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QThD6 11:45 am

Impulsive stimulated Raman excitation of coherent phonons in antimony

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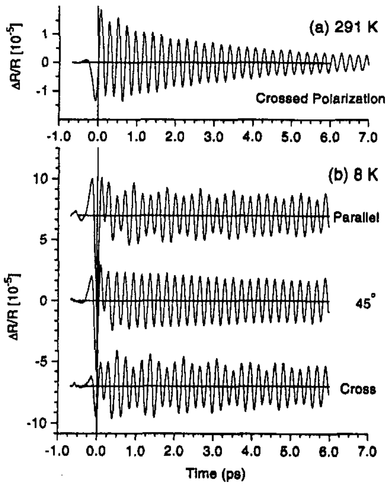
The excitation of coherent phonons in semimetals with femtosecond laser pulses has recently been investigated by several groups and accounted for by an excitation mechanism called displacive excitation of coherent phonons (DECP). DECP is considered to be different from conventional Raman processes in that it only launches fully symmetric A-phonons and that these oscillations have a cosine behavior rather than the sine behavior that is expected for impulsive stimulated raman scattering (ISRS).

In this work we re-investigated the excitation of phonons in Antimony using both time-resolved and cw-Raman techniques. The time-resolved experiments were carried out with a simple femtosecond pump-probe setup in reflection, using a self-modelocked Ti:sapphire laser at 810 nm as the pulse source. Our results agree with previous studies in that the response at room temperature is dominated by the 150 cm^{-1} A_g-mode, whereas contributions from the E_g-phonon at 112 cm^{-1} are negligible. This is illustrated in Fig. 1a, which shows the oscillating contribution to the transient reflection at room temperature after subtraction of a slowly varying background. The data can be fitted with a single-frequency damped oscillation. Below 200 K however, the E_g-phonon becomes readily observable and causes a beat structure in the transient, as can be seen in Fig. 1b. Experimental results at 8 K are shown for different polarization angles, ϕ, between the pump and probe pulses. While the fully symmetric A_g-phonon is independent of ϕ, the amplitude of the E_g-component exhibits a $\cos(2\phi)$ behavior. These findings are in agreement with Raman selection rules.



QThD6 Fig. 1 Oscillatory part of femtosecond ISRS-experiments on Sb. (a) At 300 K only the A_g-phonon mode is visible. (b) At 8 K, the E_g-phonon causes a beating structure that changes with angle between the pump and probe polarizations.

Our experiments show that the frequency, linewidth, and polarization selection rules for both phonons are consistent with results found in spontaneous Raman scattering obtained with the same excitation wavelength. We developed a theory that relates DECP to ISRS by considering a complex Raman tensor. The limiting case of a purely real, nonresonant, or complex, strongly resonant, Raman tensor then corresponds to ISRS or DECP, respectively. The phase of the excited phonons then depends on the frequency dependence of the Raman tensor. The theory predicts that the relative ratio of the intensities of the E_g and A_g modes need not be the same between spontaneous and impulsive Raman scattering, as observed in our experiments.

QThE 10:30 am
Room B2

Self-Trapping in Quadratic Media and Instabilities

George I. Stegeman, *University of Central Florida, Presider*

QThE1 (Invited) 10:30 am

Trapping of light beams and formation of spatial solitary waves in quadratic nonlinear media

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Self-focusing and self-trapping of light have been investigated since the early days of nonlinear optics.[1] Interest in this field has been maintained by the fascinating range of new phenomena encountered and their potential applications. Until very recently such effects have been pursued by use of the optical Kerr effect in third-order, or cubic, nonlinear media and the photorefractive effect. However, self-trapping of light also occurs in quadratic nonlinear media and both temporal and spatial solitons form through the mutual focusing of the waves parametrically interacting in the medium.

Self-focusing effects in quadratic nonlinear processes were long known to be possible under specific conditions, but the full extent of their implications was not fully appreciated until recently, after the so-called "cascading of second-order nonlinearities" was identified as a potential new approach for the control of light by light.[2] A remarkable exception is the work of Karamzin and Sukhorukov 20 years ago,[3] who investigated the mutual focusing of beams in parametric processes and identified its implications for the formation of solitons. Recently, spatial soliton propagation has been observed experimentally in second-harmonic generation settings in bulk potassium titanyl phosphate (KTP)[4] and in planar waveguides made of lithium niobate ($\mathrm{LiNbO_3}$).[5]

In this paper we report the outcome of our comprehensive investigations to study the dynamics of the beam trapping in both bulk crystals and planar waveguides made of quadratic nonlinear media in second-harmonic generation configurations. We address and discuss the suitable experimental conditions required to form spatial solitary waves in critical phase-matching and quasi-phase-matching settings.

We discuss the properties and stability of the relevant known families of unidimensional and two-dimensional stationary solitary waves, their evolution in the presence of small perturbations and losses, and we explore the dynamics of the excitation of spatial solitary waves under different material and excitation conditions. We show how the mutual trapping occurs in a wide variety of conditions, not necessarily close to those given by the stationary solitary waves solutions of the governing equations. Solitary waves emerge with inputs that fall very far from those solutions indeed. We specifically show the dynamics of the beam trapping with only the fundamental beam at the input face of the nonlinear crystal. We discuss the mutual beam dragging of the fundamental and second harmonic waves that occurs in the presence of linear walk-off or spatial phase modulation of the input beams, beam steering, collisions, and interaction of solitary waves, break-up and self-deflection of beams, and their potential applications.

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