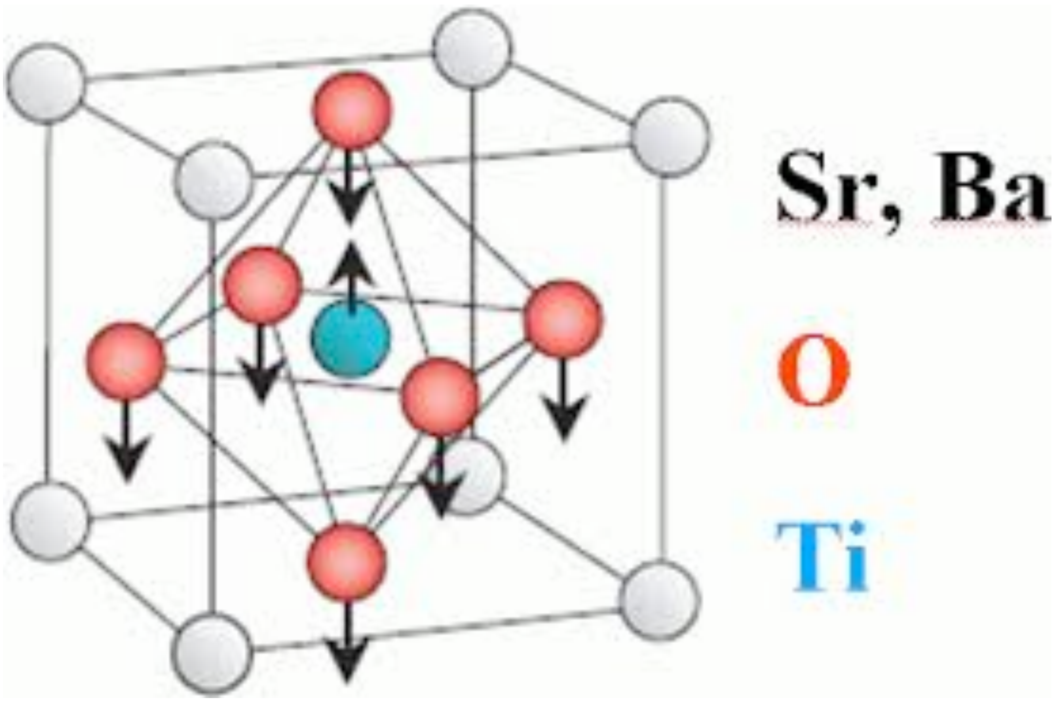




- Switching via domain nucleation and growth under applied field, takes ~ 50 ns in real devices
- Contrast with vibrational periods of ~ 1 ps = 0.001 ns

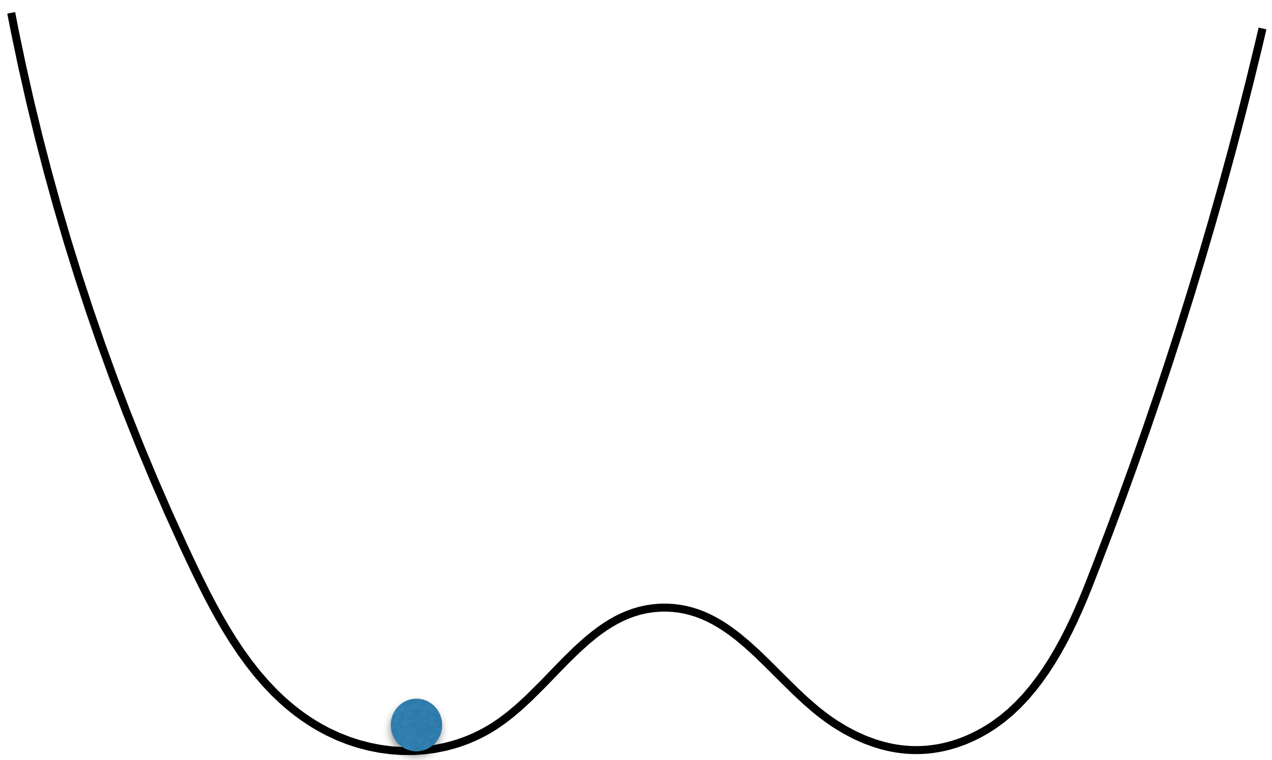
Ferroelectric memory





- Alternative path: can we homogeneously switch the entire domain at once?

Direct control over ferroic OP



- Quasi-resonant driving of an IR active vibration in a double-well potential
- Requires phase-controlled driving field, also damping
- This is “direct” control

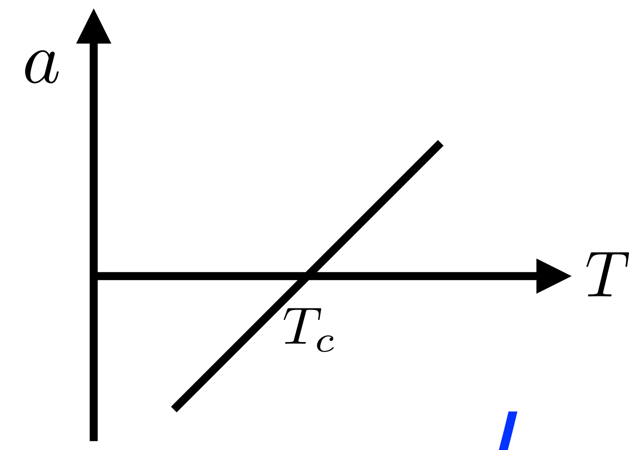
“Direct” control example 1:

