New development of XFEL experiments with synchronized optical lasers

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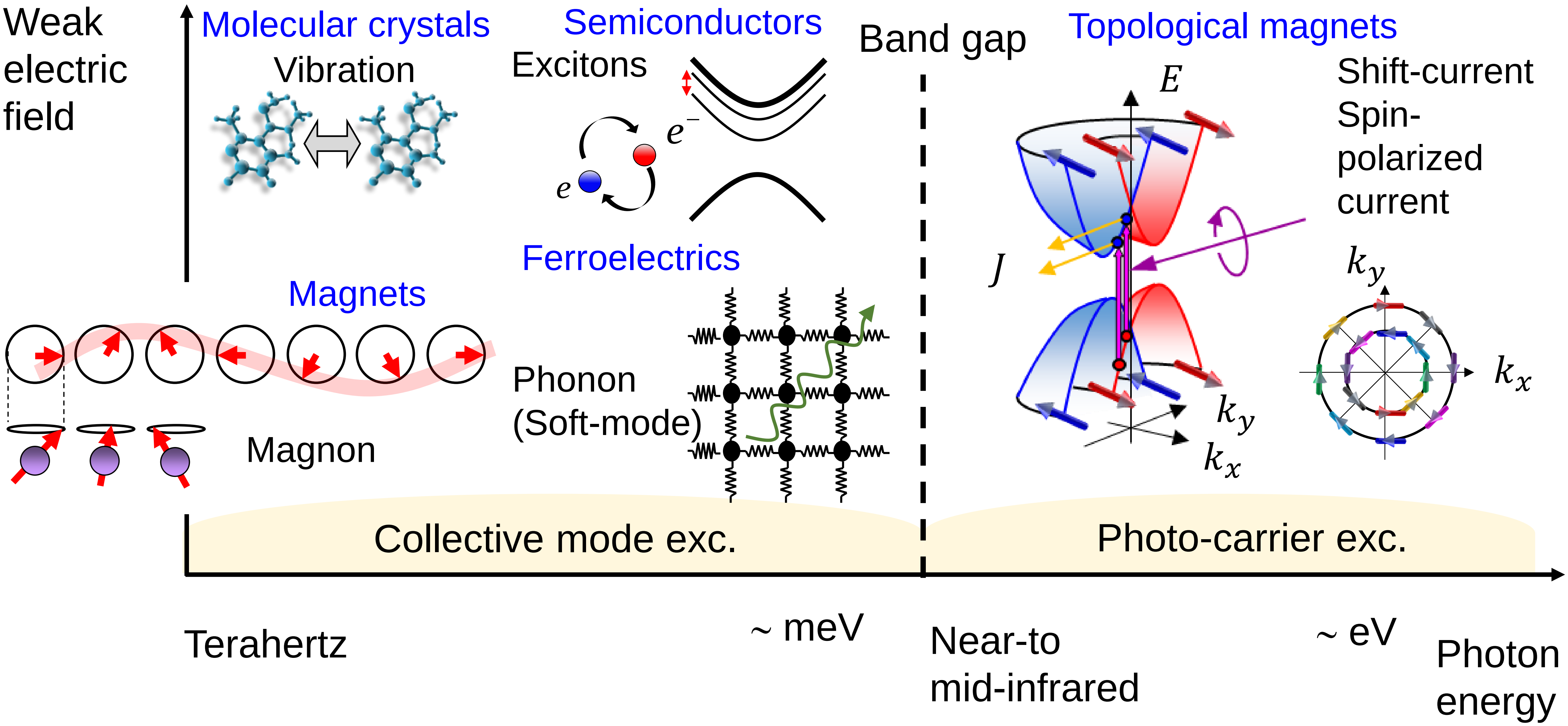
This breakout session aims to discuss the capabilities of the platforms with synchronized optical lasers in SACLA. First, we inform you about the current status of terahertz and ultrafast optics in SACLA. Next, recent activities on ultrafast photoinduced phase control of solids are presented from participants. Finally, future prospects on SACLA's experiments with synchronized optical lasers are discussed with all participants in this session.

Various optical phenomena in solids



Weak electric field

Molecular crystals

Vibration

Semiconductors

Excitons

Ferroelectrics

Magnets

Magnon

Phonon (Soft-mode)

Band gap

Topological magnets

Shift-current

Spin-polarized current

Collective mode exc.

Photo-carrier exc.

Terahertz

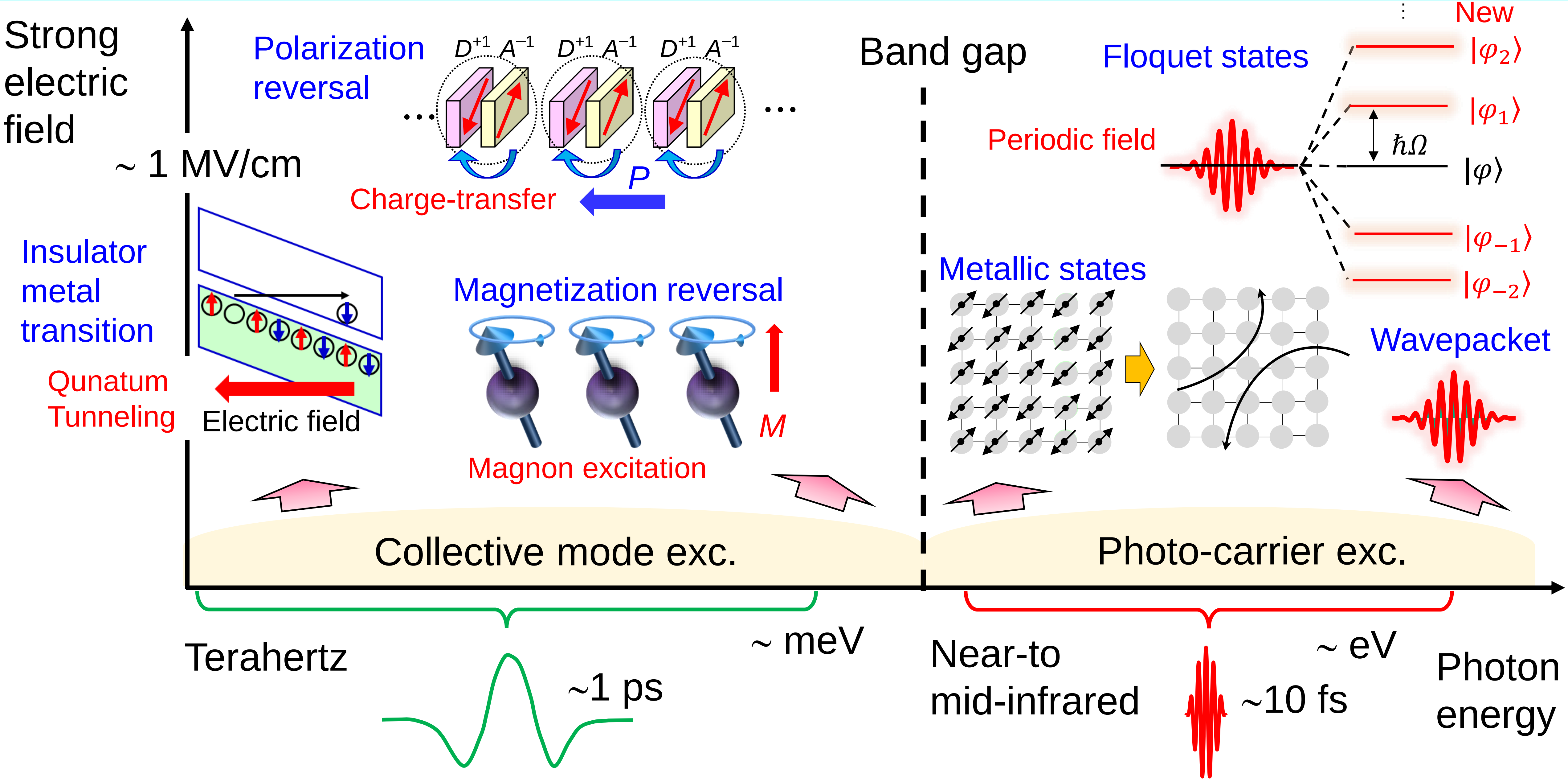
~ meV

Near-to mid-infrared

~ eV

Photon energy

New materials science by ultrafast and strong optical pulses



Strong electric field

~ 1 MV/cm

Polarization reversal

Charge-transfer

Insulator metal transition

Qunatum Tunneling

Electric field

Magnetization reversal

Magnon excitation

Collective mode exc.

