Probing phonon chirality and circular lattice motion with symmetry-selective nonlinear optical spectroscopy

Yuhan Wang[1], Yuxuan Wei[1,3], Li Huang[1] and Chuanshan Tian[1,2,*]
1. Fudan University, Shanghai 200433, China *2. Yanbian University, Jilin 133002, China*
3. University of California, California 94720, United States

Truly chiral phonons are lattice eigenmodes that combine broken mirror symmetry with circular atomic motion. They can mediate angular-momentum-selective interactions in quantum materials, yet resolving both their chirality and underlying circular motion remains challenging, especially in high-symmetry crystals. Here we show that symmetry-selective terahertz difference-frequency spectroscopy provides a phase- and polarization-resolved route to identifying truly chiral phonons in a tabletop experiment. Using α-quartz as a benchmark, we validate this approach by resolving phonon chirality via chiral-sensitive $\chi^{(2)}_{ijk}$ tensor elements ($i \neq j \neq k$), while vectorial field detection directly reveals a time-dependent polarization rotation arising from circular ionic motion and thus nonzero angular momentum. Applying the same protocol to tetragonal α-TeO$_2$, we isolate chiral E-mode resonances below 5 THz and verify their circular lattice motion, thereby resolving a symmetry-imposed ambiguity in chiral-phonon identification in fourfold-symmetric crystals. Our results establish symmetry-selective nonlinear terahertz spectroscopy as a general route to identify truly chiral phonons in condensed matter systems.