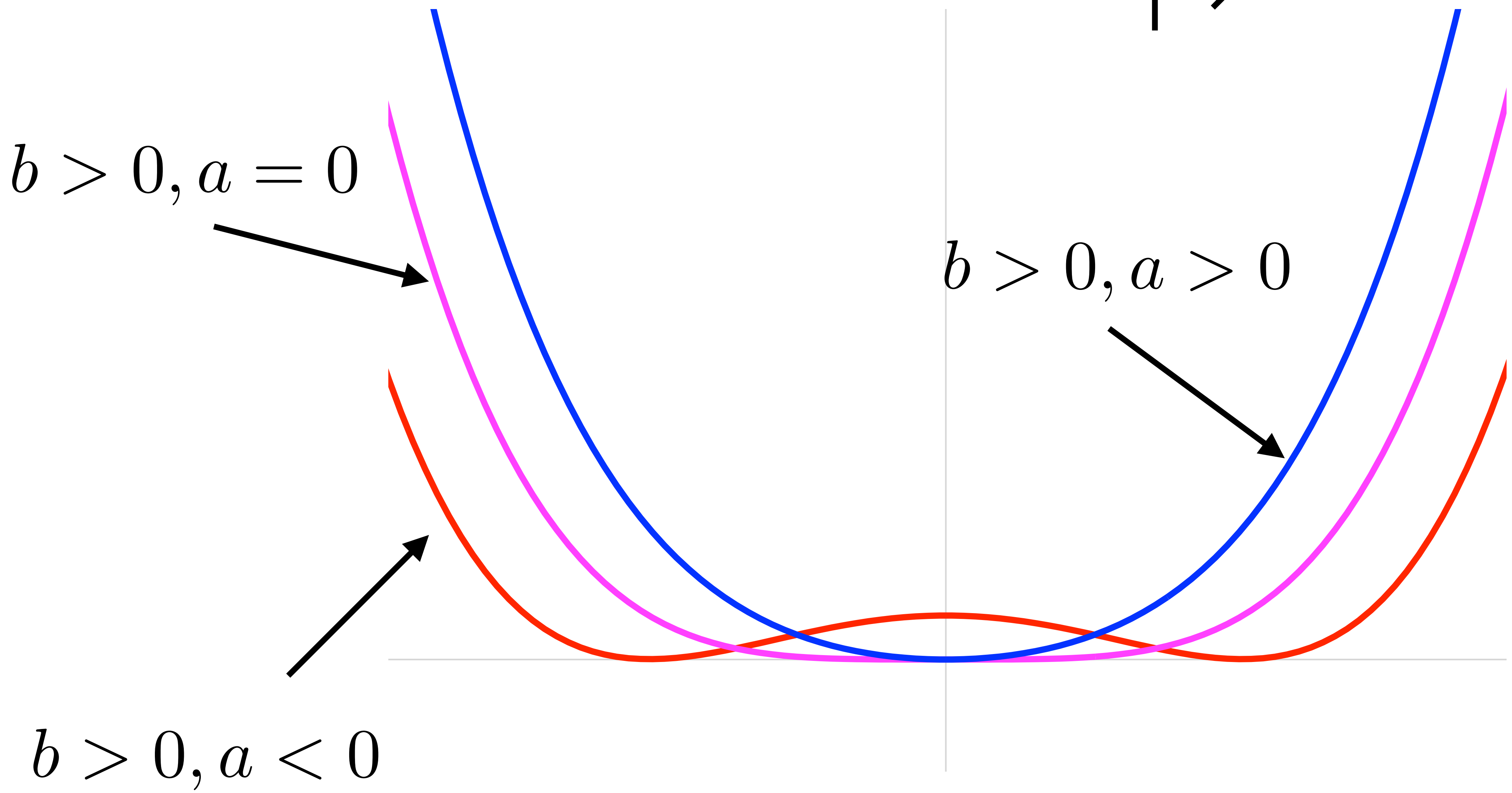
Ferroelectric polarizaiton switching

Soft mode ferroelectric

$$F(x) = ax^2 + bx^4$$

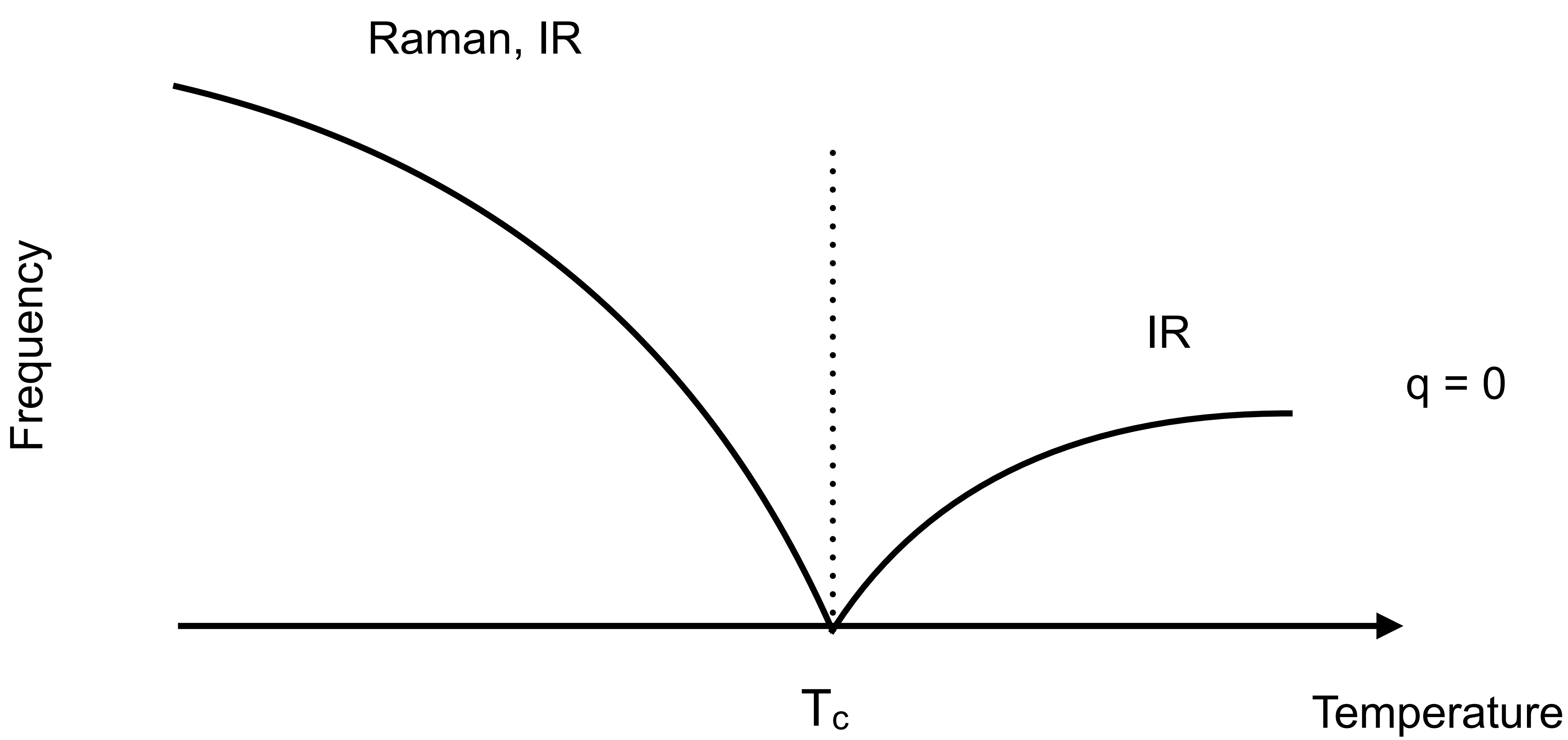
$$P \propto x$$



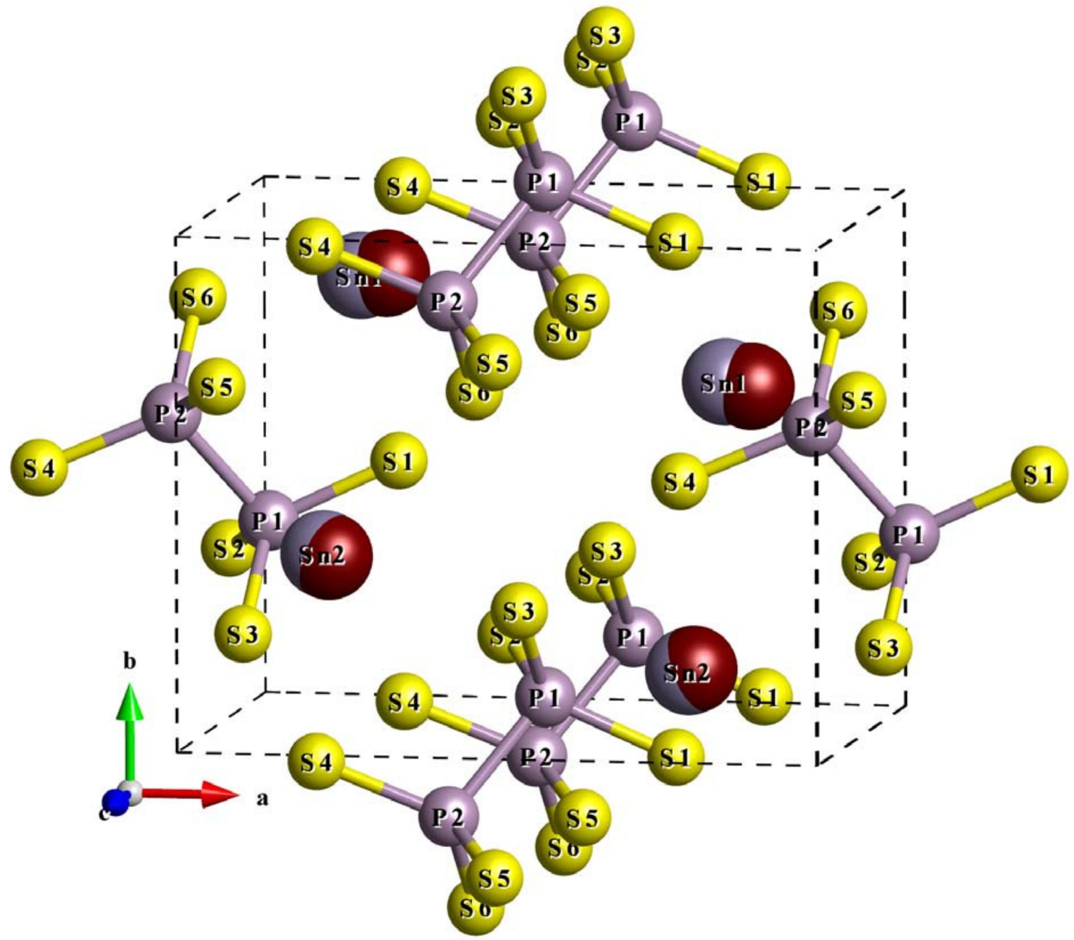


For a < 0, how fast can we change domains?

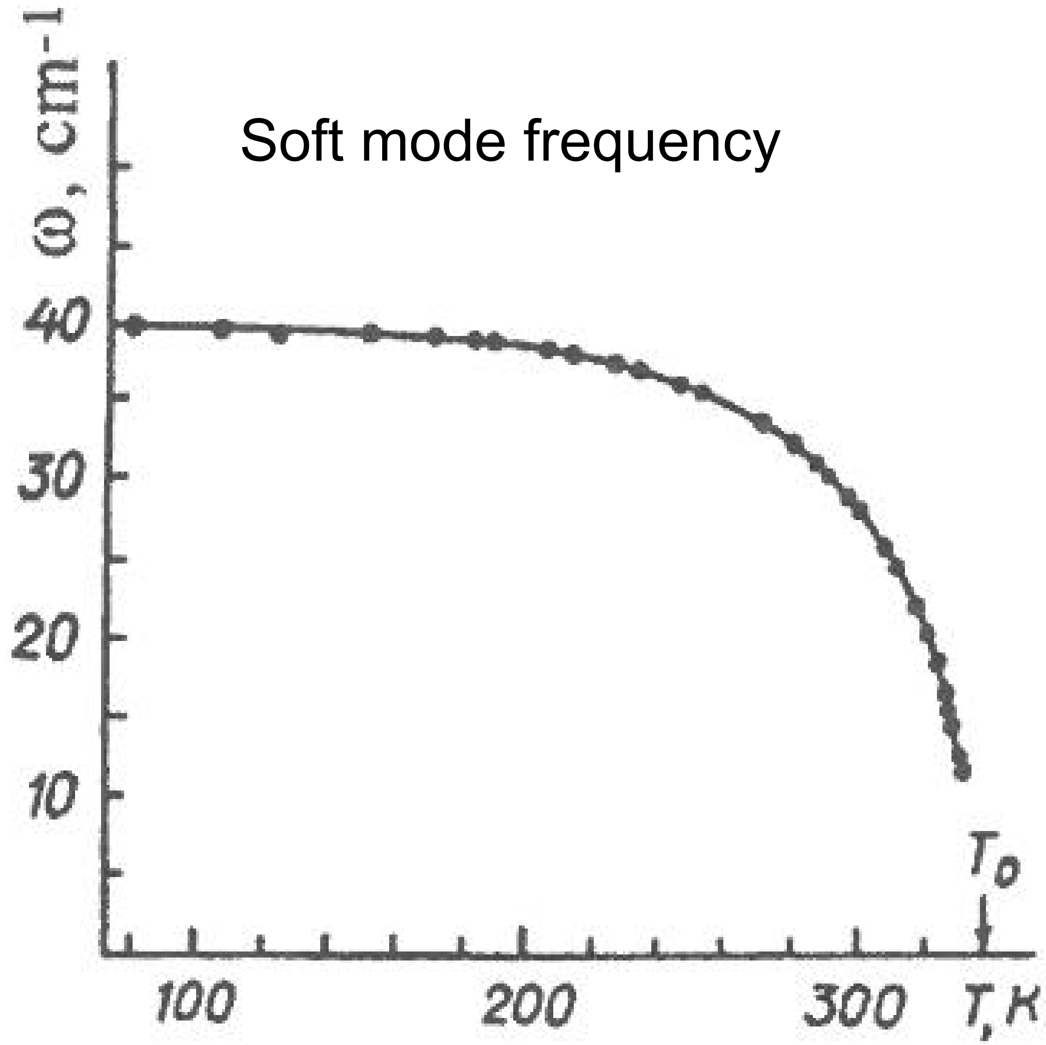
Soft mode ferroelectric



$Sn_2P_2S_6$: soft mode ferroelectric

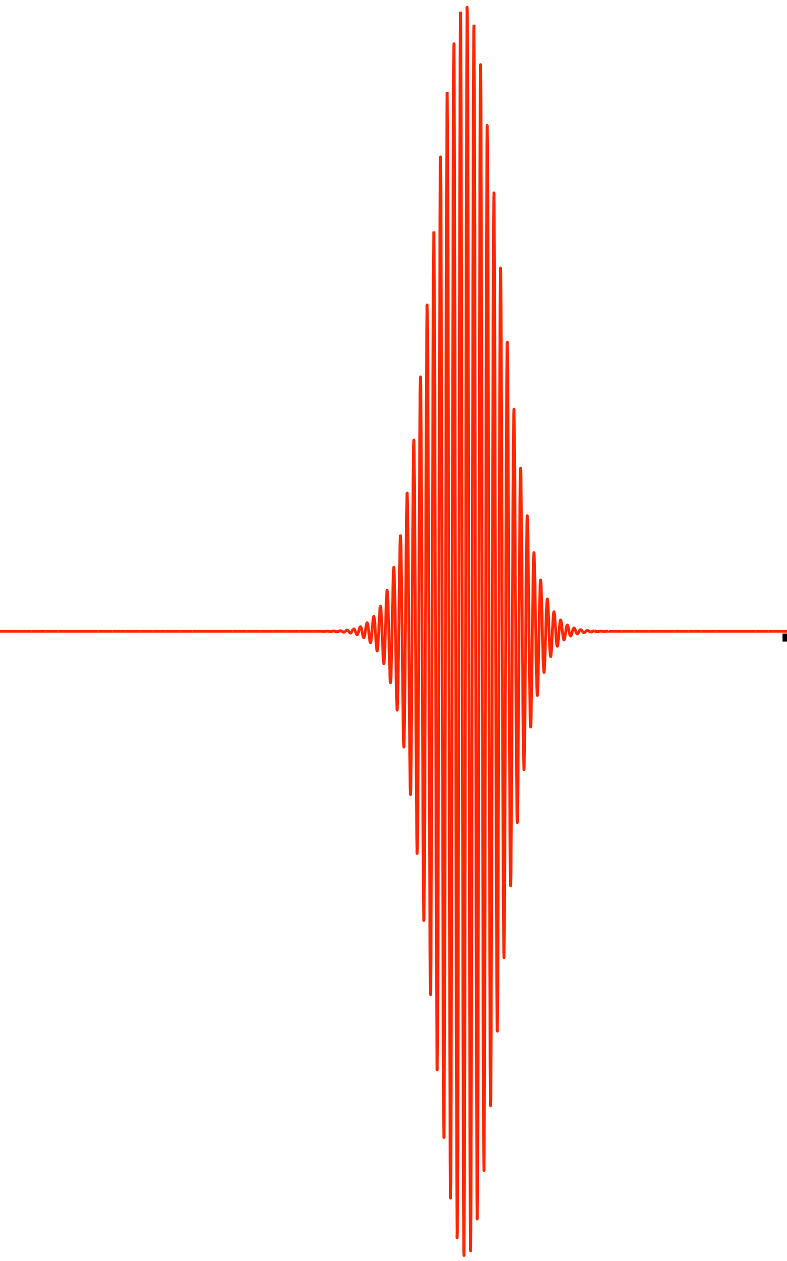


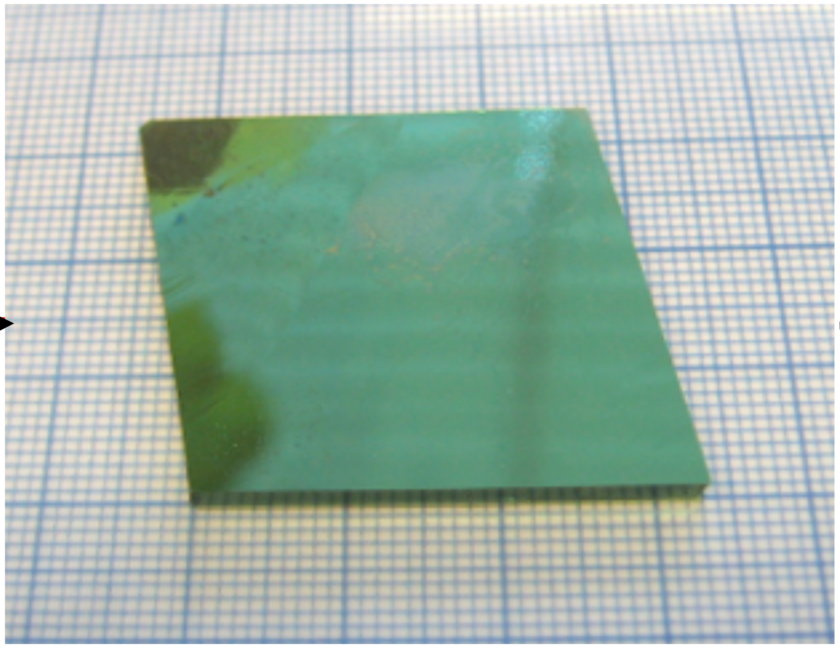
[Glukhov et al. Int. J. Mol. Sci. 2012, 13, 14356]