Band gap

Floquet states

New

Periodic field

Metallic states

Wavepacket

Photo-carrier exc.

Terahertz

~ meV

~1 ps

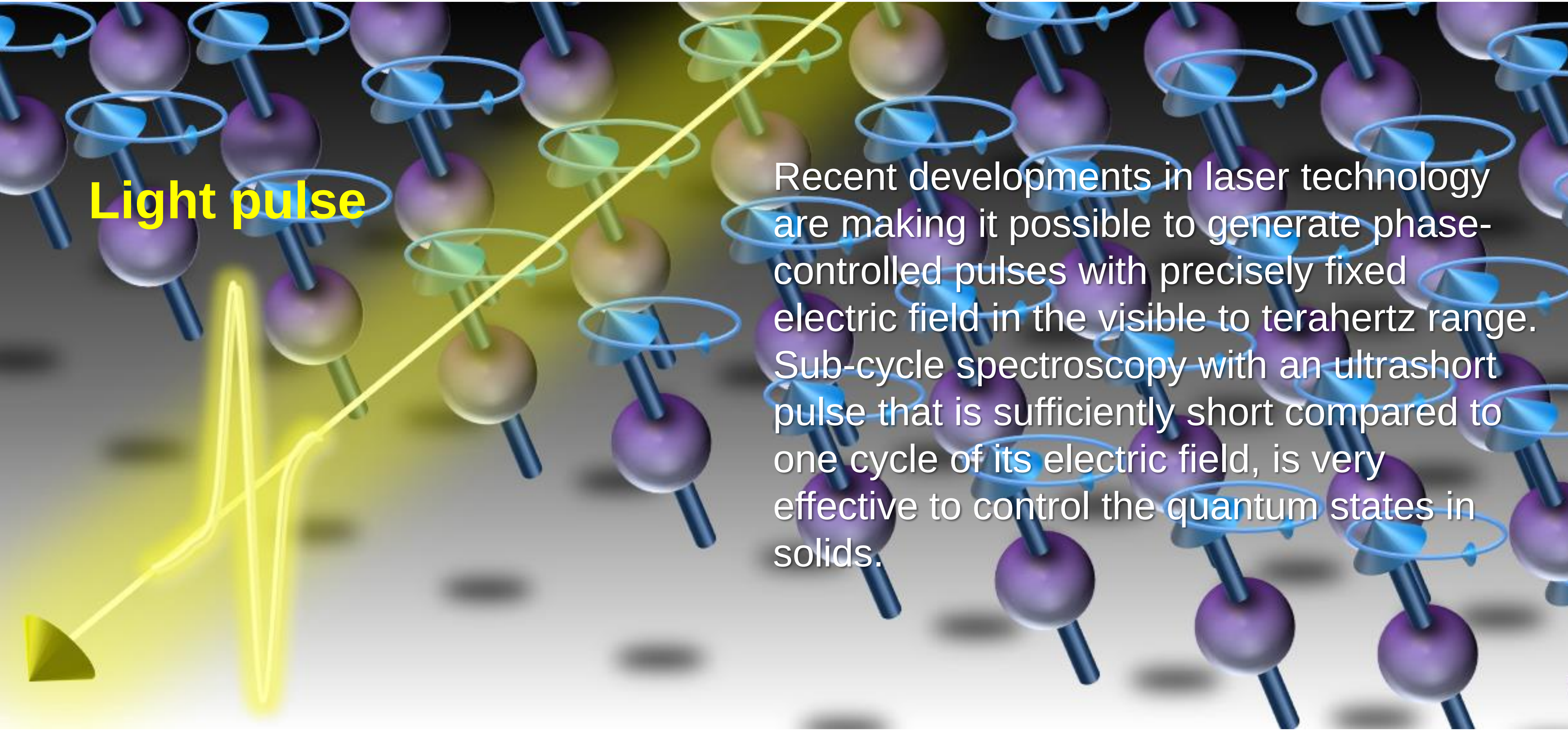
Near-to mid-infrared

~ eV

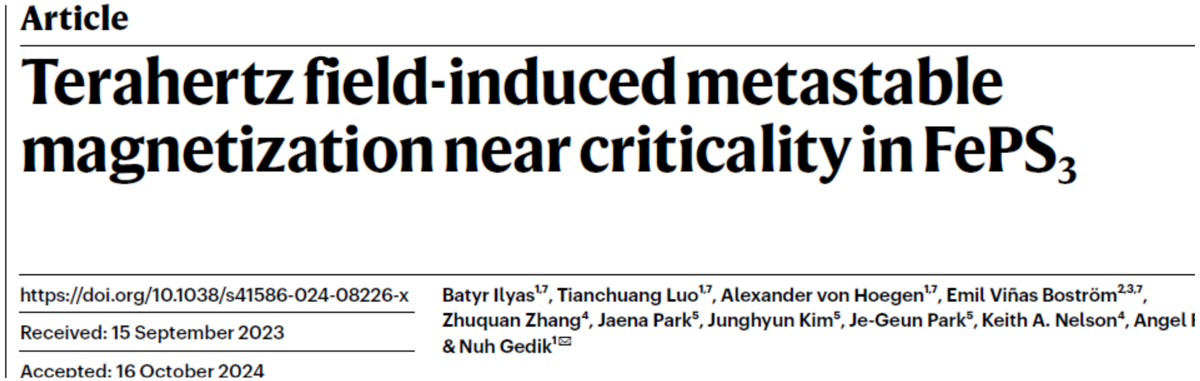
~10 fs

Photon energy

Purpose of this session: Sub-cycle spectroscopy in solids



Recent developments in laser technology are making it possible to generate phase-controlled pulses with precisely fixed electric field in the visible to terahertz range. Sub-cycle spectroscopy with an ultrashort pulse that is sufficiently short compared to one cycle of its electric field, is very effective to control the quantum states in solids.



Article

Terahertz field-induced metastable magnetization near criticality in $FePS_3$

https://doi.org/10.1038/s41586-024-08226-x

Received: 15 September 2023

Accepted: 16 October 2024

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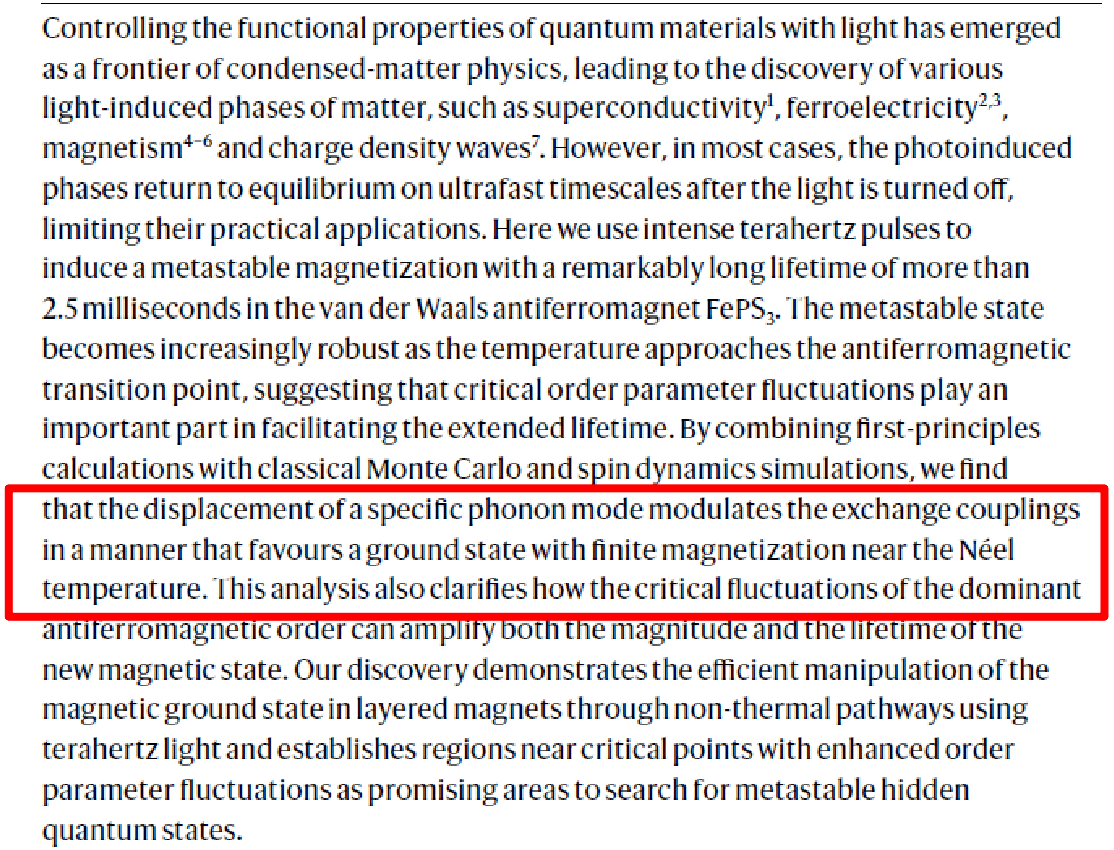
& Nuh Gedik[1]✉



