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High amplitude coherent phonons and their nonlinear behavior

We use inelastic scattering of ultrashort visible and hard X-ray pulses as a probe of the transient occupation of phonon modes constituting large amplitude strain-wavepackets. These Brillouin scattering experiments permit the time-resolved observation of the phonon dynamics including the phonon damping and nonlinear interaction. We use a variety of excitation schemes to synthesize high amplitude phonon wavepackets. By pumping a nanometric metal layer with one or multiple pulses we can shape the imprinted phonon spectrum. Such high strain fields can lead to several anharmonic phenomena like sum and difference frequency mixing, which could be termed "nonlinear phononics"in analogy to nonlinear optics. These nonlinear phenomena depend strictly on the excited phonon spectra. For high amplitude single cycle phonon pulses we have observed the self-steepening of the pulse fronts.[1] Spectrally narrow phonon wavepackets exhibit difference- and sum-frequency generation. We quantitatively model the dynamics by coupled anharmonic oscillators. New measurements of high amplitude strain pulse propagation in SrTiO3 at the phase transition of 105K reveal a superelastic behavior which is connected to the coupling of the high pressure sound pulse to the motion of domain walls. We think our new insights in the field of nonlinear phononics could help to establish new ultrafast experiments in which the direct optical excitation of a material by an ultrashort laser pulse is replaced by a highly intense ultrashort pressure pulse which excites the material triggering the dynamics by electronic excitation.

[1] Bojahr et al, PRB 2012 86(144306)

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