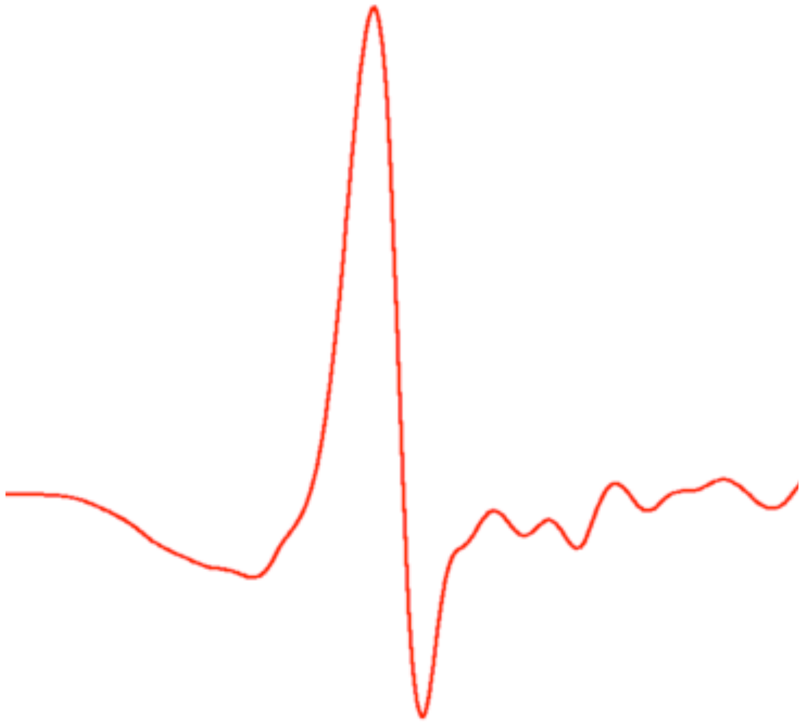
[Vysochanskii et al. Fiz. Tv. Tela 20, 90 (1978)]

THz generation

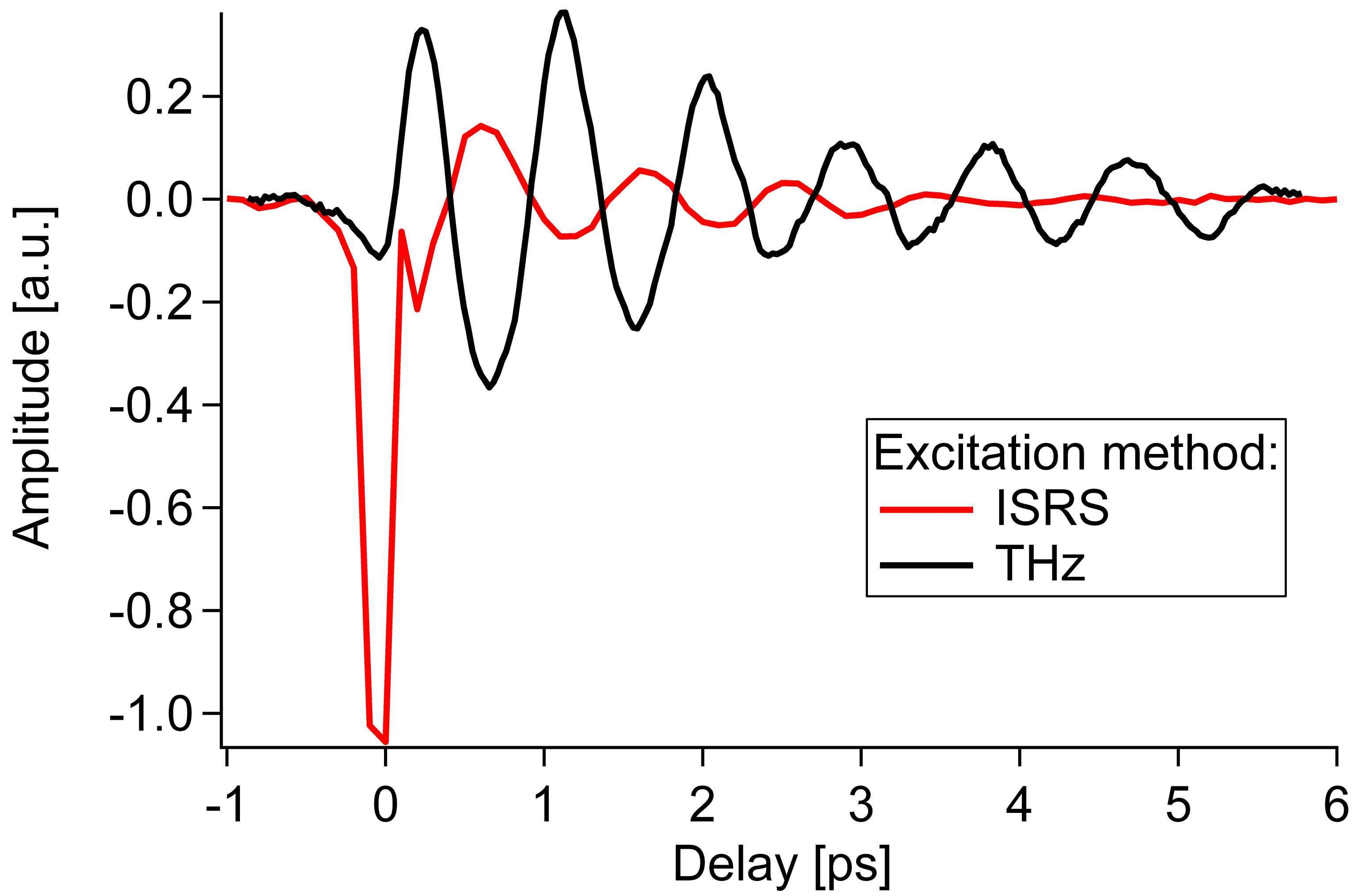




OH1



$Sn_2P_2S_6$: soft mode ferroelectric



- Optical detection of mode
  - ISRS: limited by damage to sample
  - THz (resonant): no damage, limited only by source (~150 kV/cm at present, optical rectification in OH1)

$Sn_2P_2S_6$: x-ray diffraction

ETHZ:

A. T. Huber
A. Ferrer
T. Kubacka
L. Huber
C. Dornes
J. Saari

EPFL/ETHZ:

A. Luebke
J. Rittmann

Uzhgorod:

Y. Vysochanskii

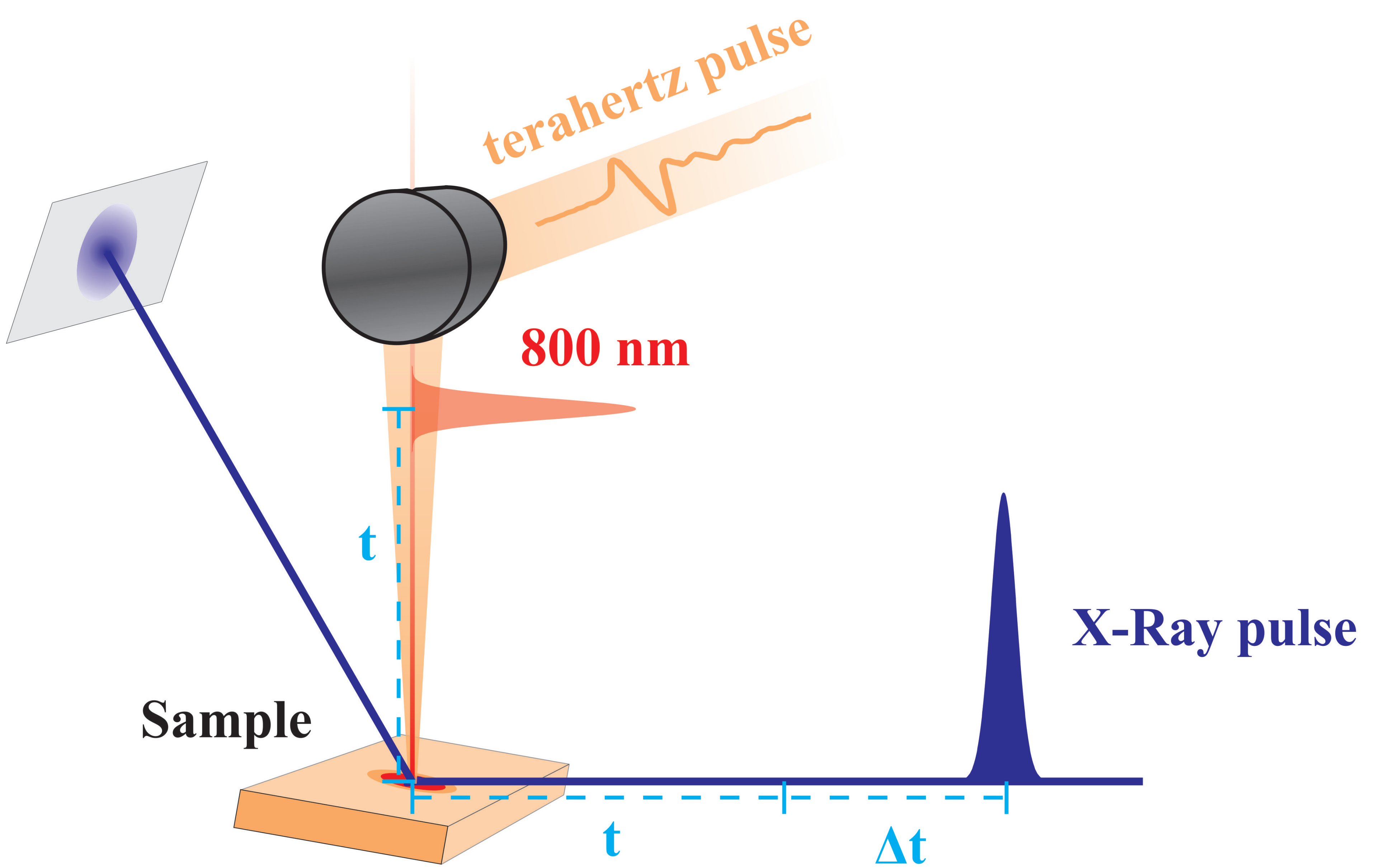
PAUL SCHERRER INSTITUT

S. Gruebel
J. Johnson
G. Ingold
S. Mariager
P. Beaud

SWISS NATIONAL SCIENCE FOUNDATION

- Electron beam slicing at Swiss Light Source
- ~ 120 fs duration x-ray pulses

$Sn_2P_2S_6$: x-ray diffraction



- 9° grazing x-rays, normal incidence THz
- Requires < 10 µm x-ray focus

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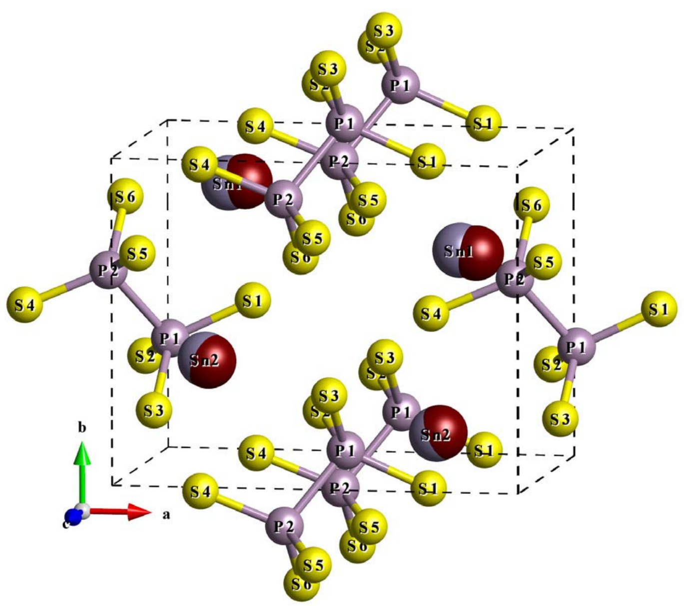
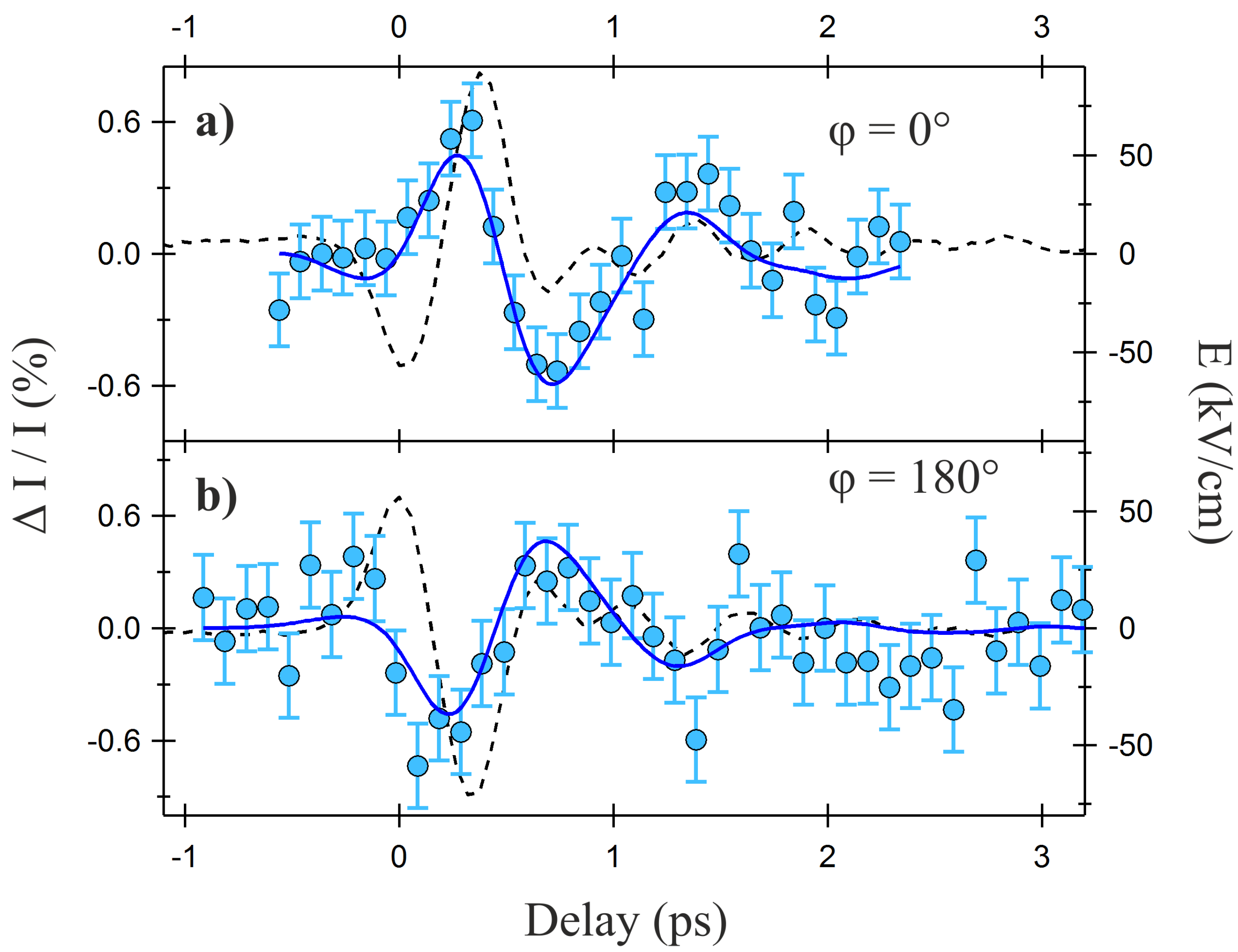
$Sn_2P_2S_6$: x-ray diffraction



(332) peak



- See ~2 pm Sn motion (7.5% of full transition)

[S. Gruebel, arXiv:1602.05435]

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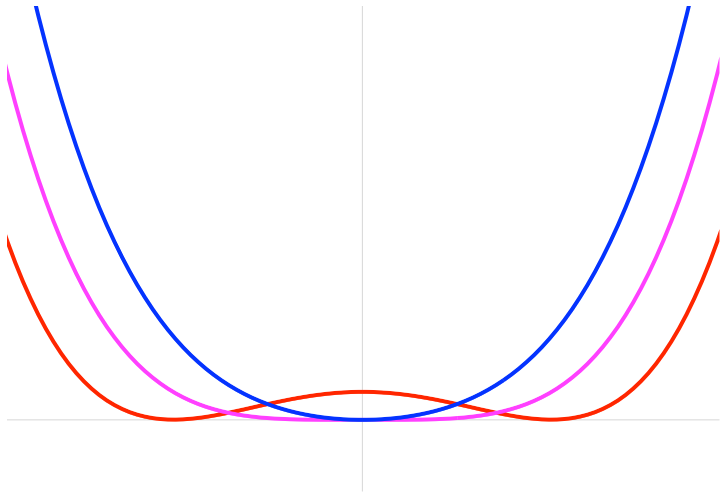
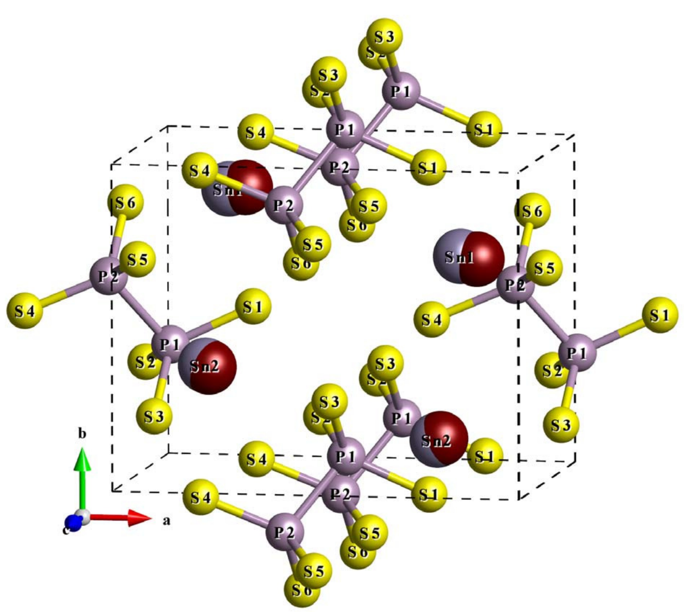
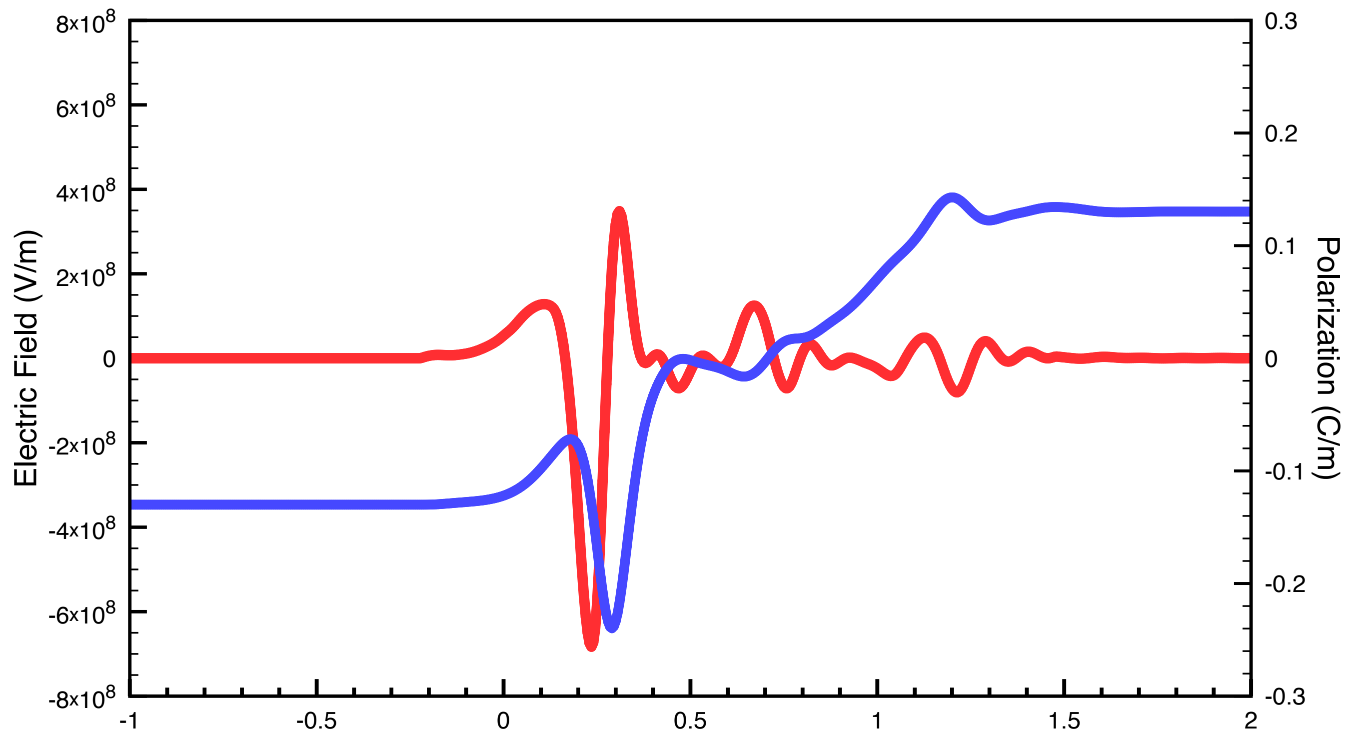
$Sn_2P_2S_6$: x-ray diffraction







- Simulated dynamics in anharmonic potential suggest flip at for ~ 6 MV/cm, single cycle pulses

[S. Gruebel, arXiv:1602.05435]

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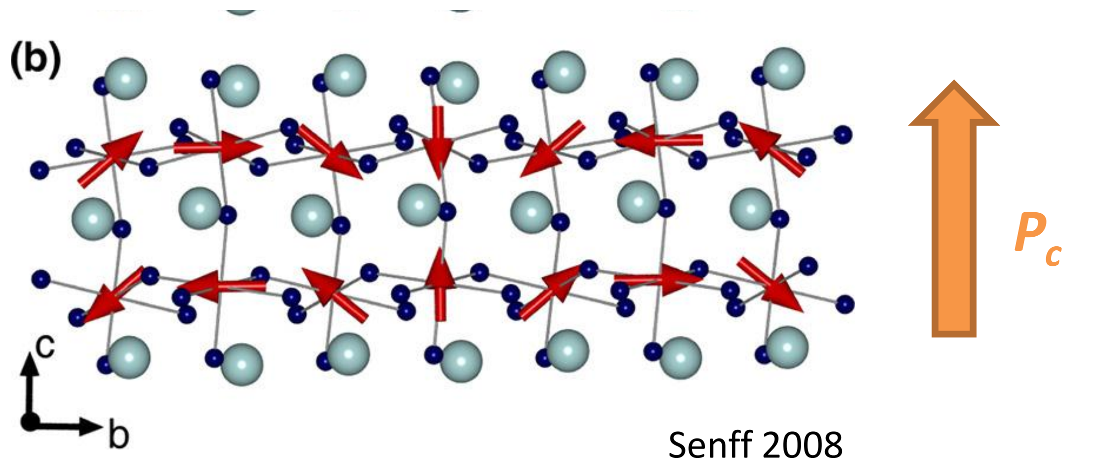
“Direct” control example 2:

Spin dynamics of a coherent electromagnon

THz excitation: path to fast control of multiferroics?