Controlling the functional properties of quantum materials with light has emerged as a frontier of condensed-matter physics, leading to the discovery of various light-induced phases of matter, such as superconductivity[1], ferroelectricity[2,3], magnetism[4–6] and charge density waves[7]. However, in most cases, the photoinduced phases return to equilibrium on ultrafast timescales after the light is turned off, limiting their practical applications. Here we use intense terahertz pulses to induce a metastable magnetization with a remarkably long lifetime of more than 2.5 milliseconds in the van der Waals antiferromagnet $FePS_3$. The metastable state becomes increasingly robust as the temperature approaches the antiferromagnetic transition point, suggesting that critical order parameter fluctuations play an important part in facilitating the extended lifetime. By combining first-principles calculations with classical Monte Carlo and spin dynamics simulations, we find that the displacement of a specific phonon mode modulates the exchange couplings in a manner that favours a ground state with finite magnetization near the Néel temperature. This analysis also clarifies how the critical fluctuations of the dominant antiferromagnetic order can amplify both the magnitude and the lifetime of the new magnetic state. Our discovery demonstrates the efficient manipulation of the magnetic ground state in layered magnets through non-thermal pathways using terahertz light and establishes regions near critical points with enhanced order parameter fluctuations as promising areas to search for metastable hidden quantum states.

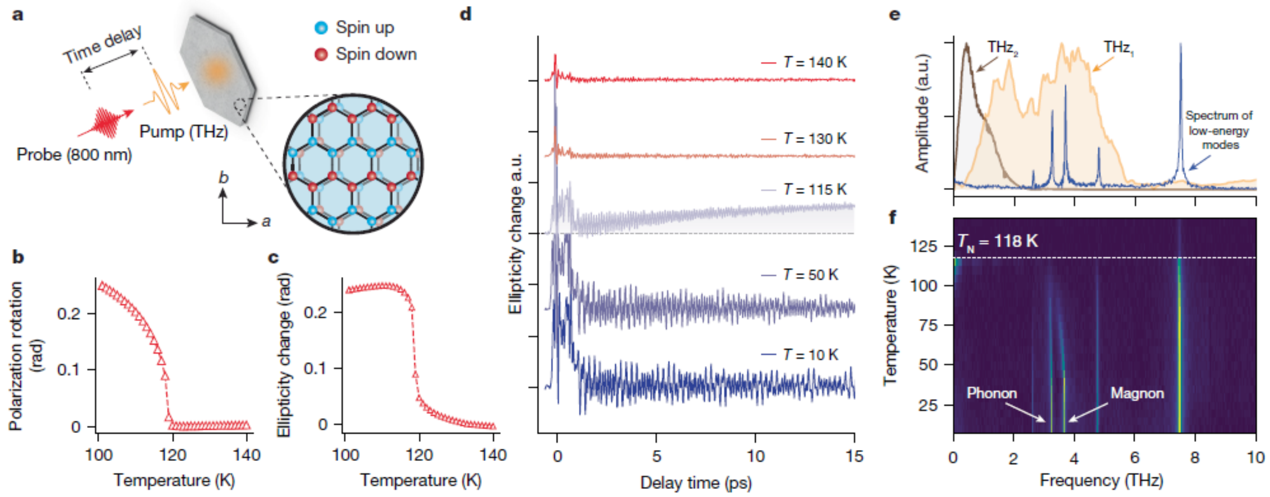
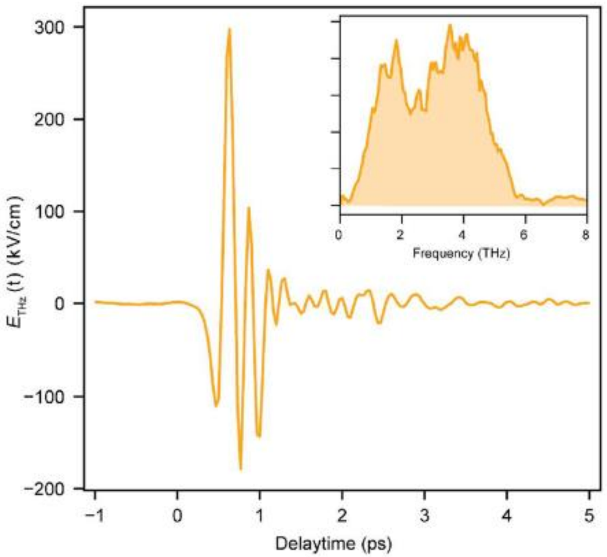
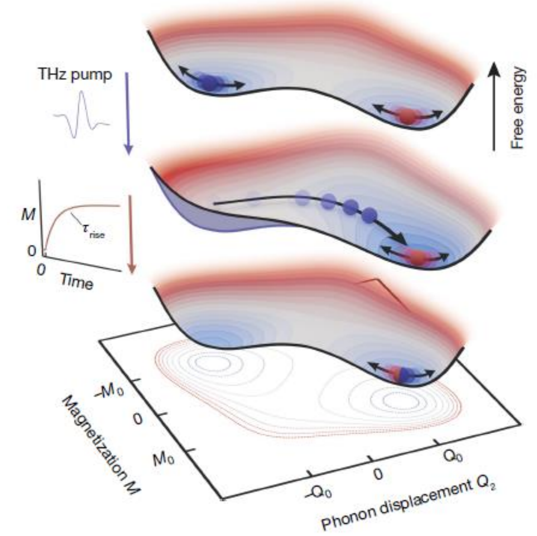


Fig. 1 | Experimental schematics and THz field-driven low-energy modes. **a**, An intense THz pulse (orange) drives low-energy collective excitations in $FePS_3$, and the induced transient changes in optical properties are probed with 800 nm probe pulse (red). Fe^{2+} ions form a hexagonal lattice and their spins arrange ferromagnetically along the zig-zag chain (*a*-axis) and antiferromagnetically between the adjacent chains (the structure in the circle). The magnetic coupling between the layers is antiferromagnetic together with a small interlayer shear distortion along the *a*-axis. **b**,**c**, Temperature dependencies of polarization rotation and ellipticity change signals in equilibrium, respectively. **d**, Probe ellipticity change signal time-traces at selected temperatures exhibit coherent oscillations induced by the THz field. As the temperature approaches the transition point (for example, at $T = 115$ K), the time traces start to build a net positive response. **e**, Fourier transform of the oscillations at $T = 10$ K (dark blue), which contains five prominent peaks that correspond to magnon and phonon modes. The spectral content of the excitation pulse is shown in orange, labelled as THz_1. For additional experiments, a THz field generated from a different source is used, with a spectrum shown in brown and labelled as THz_2, and it is not resonant with any of the modes. **f**, Temperature evolution of the Fourier spectrum. The colour bar is normalized. A notable change in the spectra occurs below the Néel temperature ($T_N = 118$ K) and the magnon mode softens on increasing temperature.



e



nature physics

ARTICLES

https://doi.org/10.1038/s41567-018-0408-1

Terahertz-driven phonon upconversion in $SrTiO_3$

M. Kozina[1*], M. Fechner[2], P. Marsik[3], T. van Driel[1], J. M. Glownia[1], C. Bernhard[3], M. Radovic[4], D. Zhu[1], S. Bonetti[5], U. Staub[4] and M. C. Hoffmann[1]

Direct manipulation of the atomic lattice using intense long-wavelength laser pulses has become a viable approach to create new states of matter in complex materials. Conventionally, a high-frequency vibrational mode is driven resonantly by a mid-infrared laser pulse and the lattice structure is modified through indirect coupling of this infrared-active phonon to other, lower-frequency lattice modulations. Here, we drive the lowest-frequency optical phonon in the prototypical transition metal oxide $SrTiO_3$ well into the anharmonic regime with an intense terahertz field. We show that it is possible to transfer energy to higher-frequency phonon modes through nonlinear coupling. Our observations are carried out by directly mapping the lattice response to the coherent drive field with femtosecond X-ray pulses, enabling direct visualization of the atomic displacements.