I. Probing Chiral Phonons in α-Quartz

Phonon Chirality

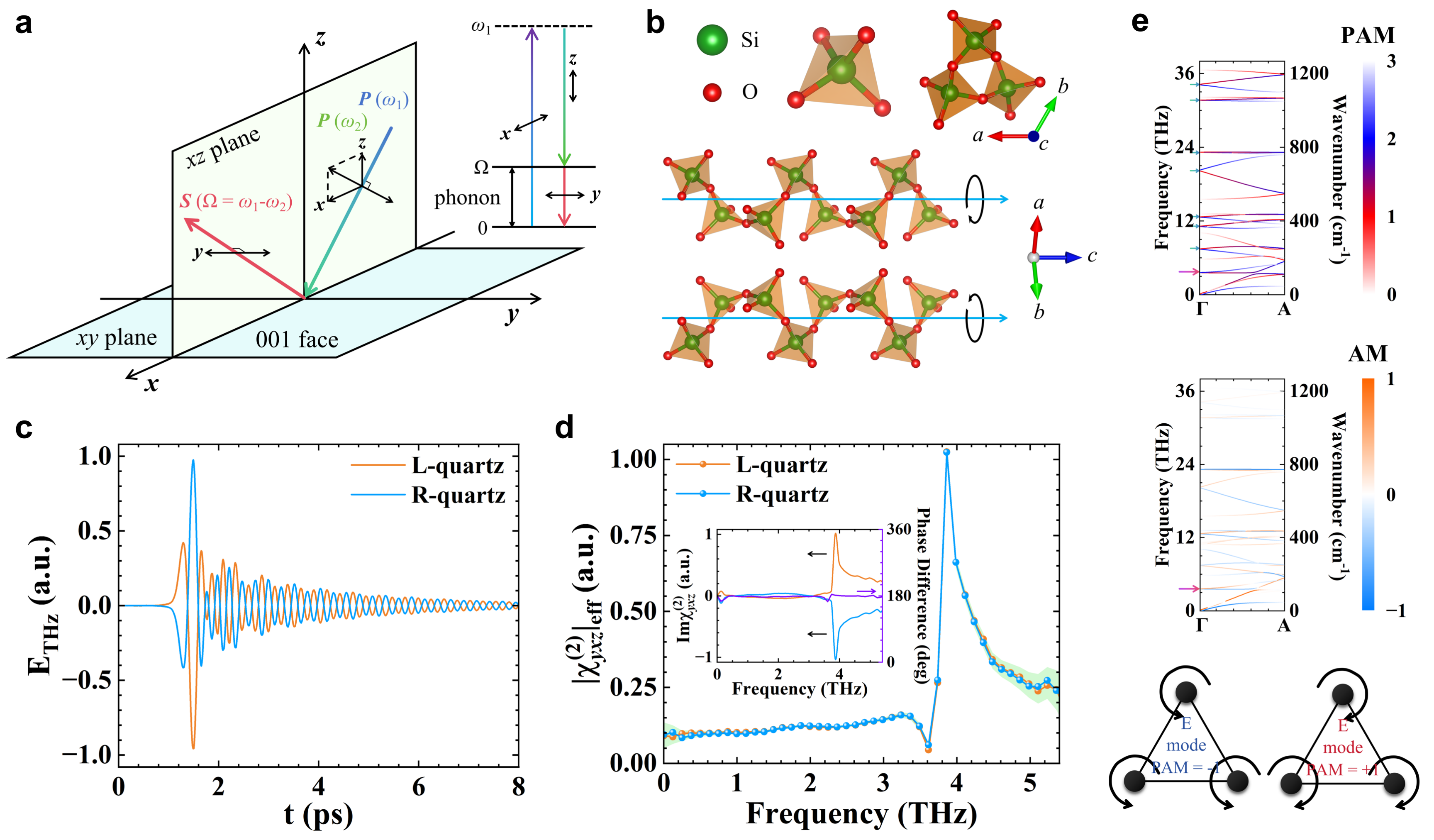

Fig. 1: Chirality-sensitive THz-DFS detection of chiral phonons in α-quartz. **a,** Schematic of the chiral terahertz difference-frequency spectroscopy (Chiral THz-DFS) geometry. The emitted terahertz field is detected in a chirality-sensitive $\chi^{(2)}_{yxz}$ channel. **b,** Crystal structure of α-quartz (SiO$_2$), showing its enantiomorphic lattice[1]. **c,** Emitted terahertz waveforms from opposite enantiomers exhibit a global sign reversal. **d,** The nonlinear susceptibility and its imaginary part (inset) show a π-phase difference between enantiomers, while the resonance frequency remains unchanged. **e,** Calculated[2] phonon dispersion[3] of α-quartz along the Γ-A direction, with color indicating pseudo-angular momentum (PAM, top) and real angular momentum (AM, middle). Only doubly degenerate E modes, indicated by red and green arrows, carry nonzero PAM and finite AM near Γ point. Bottom: schematic illustration of circular motion for E modes with opposite angular momentum.