$TbMnO_3$

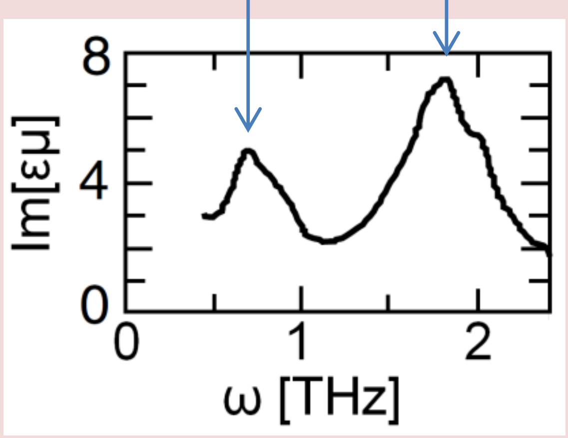


Senff 2008

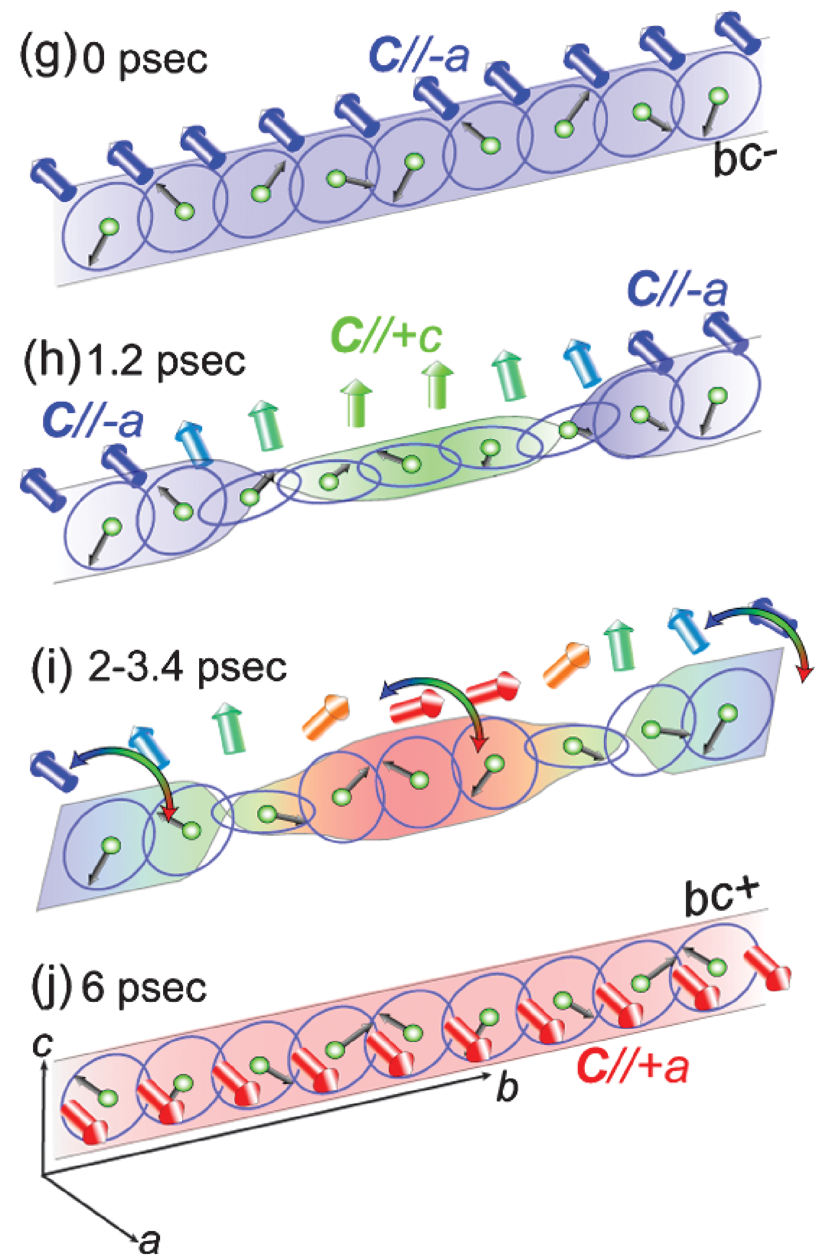
excitations due to the electromagnetic coupling:

higher-harmonic, ellipticity, phonons 0.7 THz

spin-spiral excitation, 1.8 THz

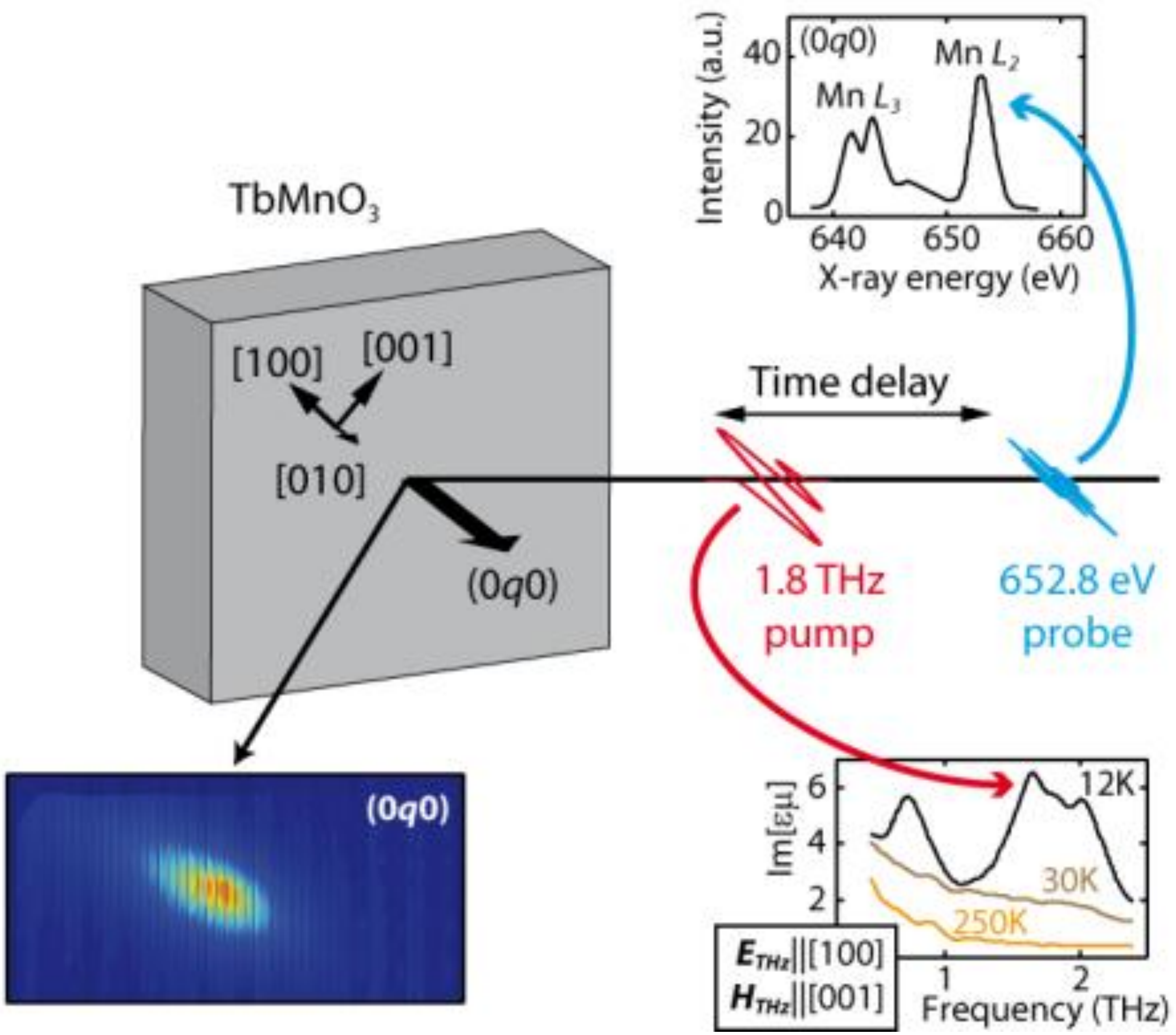


[Y. Takahashi et al., PRL **101**, 187201 (2008)]



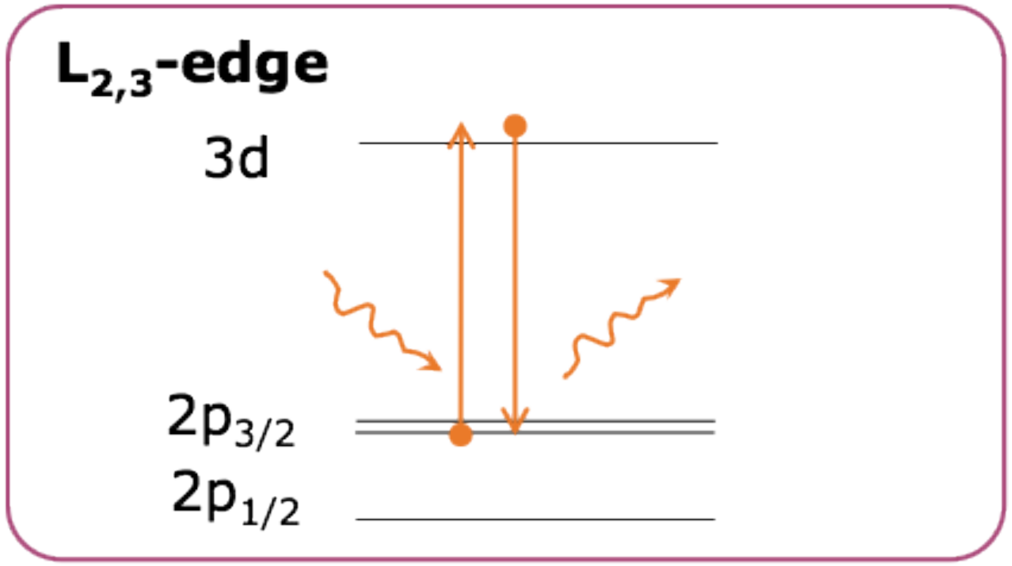
[Mochizuki & Nagaosa, PRL **105**, 147202 (2010)]

Experiment concept



Pump electromagnon with THz, watch spins with resonant x-ray diffraction

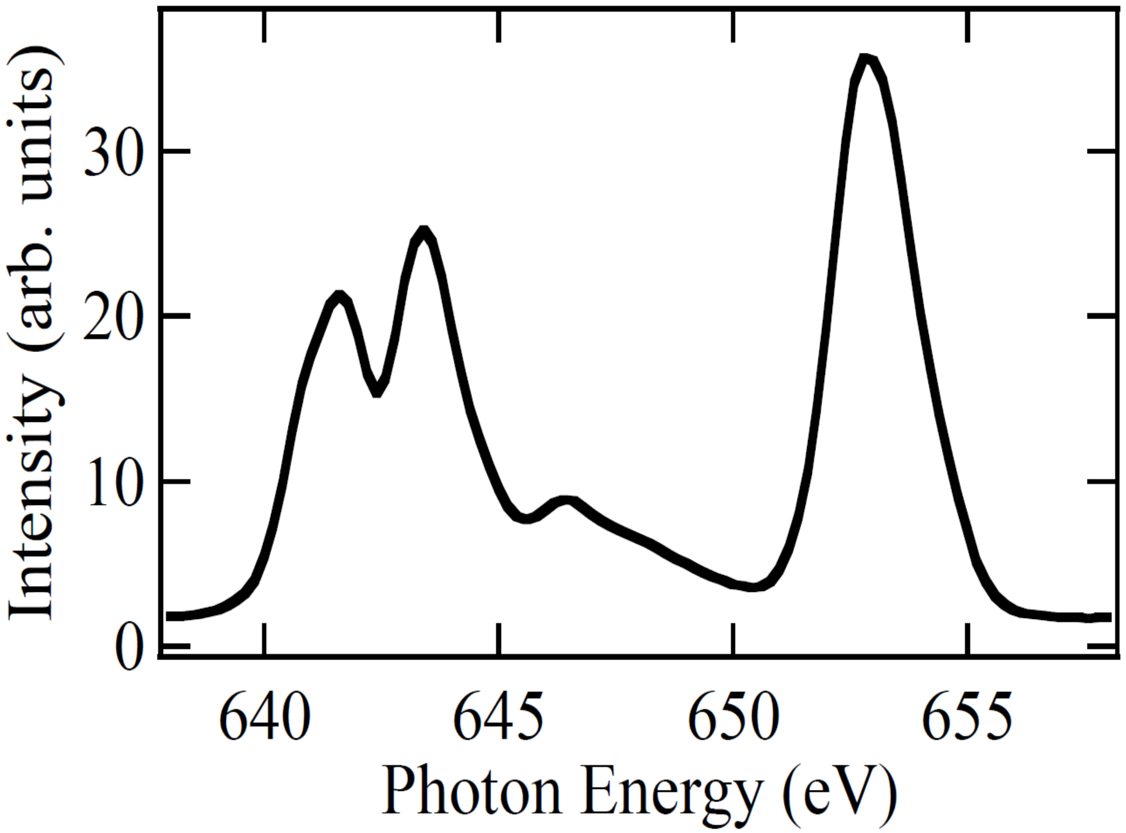
X-ray pulses: probe spin order



$$\left\langle \mathbf{T}_q^k \right\rangle \propto \sum_n \frac{\langle g| O |n\rangle \langle n| O^* |g\rangle}{E_n - E_g - \hbar\omega + i\Gamma}$$