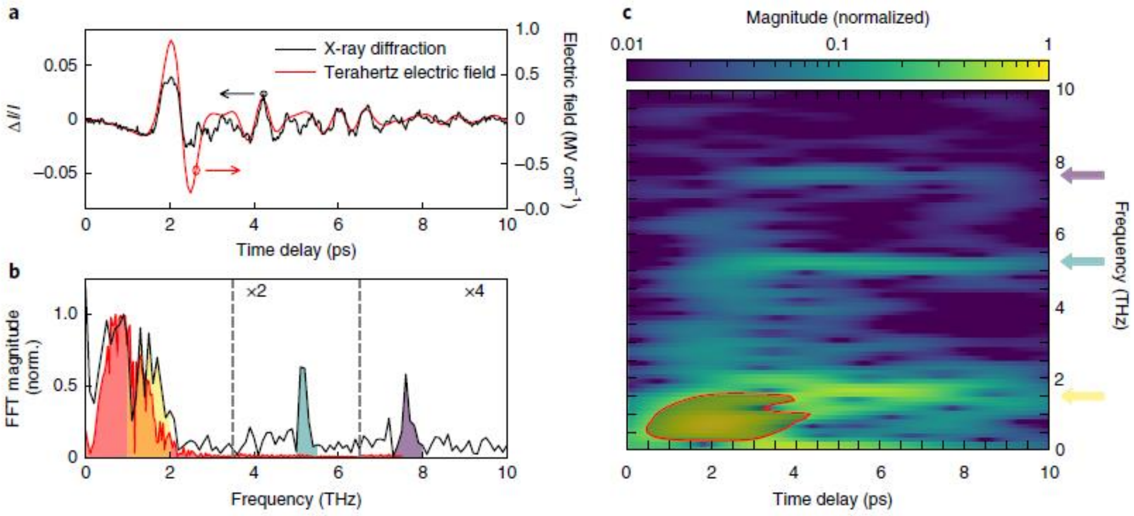


Fig. 2 | Time-resolved XRD data at 100 K. **a**, Intensity change of the (2 −2 3) peak in STO (black) as function of time delay compared to the terahertz pump field (red). The traces are shifted to overlap in time as their relative timing is known only within 1 ps. **b**, Fourier transform of the time-domain X-ra data shows distinct peaks, identified as the soft mode at 1.5 THz, the TO_2 mode at 5.15 THz and the TO_3 mode at 7.6 THz. The spectrum of the terahertz excitation pulse is shown in red. **c**, A short-time Fourier transformation with a 2.5 ps Kaiser window reveals the immediate onset of oscillations. The area outlined in red indicates the spectrogram of the terahertz pump pulse (contour at 30% of peak magnitude).

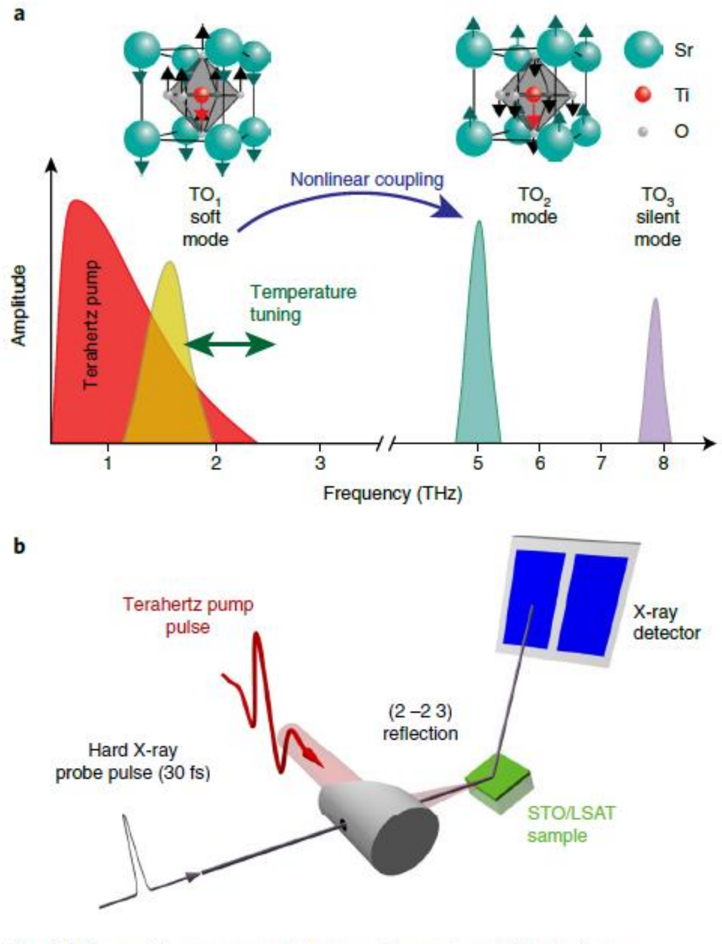


Fig. 1 | Phonon frequency spectrum and experimental overview. **a**, Strong terahertz radiation (red) interacts with the STO soft-mode phonon (yellow). The degree of resonant overlap can be tuned by temperature. Energy is exchanged with higher-frequency phonon modes (turquoise, purple) through nonlinear coupling. The STO unit cell and two lowest-frequency zone-centre TO eigenmodes are indicated at the top of the figure. **b**, Phonon motion is probed in the time domain with ultrafast XRD in reflection geometry.

$$\ddot{Q}_i + \frac{\partial V}{\partial Q_i} + \gamma_i \dot{Q}_i = Z_i^* \tilde{E}_{\mathrm{THz}}$$