Circular Lattice Motion

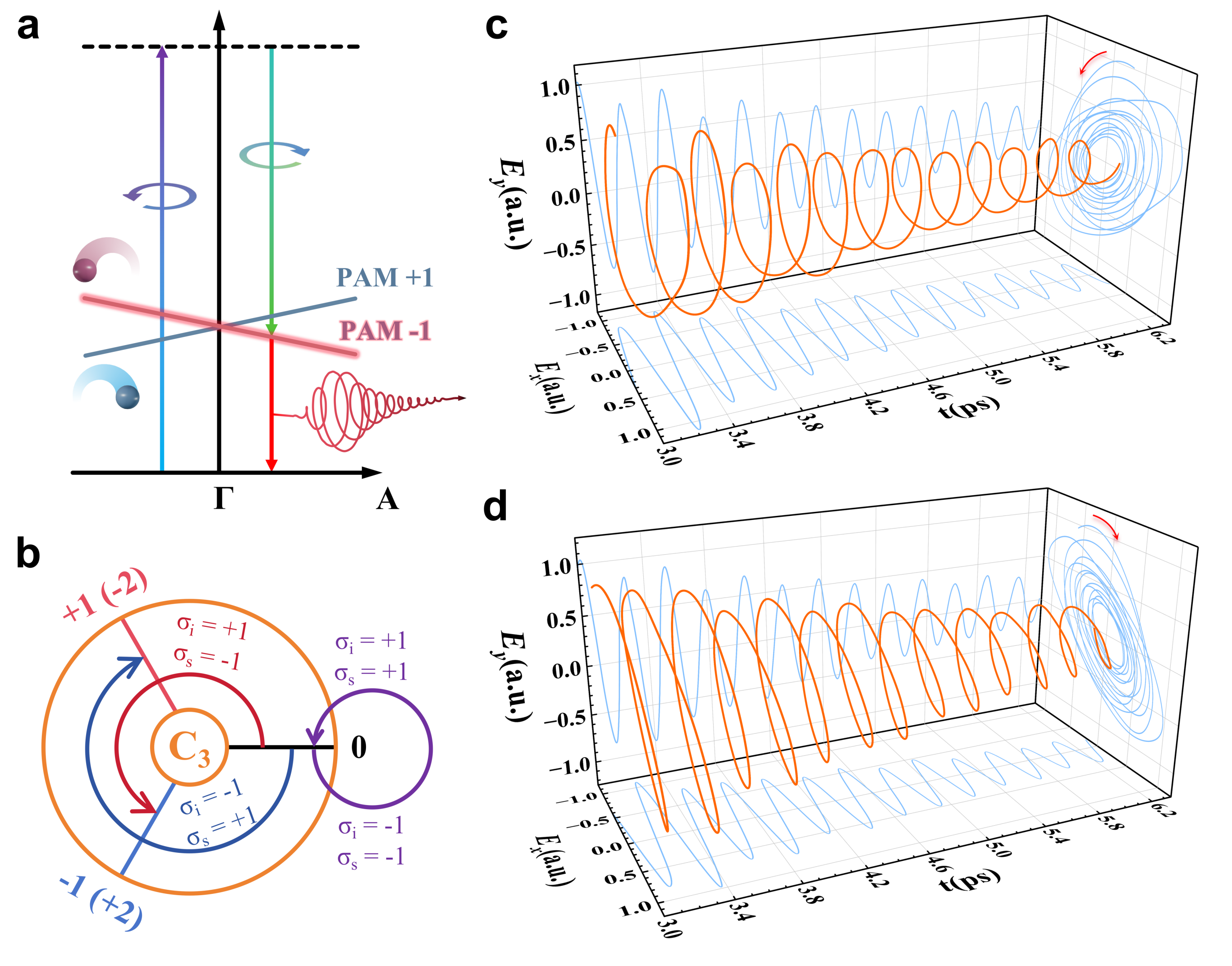

Fig. 2: Angular-momentum transfer and circular lattice motion in α-quartz. **a**, Circularly polarized difference-frequency excitation transfers pseudo-angular momentum (PAM) from light to lattice vibrations, selectively driving phonons with PAM = ±1. The resulting circular motion (non-zero AM) radiates a terahertz field whose polarization rotates in time. **b,** Selection rules under C$_3$ symmetry link the helicities of the incident fields to the PAM of the excited phonons. **c,** The emitted terahertz field exhibits a rotating polarization in time, directly visualizing coherent circular lattice motion. **d,** Reversing the pump helicities inverts the rotation sense of the terahertz polarization, evidencing controlled reversal of phonon angular momentum.

