Journal Club: Two-Phonon Quantum Coherences in Indium Antimonide Studied by Nonlinear Two-Dimensional Terahertz Spectroscopy

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May 30, 2016

Two-Phonon Quantum Coherences in Indium Antimonide Studied by Nonlinear Two-Dimensional Terahertz Spectroscopy

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(Received 25 January 2016; published 28 April 2016)

We report the first observation of two-phonon quantum coherences in a semiconductor. Two-dimensional terahertz (THz) spectra recorded with a sequence of three THz pulses display strong two-phonon signals, clearly distinguished from signals due to interband two-photon absorption and electron tunneling. The two-phonon coherences originate from impulsive off-resonant excitation in the nonperturbative regime of light-matter interaction. A theoretical analysis provides the relevant Liouville pathways, showing that nonlinear interactions using the large interband dipole moment generate stronger two-phonon excitations than linear interactions.

DOI: 10.1103/PhysRevLett.116.177401

Phase-coherent superpositions of the wave functions of different quantum states represent a fundamental elementary excitation of matter. In a basic generation scheme, an optical field interacting resonantly with the transition dipole between two levels induces a quantum coherence connected with a coherent optical polarization. Such one-quantum coherences have been generated, optically

In this Letter, we report the first time-resolved study of two-phonon coherences. We apply a novel method of nonlinear two-dimensional (2D) terahertz (THz) spectroscopy t*o measure the coherent THz emission of two-phonon excitations in bulk InSb in amplitude and phase and identify the relevant nonlinear interaction mechanisms.

Our experiments are based on phase-resolved 2D col-

Quantum phononics

Quantum phononics:

- squeezed phonons
- entangled phonons

Some references:

- M. Artoni and J. L. Birman, Opt. Commun. **104**, 319 (1994).
- S. L. Johnson *et al*, Phys. Rev. Lett. **102**, 175503 (2009).
- J. Hu, O. V. Misochko, and K. G. Nakamura, Phys. Rev. B **84**, 224304 (2011).
- K. C. Lee *et al*, Nat. Photonics **6**, 41 (2012).
- O. V. Misochko, Usp. Fiz. Nauk **183**, 917 (2013) (Sov. Phys. Usp. **56**, 868 (2013)).

Selected for a Viewpoint in *Physics*
PRL **102,** 175503 (2009) PHYSICAL REVIEW LETTERS week ending 1 MAY 2009

Directly Observing Squeezed Phonon States with Femtosecond X-Ray Diffraction

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(Received 13 December 2008; published 27 April 2009)

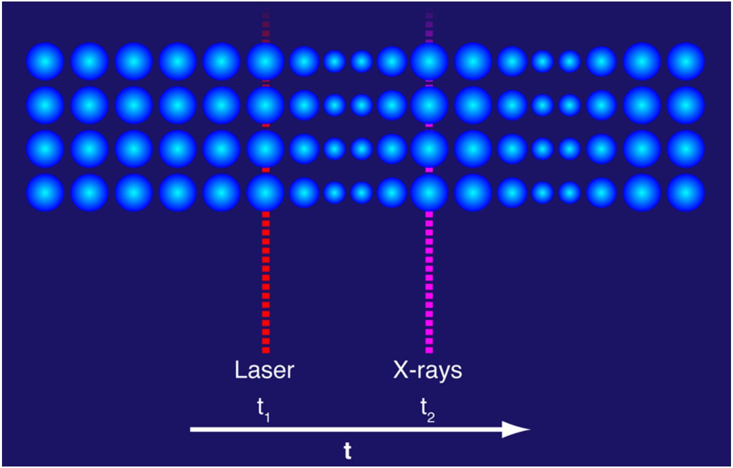
Squeezed states are quantum states of a harmonic oscillator in which the variance of two conjugate variables each oscillate out of phase. Ultrafast optical excitation of crystals can create squeezed phonon states, where the variance of the atomic displacements oscillates due to a sudden change in the interatomic bonding strength. With femtosecond x-ray diffraction we measure squeezing oscillations in bismuth and conclude that they are consistent with a model in which electronic excitation softens all phonon modes by a constant scaling factor.