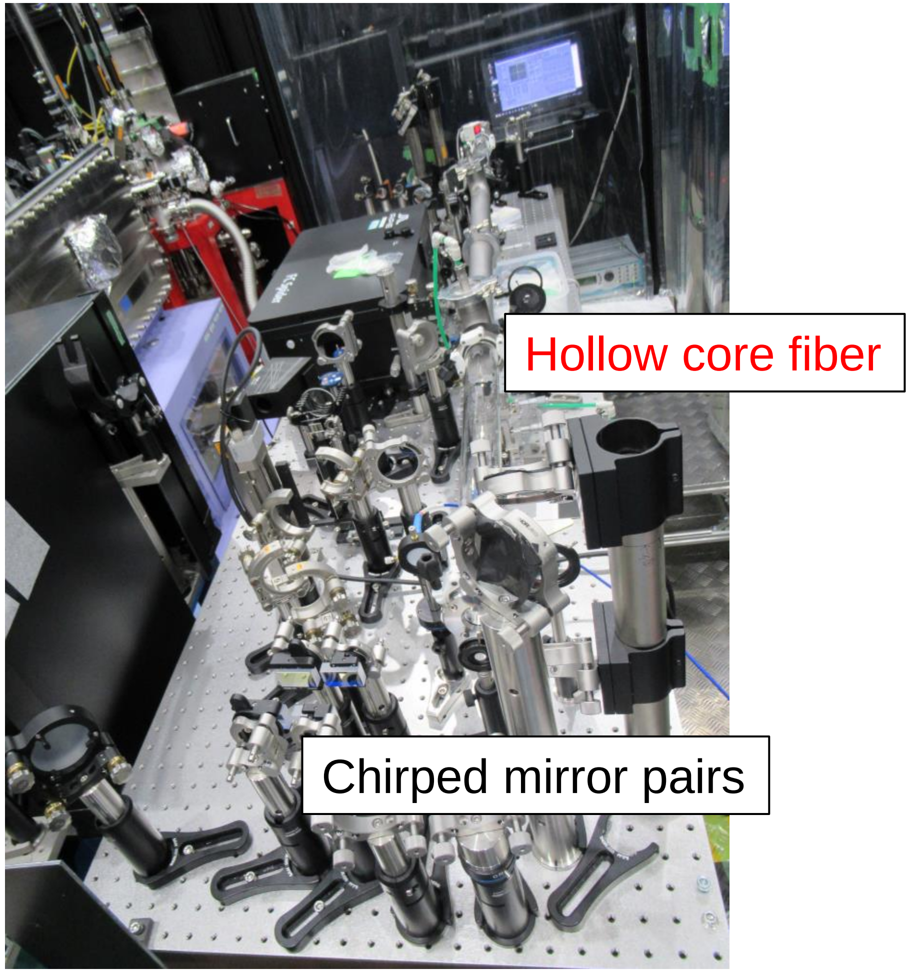
$$\ddot{Q}_1 + \gamma_1 \dot{Q}_1 + \tilde{\omega}_1^2 Q_1 = Z_1^* \tilde{E}_{\mathrm{THz}}$$

$$\ddot{Q}_2 + \gamma_2 \dot{Q}_2 + \omega_2^2 Q_2 = Z_2^* \tilde{E}_{\mathrm{THz}} - \psi_{12} Q_1^3$$