- Experiment at LCLS
- Pulses of < 80 fs duration
- Time-stamping for < 250 fs resolution

- (0q0) reflection at Mn L-edges: only magnetic order



[Beye et al. Appl. Phys. Lett. 100, 121108 (2012)]

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Experiment team: $TbMnO_3$

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T. Kubacka
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SLAC:

M. Hoffmann
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W. Schlotter
G. Dakovski

LBNL:

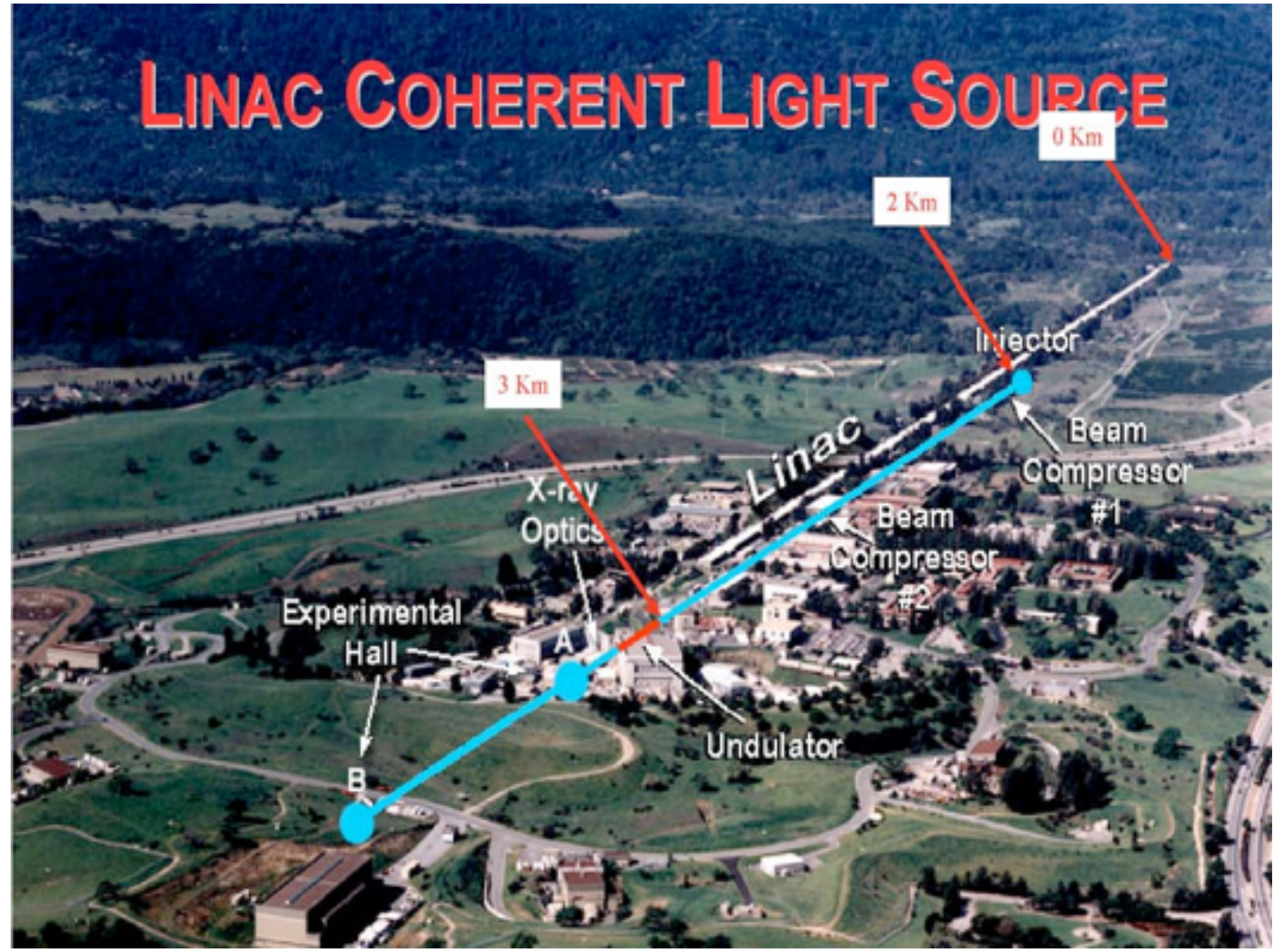
Y.-D. Chuang

Stanford:

W.-S. Lee
R. G. Moore

Johns Hopkins:

S. M. Koohpayeh



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C. Vicario
G. Ingold

Ch. Hauri
S. Gruebel
P. Beaud
L. Patthey

must

LCLS

FNSNF
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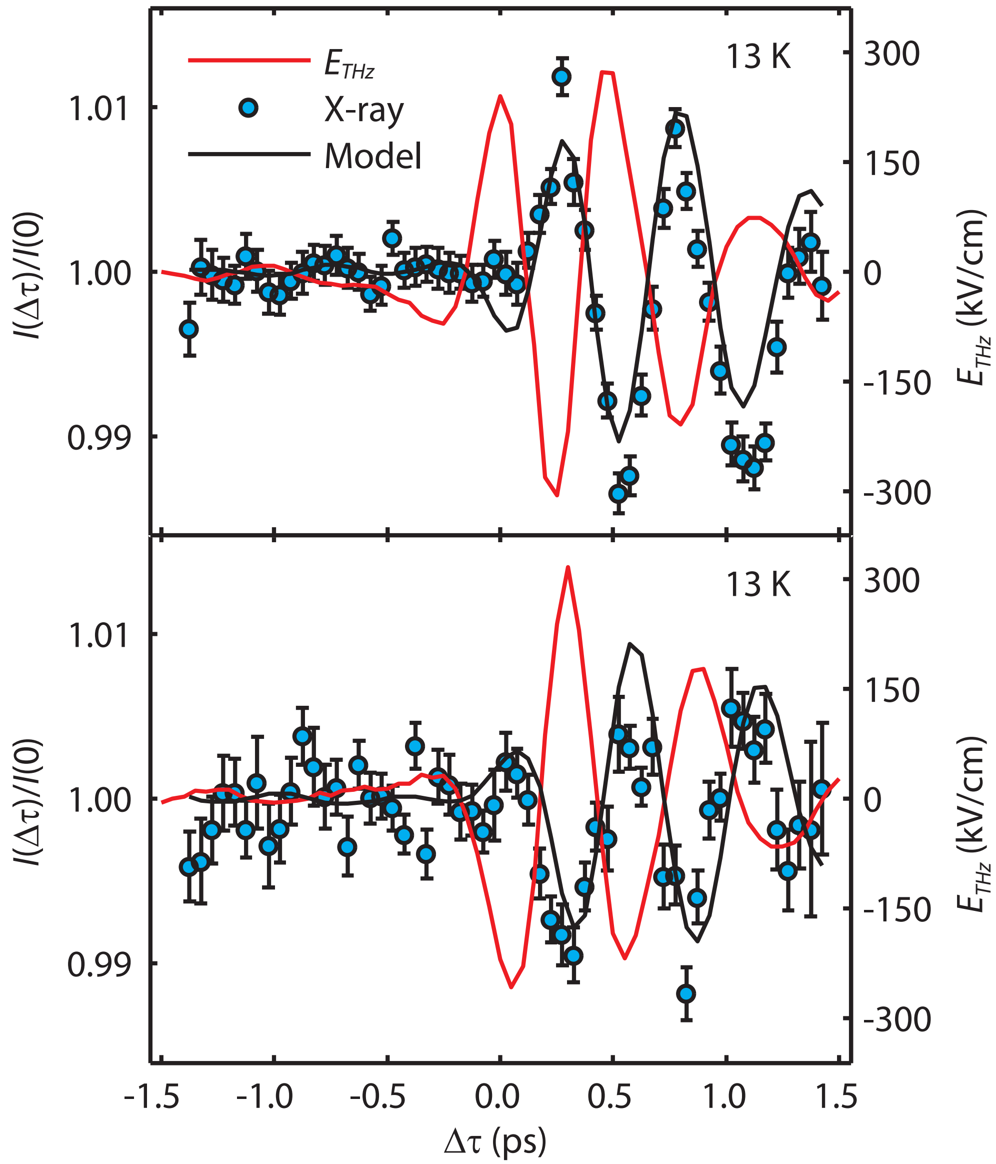
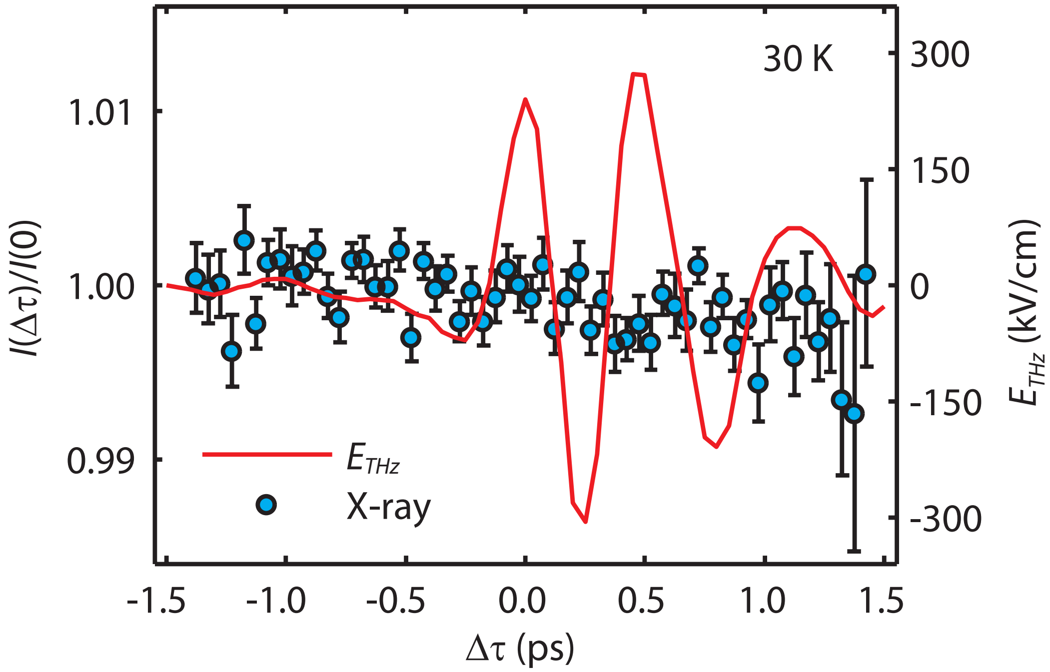
Results: coherent electromagnon

- E-field of THz → coherent spin response
- Measured spin response delayed by half cycle
- Response suppressed in non-multiferroic phase