DOI: 10.1103/PhysRevLett.102.175503 PACS numbers: 63.20.K−, 61.80.Ba, 78.47.J−

Squeezed states, where the variance of two conjugate variables oscillate out of phase in time, are a centerpiece of modern quantum optics [1,2]. The physics of squeezing is not limited to the electromagnetic field, however, and indeed there has been much interest in the squeezing of

factor, is sensitive to statistical correlations among phonon coordinates via $\langle \hat{Q}_{\mathbf{k}s} \hat{Q}_{-\mathbf{k}s'} \rangle$. For the case $s' = s$ this is a measurement of the coordinate variance, provided $\langle \hat{Q}_{\mathbf{k}s} \rangle = \langle \hat{Q}_{-\mathbf{k}s} \rangle = 0$. Terms with $s' \neq s$ correspond to covariances associated with "combination modes" [6].

Squeezed phonon states in a crystal of bismuth



Coherent oscillating states in matter

Video:

http://phys.org/news/2016-05-quantum-swinga-pendulum.html

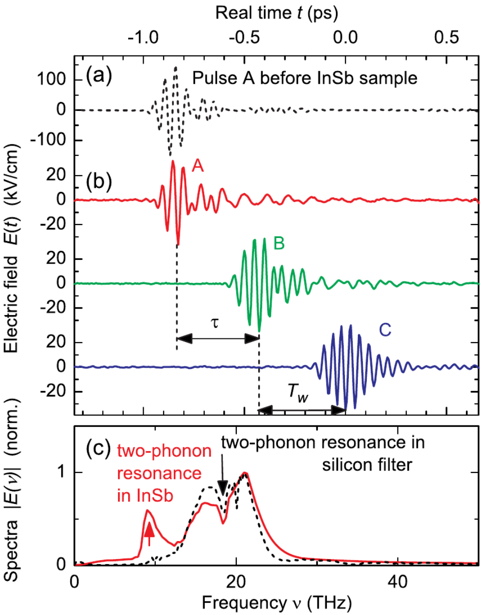
Experiment

- A 70 μm thick (100)-oriented InSb single crystal with a low n-type doping at room temperature
- A sequence of three THz pulses interacting with the sample
- The pulses incident on and transmitted through the sample and the THz emission from the sample are detected by phase-resolved electro-optic sampling

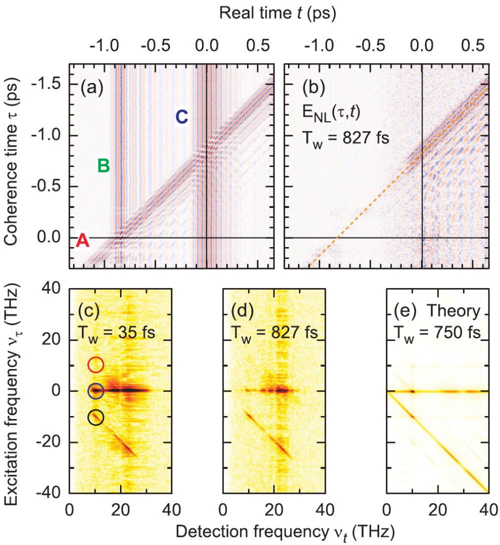
Experiment

- The signal contains the linear response and one-, two-, and three-pulse nonlinear responses.
- Seven transients are determined, one when all three pulses ABC interact with the sample, three for the pairs AB, BC, and CA, and three for the single pulses A, B, and C.
- The pure three-pulse nonlinear field emitted by the sample is calculated

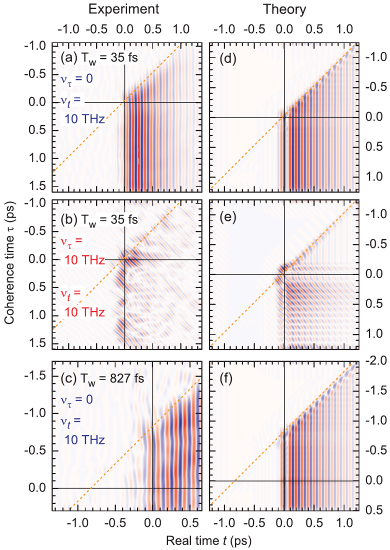
Incident and transmitted electric fields



Two-dimensional THz spectroscopy on InSb



Experimental time-domain 2D signals on different two-phonon peaks



Summary

In conclusion, we have applied three-pulse 2D THz spectroscopy to map the dynamics of two-phonon coherences in a bulk semiconductor. In the nonperturbative limit of light-matter interaction, higher-order contributions in the driving fields strongly dominate over the linear two-phonon response. Our work paves the way towards 2D THz spectroscopy of low-energy excitations like phonons and magnons, and of transitions between ground and excited states of excitons and impurities with multiple-pulse sequences.

Thank you for your attention!