SACLA laser facility up-date

In this term, we developed two pumping systems combined with XFEL

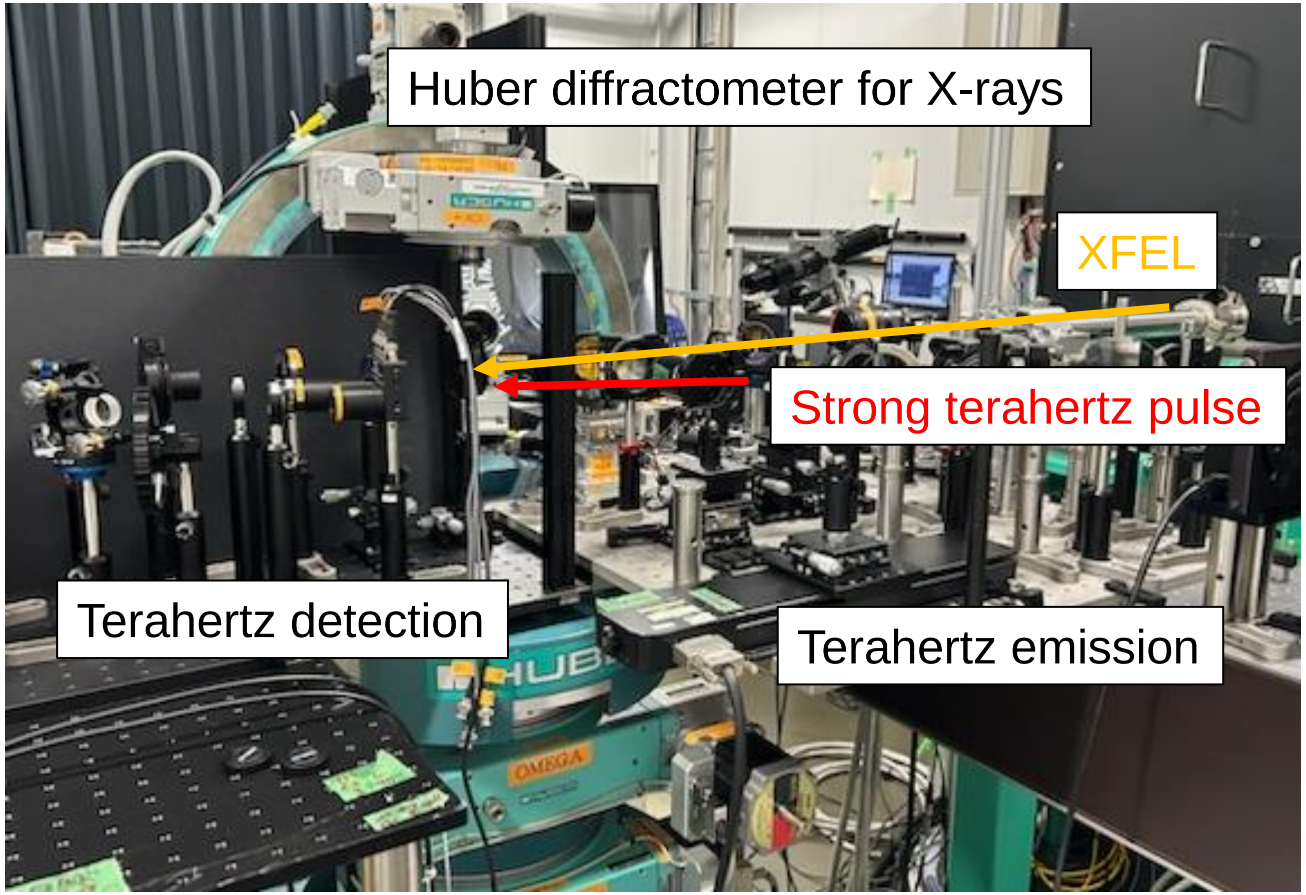
8 fs 800 nm laser pump system



Hollow core fiber

Chirped mirror pairs

Strong terahertz pump system



Huber diffractometer for X-rays

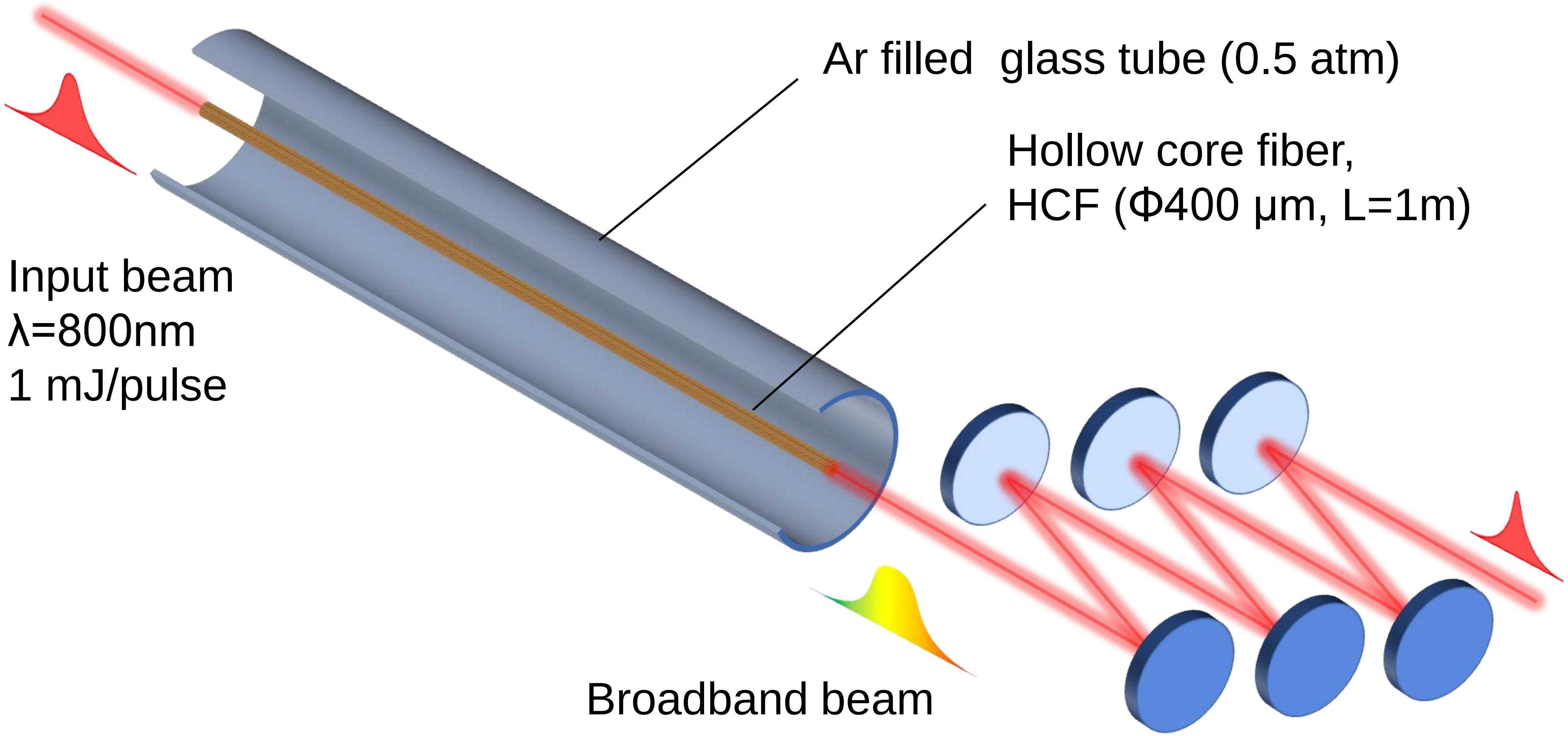
XFEL

Strong terahertz pulse

Terahertz detection

Terahertz emission

NIR Few-cycle pulses at SACLA



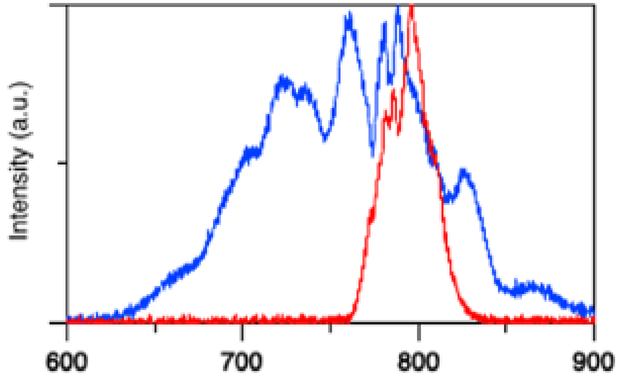
Ar filled glass tube (0.5 atm)

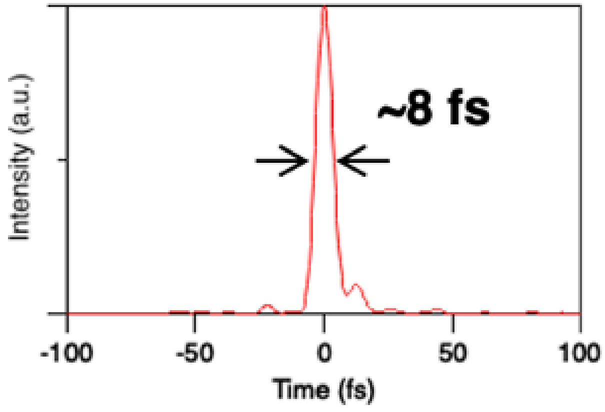
Hollow core fiber,
HCF (Φ400 μm, L=1m)

Input beam
λ=800nm
1 mJ/pulse

Broadband beam

Chirped mirror pairs





Compressed beam
λ=800nm
0.5mJ/pulse

- Conventional pulse compression using HCF filled with rear gas
- Available for BL1(EH4a) and BL3(EH2)
- Typical pulse duration / pulse energy: ~8 fs / 0.5 mJ/pulse

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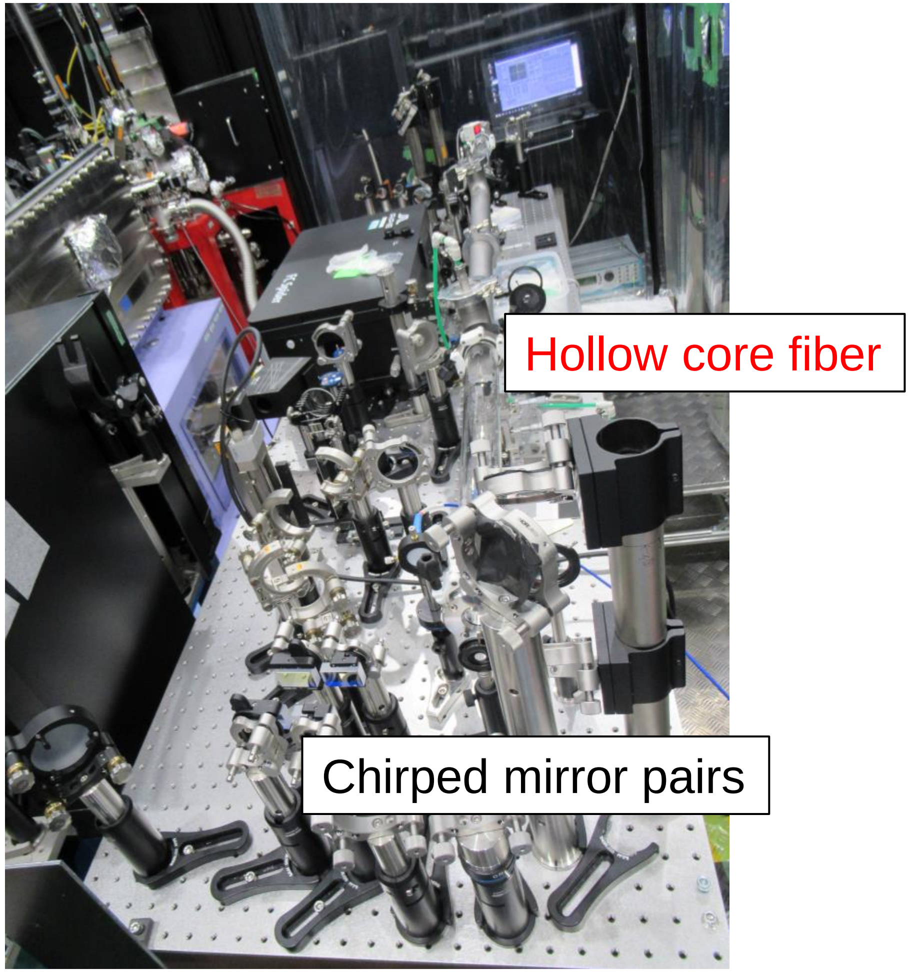
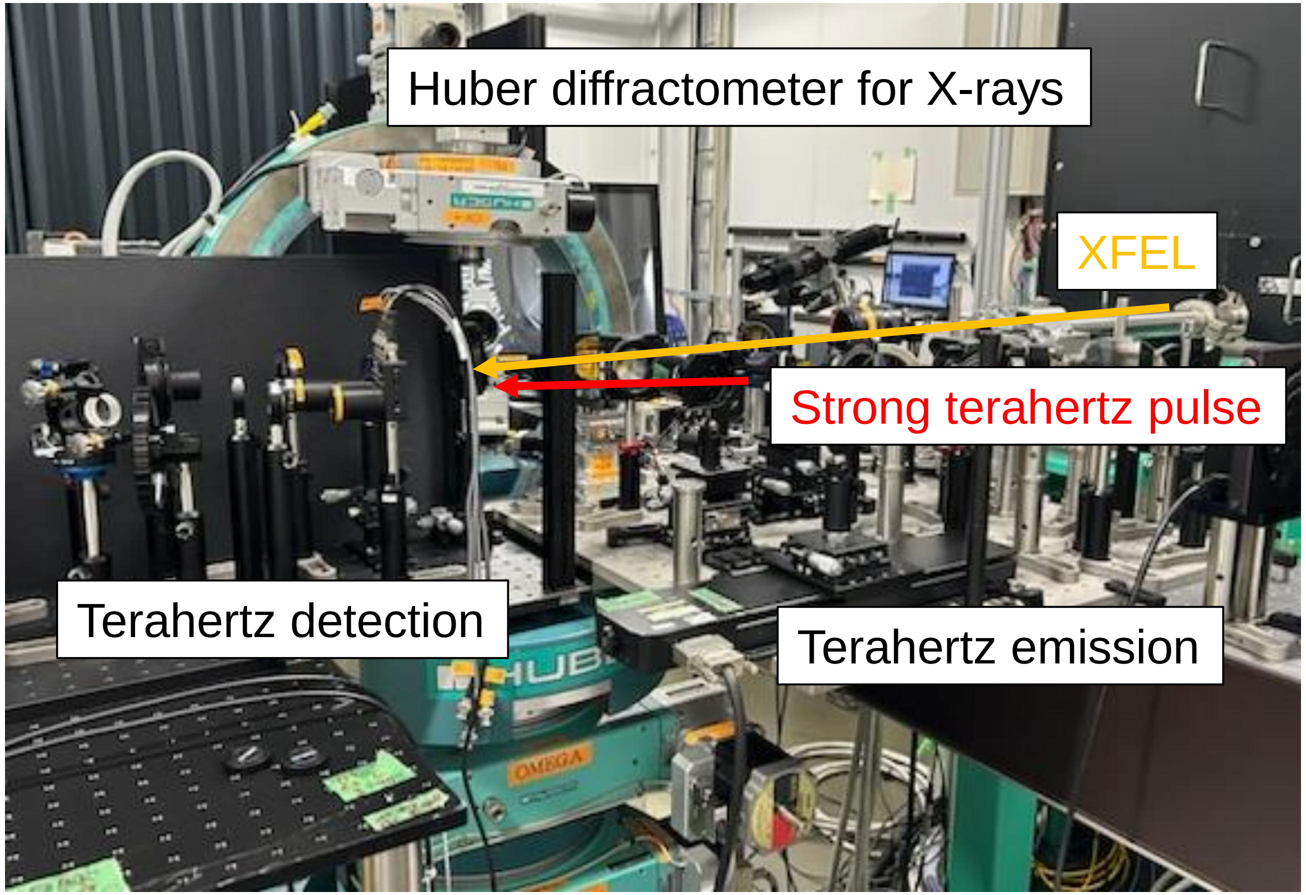


