Two-phonon quantum coherences in InSb observed by two-dimensional three-pulse THz spectroscopy

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Two-phonon quantum coherences are observed in the narrow-gap semiconductor InSb by two-dimensional terahertz spectroscopy using three nonresonant THz pulses. The two-phonon signals originate from impulsive excitation in the nonperturbative regime of light-matter interaction mediated by the large interband dipole moment.

The fundamental laws of quantum mechanics allow for coherent superpositions of quantum states as solutions. Much work has been done on one-quantum coherences, generated, e.g., by the resonant absorption of light between the ground and excited state of a quantum system. In contrast, much less is known about two-quantum coherences using nonresonant excitation. Prominent examples of two-quantum excitations are phonon combination tones in solids. In the narrow-gap semiconductor InSb such combination modes have been observed both in infrared absorption [1] and in second-order Raman scattering [2], but their coherent dynamics has not been studied so far.

In our experiment we use three pulses with a center frequency of 20 THz generated by optical rectification from the output of a femtosecond Ti:sapphire laser system. Their electric fields as

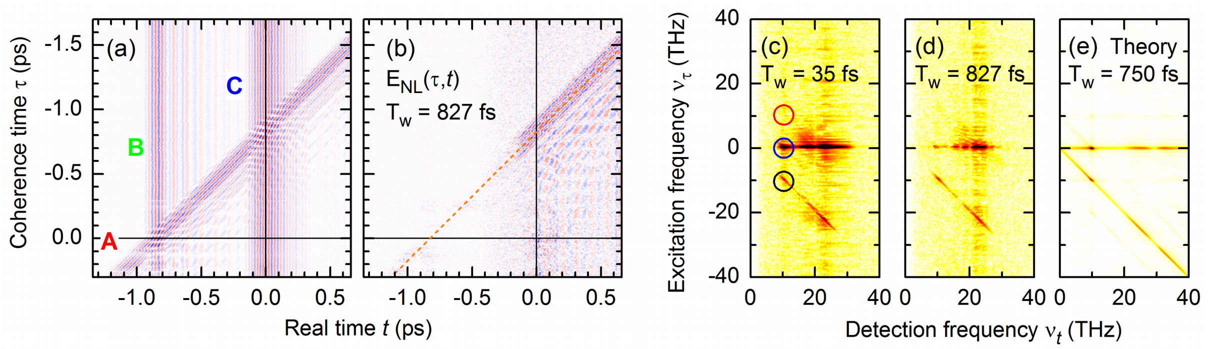

Fig.1 (a) Electric field transients of the three THz pulses A, B, and C for a waiting time T_w = 827 fs as a function of real time t and coherence time τ. (b) Corresponding nonlinear signal. (c), (d) Two-dimensional Fourier transforms of the nonlinear signal for waiting times of (c) 35 and (d) 827 fs. (e) Theoretical result for T_w = 750 fs.

functions of real time t and of the delays between the pulses, the coherence time τ and the waiting time T_w, are measured by electro-optic sampling [Fig. 1(a)]. The nonlinear signal [Fig. 1(b)] is determined from measuring all possible combinations of the three pulses [3]. The Fourier transforms [Figs. 1(c),(d), circles] show strong nonlinear signals at the detection frequency ν_t = 10 THz, the frequency of the two-phonon resonance in InSb, although the incident pulses have nearly no intensity at this frequency. As a theoretical calculation [Fig. 1(e)] shows, at our field amplitude of 50 kV/cm we are in the nonperturbative limit of light-matter interaction, so that higher-order contributions in the driving fields strongly dominate over the linear two-phonon response.

[1] S. J. Fray *et al.*, Proc. Phys. Soc. **76**, 939 (1960); E. S. Koteles *et al.*, PRB **9**, 572 (1974).
[2] W. Kiefer *et al.*, PRB **12**, 2346 (1975).
[3] W. Kuehn *et al.*, JPCB **115**, 5448 (2011); PRL **107**, 067401 (2011).