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Coherent spectroscopy of high wave-vector phonons

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Hard x-ray free-electron lasers have emerged as an invaluable tool for studying materials dynamics near and far from equilibrium. Advanced nonlinear x-ray methods will allow us to greatly expand on our ability to understand and control materials properties at the atomic-scale. In this talk I will present results of all time-domain x-ray scattering-based spectroscopies to study transient lattice dynamics in photo-excited materials spanning the Brillouin zone using both optical and x-ray excitation. Ultrafast optical excitation typically produces broad-band coherences in the mean-square displacements of the ions that we subsequently resolve in time and momentum via femtosecond diffuse scattering [1,2]. I will briefly discuss the mechanism of excitation [3] whereby we can measure the excited state phonon-dispersion [4] and momentum-resolved anharmonic decay channels [5] using photo-excited bismuth as an example. We have also shown that to some extent selective-excitation using optical pulses can be used to control which modes are excited [3,6]. Finally, I will discuss progress towards arbitrary x-ray selective excitation using atomic-scale transient gratings including preliminary results of x-ray pump, x-ray probe experiments on cubic perovskites.
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