Strong terahertz pump system

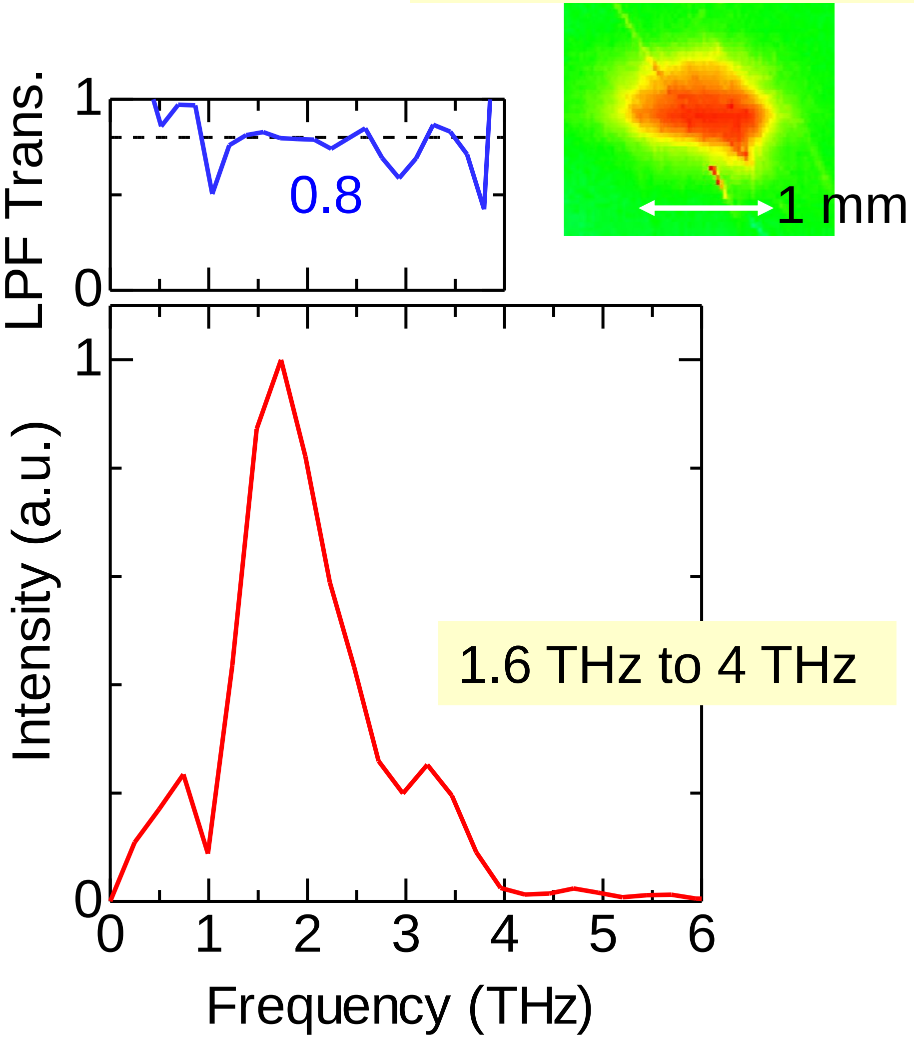


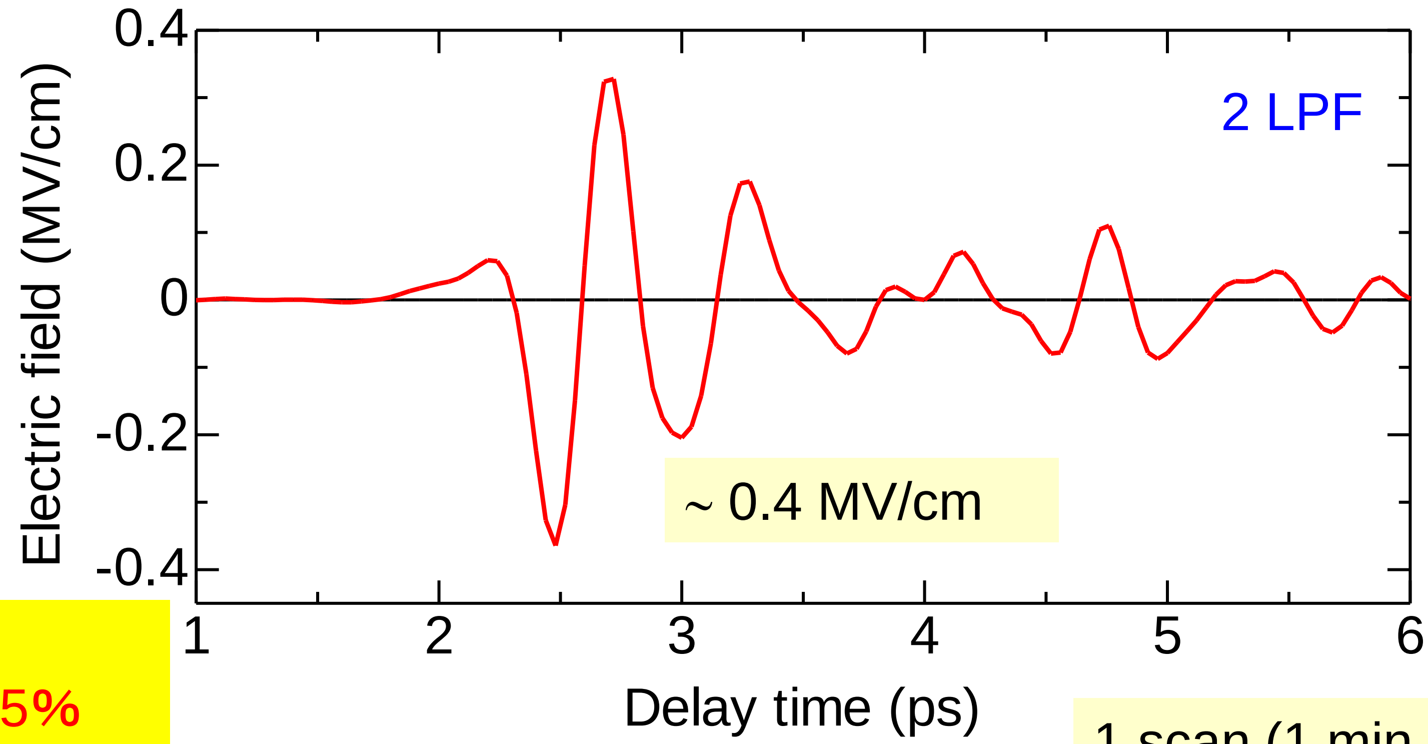
Strong terahertz electric field for pumping pulse

