

Sensing Coherent Phonon Dynamics in Solids with Delayed Even Harmonics

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In a pump–probe setup with variable delay, we theoretically investigate how even harmonics reveal coherent phonon dynamics. If pump and probe pulses overlap temporally, the spatial interference effect resulting from a non-coaxial pump-probe setup suppresses harmonic yields.

At longer delays, odd-harmonic yields oscillate in phase at the optical phonon frequency, whereas even harmonics exhibit order-dependent phase-shifted oscillations. We identify a responsive range of even harmonic orders, in which the delay of yield oscillations is highly sensitive to subtle features of phonon dynamics and electron–electron interactions. Hence, even harmonics can serve as a sensor for microscopic effects in systems with dynamically broken inversion symmetry.