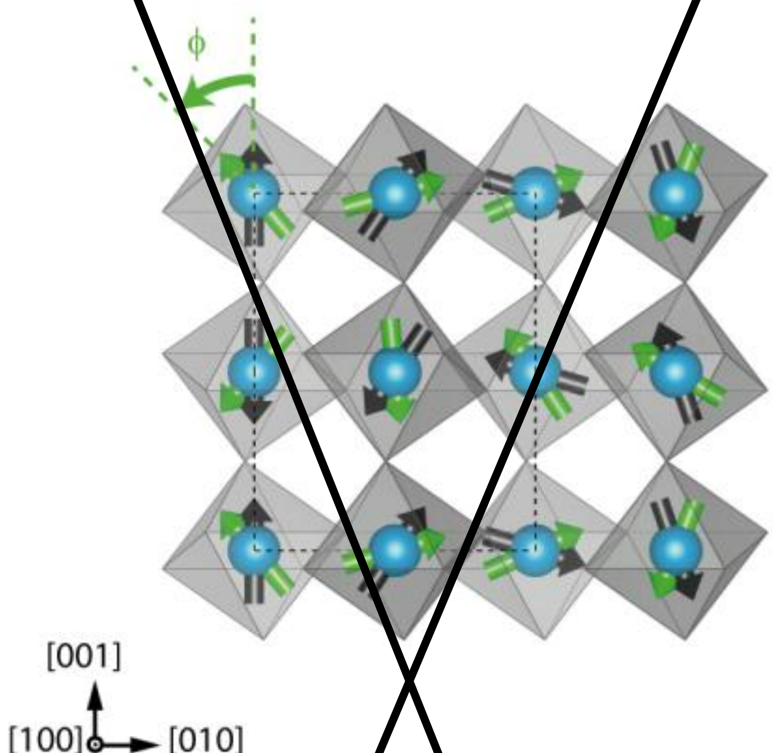


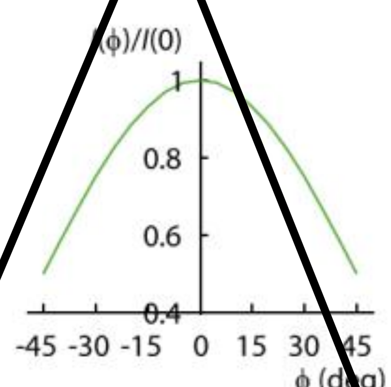


[T. Kubacka et al., Science **343**, 1333 (2014)]

Analyzing the motion

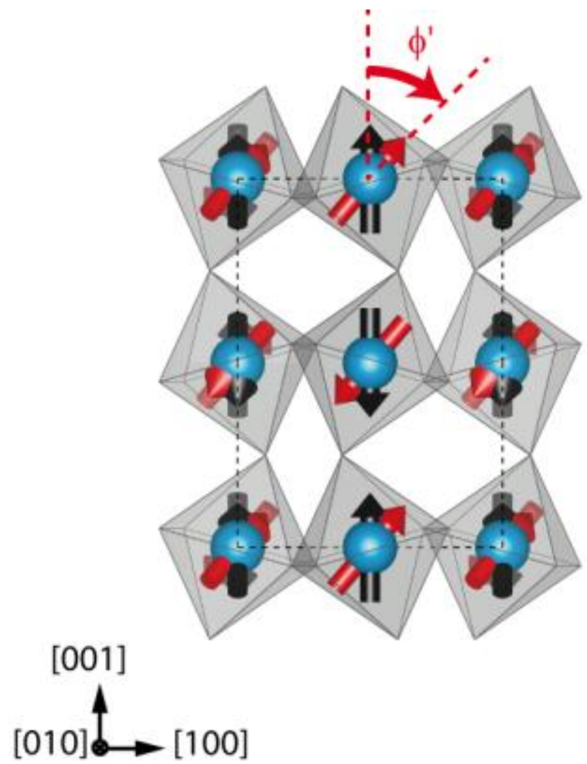
In-plane rotation

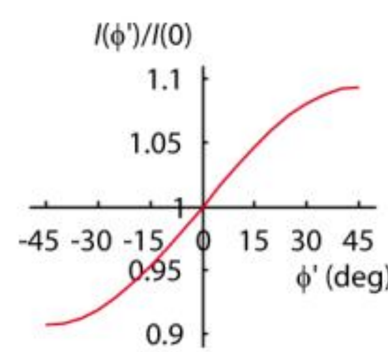




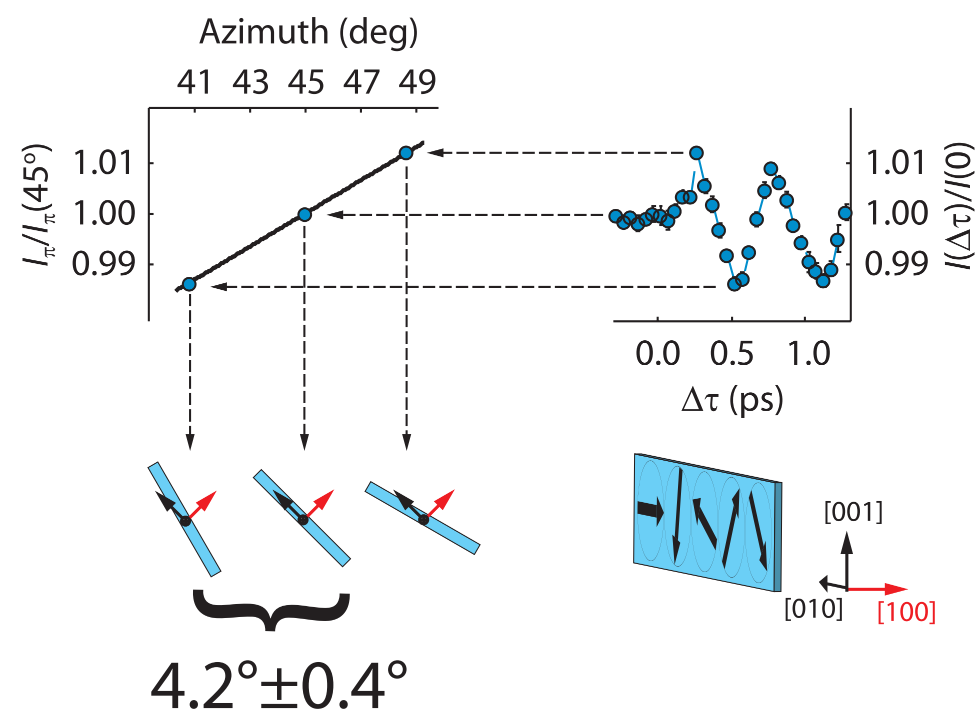
Even function

Rotation of spin planes





Odd function



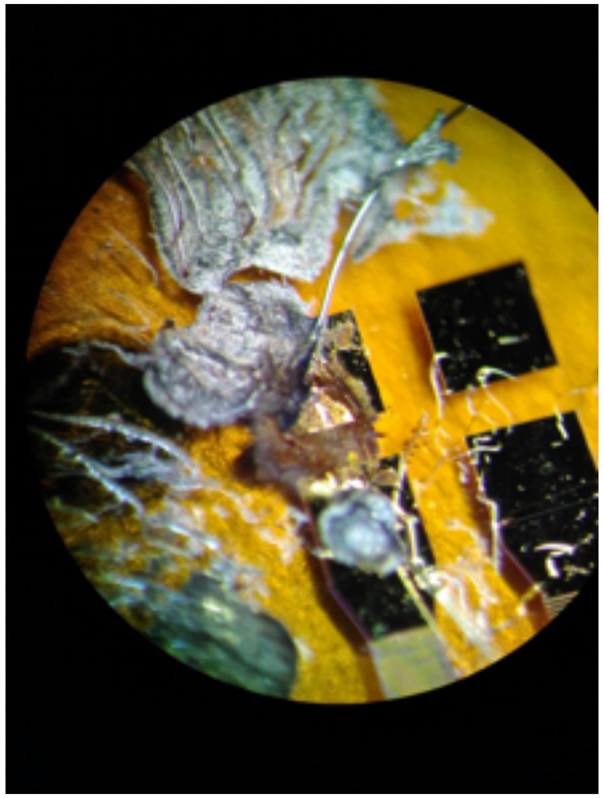
⇒ 4.2 ± 0.4 degree rotation of spin planes

⇒ Switching at ~ 10 MV/cm (extrapolation)

[T. Kubacka et al., Science **343**, 1333 (2014)]

Scaling this up?

- Low-field experiments on ferroelectrics and multiferroics with “single/few cycle” THz seem both consistent with multi-MV/cm fields



- Problem: High fields can lead to dielectric breakdown
- Material dependent, but ~ 1 MV/cm is “danger zone”

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Outlook

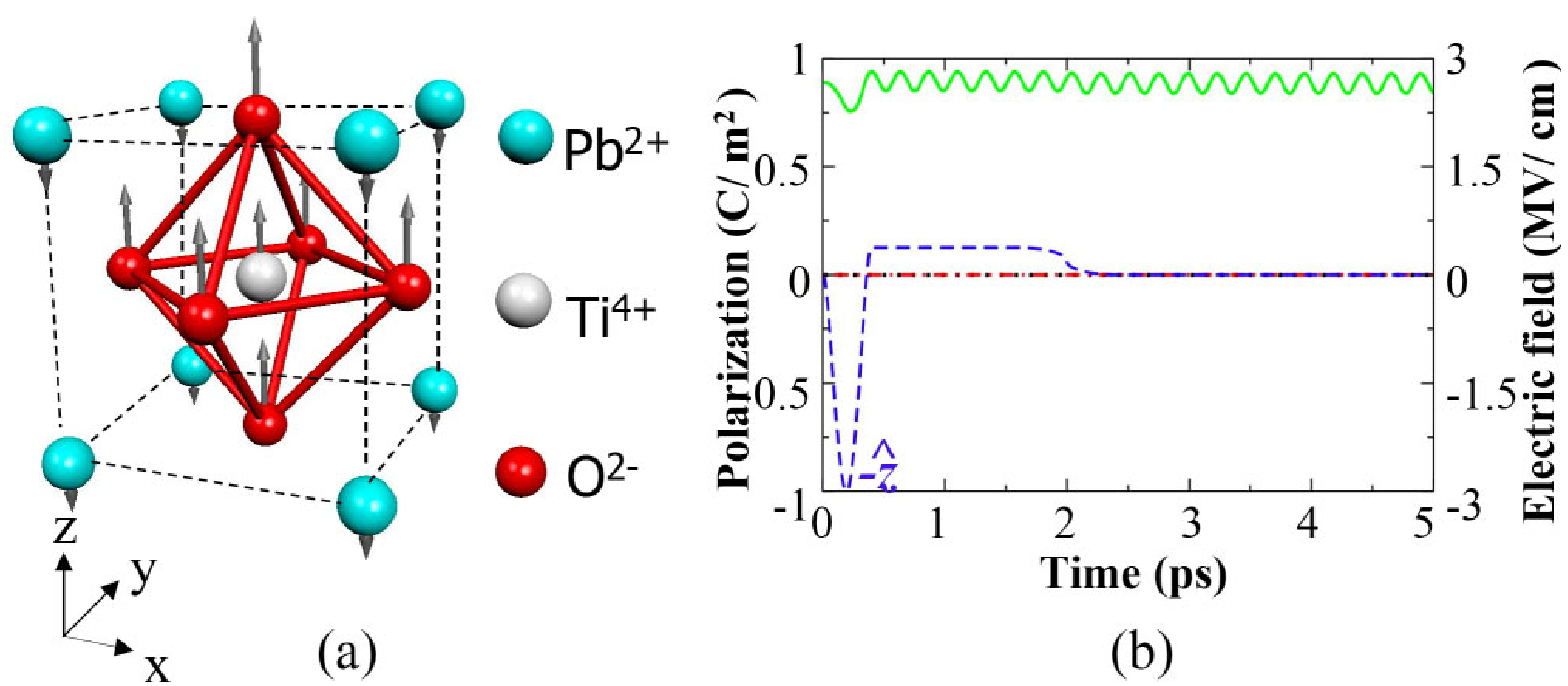
- Possible “way out”: work with modes that have (initially) low damping
- Spread interaction out over several vibrational cycles
- Requires generation of trains / tunable bandwidth / chirping

Outlook

PRL **102,** 247603 (2009) PHYSICAL REVIEW LETTERS week ending 19 JUNE 2009

Collective Coherent Control: Synchronization of Polarization in Ferroelectric $PbTiO_3$ by Shaped THz Fields

Tingting Qi,[1] Young-Han Shin,[1] Ka-Lo Yeh,[2] Keith A. Nelson,[2] and Andrew M. Rappe[1]
[1]*The Makineni Theoretical Laboratories, Department of Chemistry, University of Pennsylvania, Philadelphia, Pennsylvania 19104–6323, USA*
[2]*Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*
(Received 16 November 2008; published 19 June 2009)



- Theory study of FE switching
- Is a single half-cycle really the best?