Emission
DSTMS (4-N,N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate)
Exc. 1550 nm, 30 fs, Rep. 60 Hz
Laser power 416 μJ
Φ ~ 4 mm (non-focus) (~827 μJ/cm^2)

Balanced Det.
GaP 100 μm
Gated 800 nm
30 fs

Spot size
~ 1.5 mm × 1.3 mm





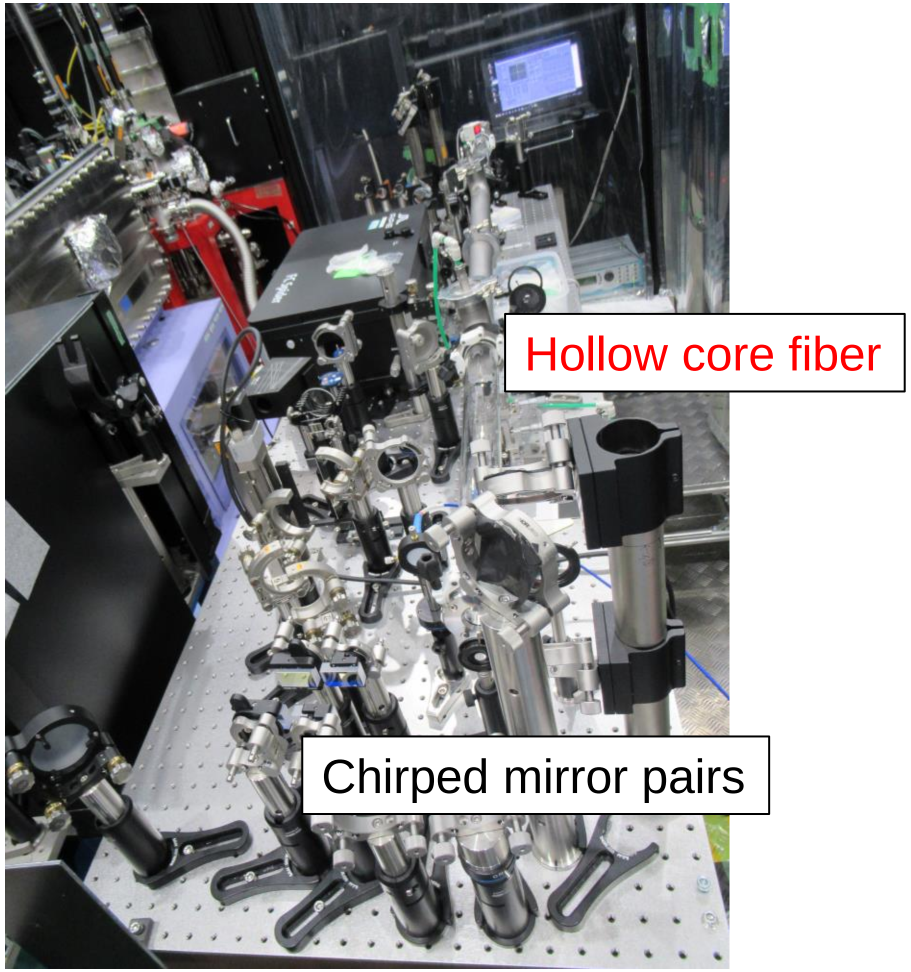
QE
0.15%
ML 0.55

1 scan (1 min.)

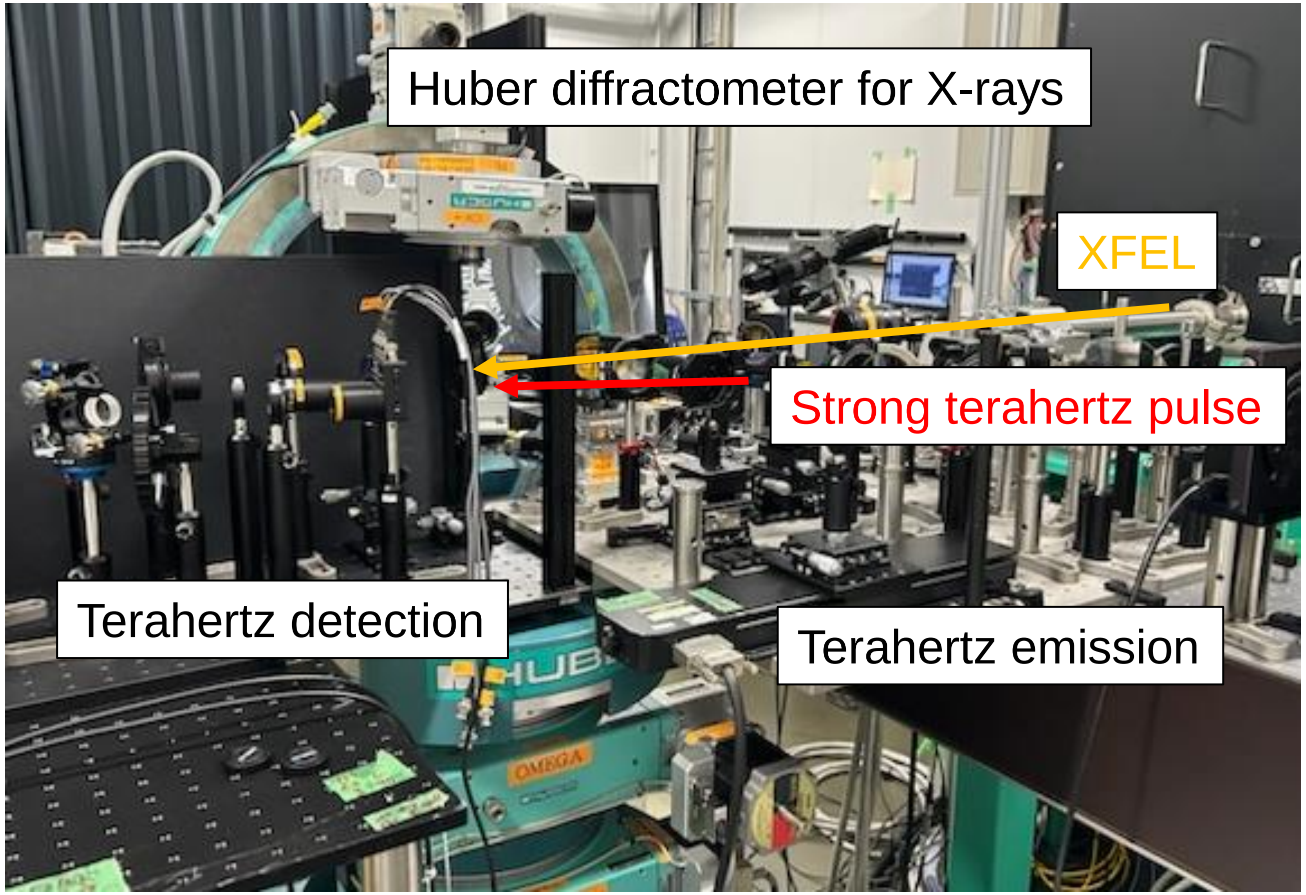
Summary

In this term, we developed two pumping systems combined with XFEL

8 fs 800 nm laser pump system



Strong terahertz pump system



Research highlights

Photoinduced sequential dynamics in a halogen-bonded hybrid system by complementary ultrafast optical and electron probes

Tadashiko Ishikawa (Institute of Science Tokyo)

Photoinduced charge/spin dynamics studied by time-resolved x-ray absorption spectroscopy

Hiroki Wadati (University of Hyogo)

Terahertz enhancement of electronic-ferroelectric polarization traced by time-resolved X-ray and nonlinear optics experiments

Hirotake Itoh (Kwansei Gakuin Univ.)