II. Chirality and Circular lattice motion in α-TeO2

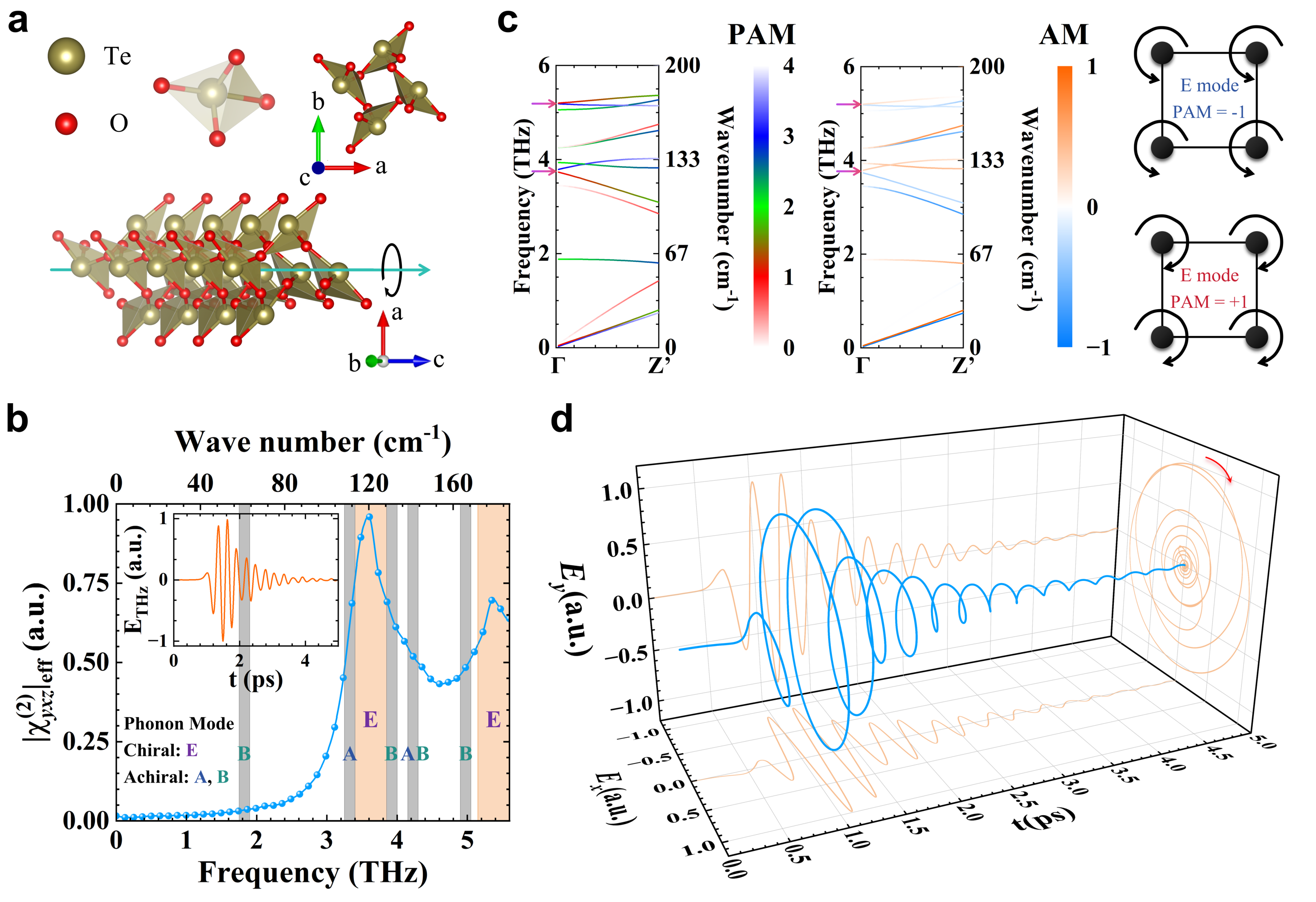

Fig. 3: Symmetry-selective identification of chiral phonons in tetragonal α-TeO$_2$. **a**, Crystal structure of α-TeO$_2$, showing the helical arrangement of Te-O tetrahedra in a tetragonal (C$_4$) lattice. **b**, Time (inset) and frequency domain THz-DFS in a chiral-sensitive channel. Only E-symmetry phonons appear, while A- and B-type (achiral) modes remain silent. **c**, Calculated phonon dispersion with PAM and AM. Only doubly degenerate E modes carry PAM = ±1 and finite AM. **d,** The emitted terahertz field exhibits a rotating polarization in time, demonstrating coherent circular lattice motion.