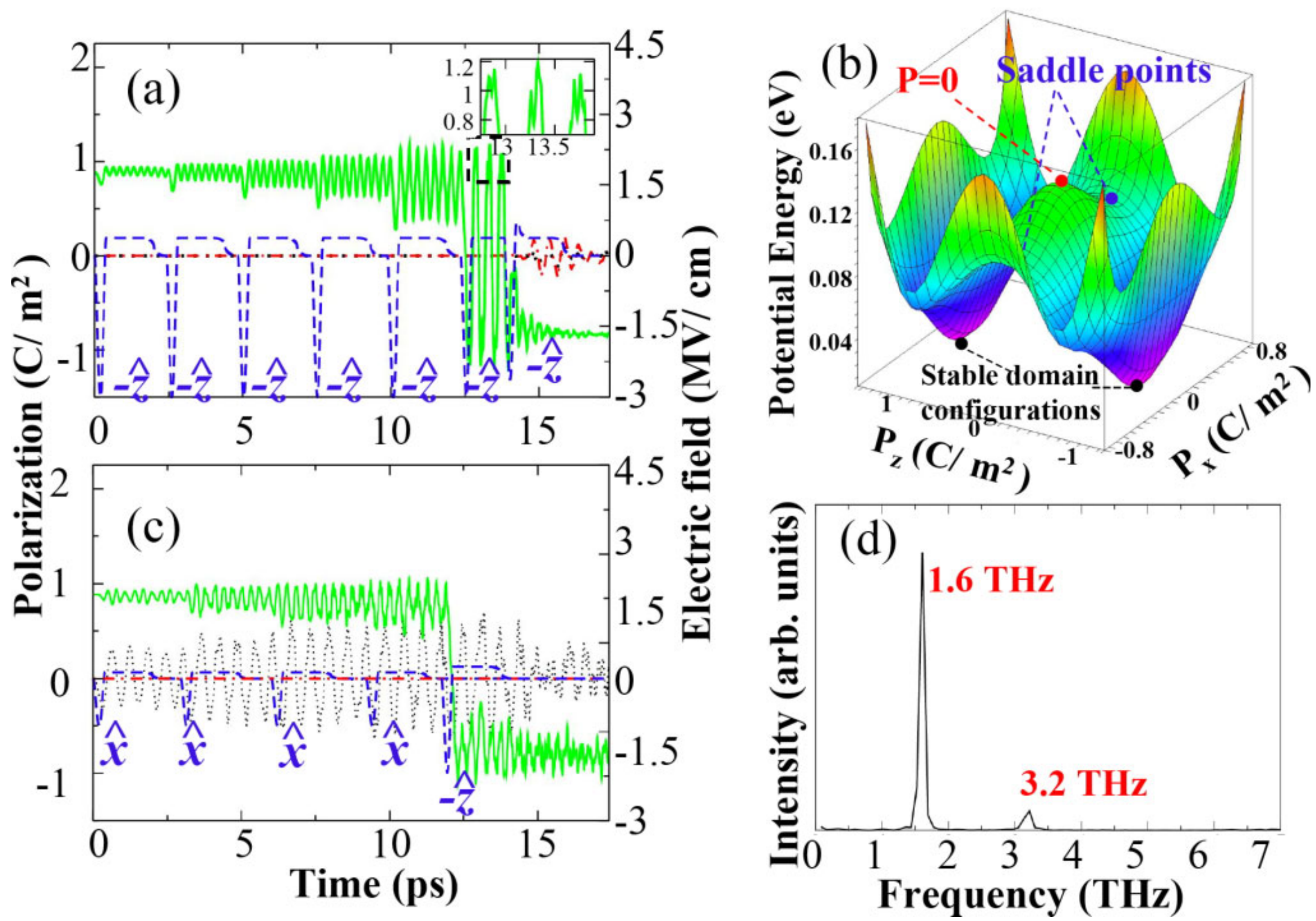
Outlook

PRL **102,** 247603 (2009) PHYSICAL REVIEW LETTERS week ending 19 JUNE 2009

Collective Coherent Control: Synchronization of Polarization in Ferroelectric $PbTiO_3$ by Shaped THz Fields

Tingting Qi,[1] Young-Han Shin,[1] Ka-Lo Yeh,[2] Keith A. Nelson,[2] and Andrew M. Rappe[1]
[1]*The Makineni Theoretical Laboratories, Department of Chemistry, University of Pennsylvania, Philadelphia, Pennsylvania 19104–6323, USA*
[2]*Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*
(Received 16 November 2008; published 19 June 2009)

- Sequence of 7 half-cycle pulses make switching feasible
- Also tricks with polarization possible
- Key: increase spectral weight overlap with modes



Outlook

PRL **105,** 203003 (2010) PHYSICAL REVIEW LETTERS week ending 12 NOVEMBER 2010

Ladder Climbing on the Anharmonic Intermolecular Potential in an Amino Acid Microcrystal via an Intense Monocycle Terahertz Pulse

Mukesh Jewariya,[1,2] Masaya Nagai,[1,3,*] and Koichiro Tanaka[1,2,4]
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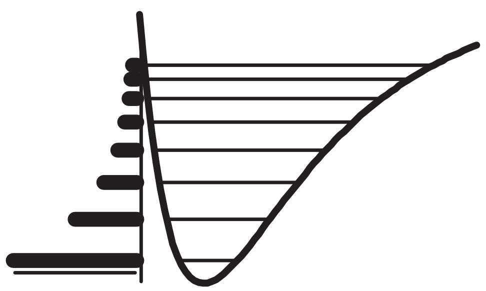
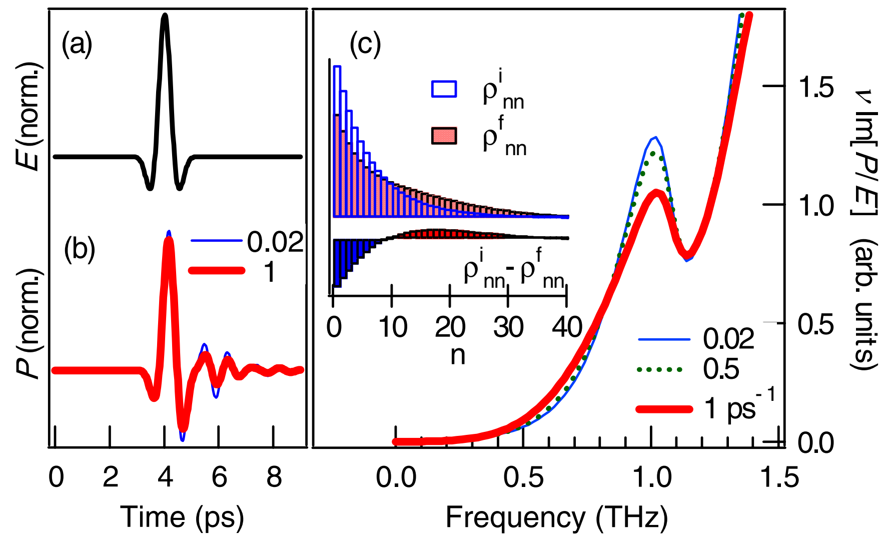
[2]*Core Research for Evolutional Science and Technology, Japan Science and Technology Agency, Japan*
[3]*Precursory Research for Embryonic Science and Technology, Japan Science and Technology Agency, Japan*
[4]*Institute for Integrated Cell-Material Sciences, Kyoto University, Kyoto 606-8501, Japan*
(Received 1 June 2010; published 11 November 2010)

arginine



- Amplitude-dependent vibrational frequency
- Shown in an amino acid crystal
- Indicates pulse shaping important

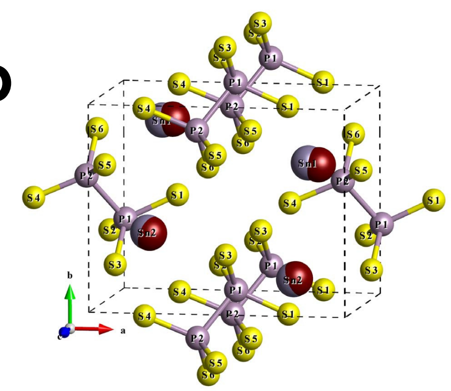


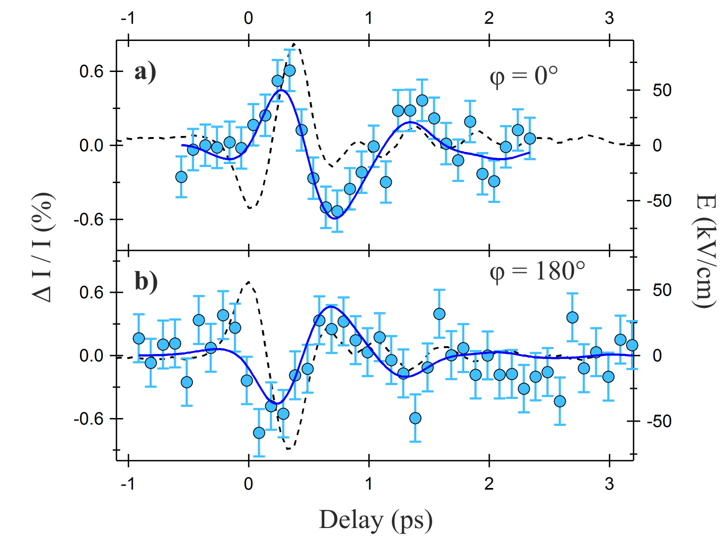
Ideal THz source characteristics

- Center frequency tunable from 0.5-18 THz
  - Covers most strong vibrational lines, (electro)magnons
- Phase stability critical (better than 10%)
- Bandwidth tunable from ~ 50% to ~5%
  - Idea is to match material
- Ability to chirp frequency by up to 30%
  - Ladder climbing
- Ability to shape polarization with time interesting, maybe not essential
- Peak fields not really the best quality factor, rather pulse energy > 0.01-0.1 mJ (depends strongly on spectral overlap with resonance)

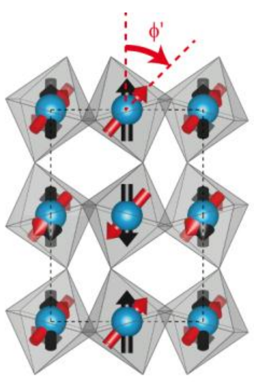
Summary

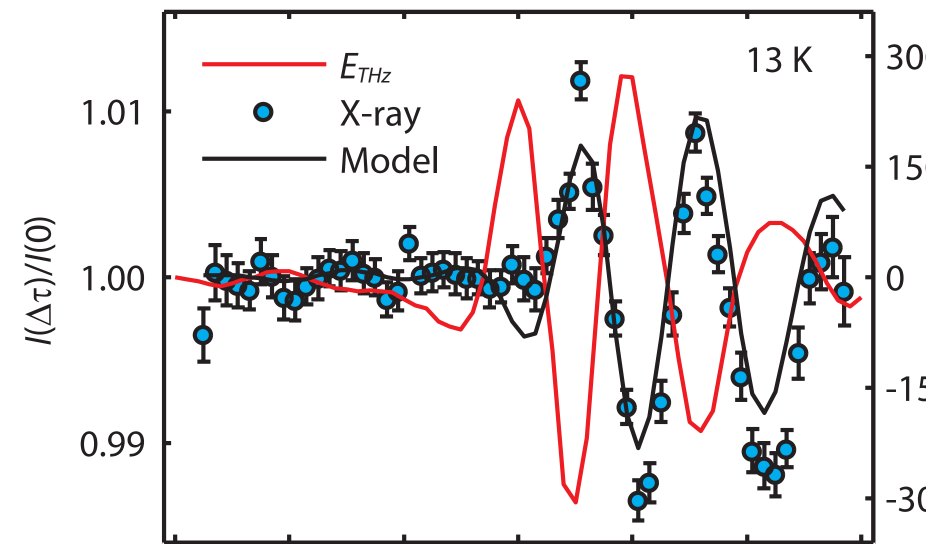
- THz can potentially drive OP dynamics directly





  - Driving soft mode in ferroelectric, consistent with switching at ~ 0.8 MV/cm
  - Drive spin structure changes with E-field, switching expected at ~ 10 MV/cm





- Main barrier is damage from avalanche ionization
- Tunable bandwidth/shaping as solution?

Acknowledgments

SLAC/SIMES

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