Terahertz dynamics of quantum materials

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Ultrafast Dynamics...
@UltrafastSU
What are quantum materials?

- Materials where *emergent* properties appear (cannot be predicted by properties of single atoms)

**Examples:**
- Magnetism
- Superconductivity
- Strongly correlated phenomena
- Topologically protected states

- Interplay of many degrees of freedom important (lattice, spin, electrons): many occurs at THz frequencies (meV energies)
Why ultrafast x-ray probes are important?

- femtoseconds are the time-scales of electronic and atomic motion

- paradigm shift where we can look *directly* at quantum mechanics in time and space

- new fundamental understanding of open problems in physics of quantum materials
Why high rep-rate XFELs for condensed matter?

- Ultrafast imaging with nanometer resolution
- Electron and spin spectroscopy with femtosecond resolution
- High-sensitivity time-resolved x-ray diffraction
Outline

- **Three scientific cases:**
  - Imaging of ultrafast magnetism
  - Nonlinear phonon dynamics
  - Dynamical multiferroicity

- A “dream” SASE4/SASE5 implementation at European XFEL
Why magnetism and why ultrafast?

Why the Future of Data Storage is (Still) Magnetic Tape

Disk drives are reaching their limits, but magnetic tape just gets better and better

- 90%+ storage in data-centers worldwide is magnetic (cheap and reliable)
- Bottle-neck for computing speed is storage
- Data-centers are very energy-hungry (most energy goes into heat and cooling):
  - 5% world electricity, 2-3% emission (now)
  - 20% world electricity (2025)
Ultrafast magnetism


X-ray magnetic holography: some details


X-ray holography of ultrafast magnetism: femtosecond movies at the nanoscale

Interference pattern on DSSC detector

Sample with reference structure

Skyrmion

S. Bonetti et al, XFEL Proposal 2222, beam time allocated May 2019
SrTiO$_3$ (STO): perovskite with soft phonon mode

Idea: drive the soft phonon and probe the atomic motion directly

Collaboration with Matthias Hoffmann and Mike Kozina, Stanford / SLAC

X-ray detector

SrTiO$_3$/LSAT sample

THz pump pulse

Hard x-ray probe pulse (30 fs)

(2 -2 3) reflection

Mechanism: nonlinear (anharmonic) coupling

- 1.3 pm motion (15% of full displacement in the ferroelectric BaTiO$_3$ at RT)
- Drive 1 THz mode (TO$_1$), excite 5 THz mode (TO$_2$)
- Excite also “silent” mode at 8 THz (TO$_3$): access of symmetry-forbidden modes via nonlinear coupling

Next step: quantum paraelectric phase (<35 K) and dynamical multiferroicity

Collaboration with Sasha Balatsky, NORDITA

Effective magnetic field from driven phonons

- Two orthogonal phonons at $f_1 = 16$ and $f_2 = 17$ THz

- Effective circular orbit of the ions, at the beating frequency $f_1 - f_2$, resonant with magnon in the system

- Effective magnetic field produced

Dream set-up for quantum materials

- **Soft x-ray undulator**
- **Hard x-ray undulator**
- **THz undulator**
- End-station

Seeding arrows point to each component.
High-rep rate machine goals

- Simultaneous spectroscopy and diffraction (correlated materials, e.g. dynamical multiferroics, electron-phonon coupling in superconductors)

- Combined hard and soft x-ray ultrafast imaging (antiferromagnets with THz resonances)

- Dirac materials?
Thank you for your attention!