III. Chiral Phonon Modes in C$_3$ and C$_4$ crystal

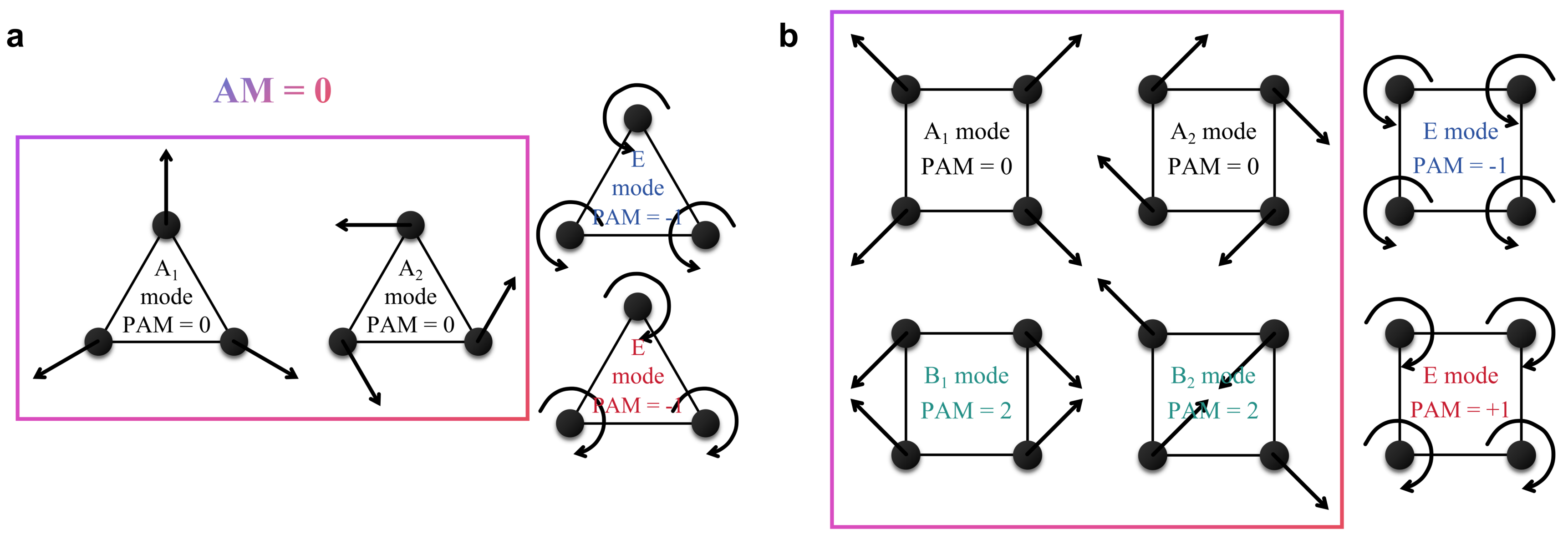

Fig. 4: Schematic illustration of motion for different modes. a-b, Schematic illustration of motion for different modes in C$_3$ (a) and C$_4$ (b) crystal, the highlighted area is AM = 0 mode.

IV. Conclusion

We establish terahertz difference-frequency time-domain spectroscopy as a symmetry-selective, phase- and polarization-resolved route for identifying truly chiral phonons. The key advance is that a single optical platform unifies two complementary observables: an enantiomer-odd phase criterion that provides a stringent fingerprint of phonon chirality, and a polarization-resolved vector-field readout that directly reveals real-space circular lattice motion and thus angular momentum (AM). By combining these two criteria, the method distinguishes chirality from angular-momentum-carrying phonons and resolves a long-standing challenge in identifying chiral phonons in high-symmetry crystals. Because the response is mediated by electric-dipole-allowed lattice excitations, the approach further offers high sensitivity in the terahertz regime. More broadly, it establishes a general framework for driving and quantifying helical lattice motion, and opens new opportunities for exploring how lattice angular momentum couples to electronic, magnetic and topological excitations in quantum materials.

Reference:

1. Momma, K. & Izumi, F. Journal of Applied Crystallography 44, 1272-1276 (2011).
2. Verstraete, M. J. et al. The Journal of Chemical Physics 163 (2025).
3. Yang, Y. et al. Nature Physics (2026).