The Mechanism of Coherent Phonon Generation

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Coherent acoustic phonons in solids can be generated by impulsive optical excitation through ultrafast heating of lattice and electrons. The former is the thermal lattice expansion related to anharmonicity of ion-ion interaction. The latter is the thermal pressure of free electrons. Previous studies using fs optical probes indicate that both driving forces contribute the phonon generation. However, these optical measurements are indirect probes of lattice motions. Here, we report the study of coherent acoustic phonon generation mechanism by directly measuring the associated structural dynamics with femtosecond electron diffraction (FED). FED provides a quantitative measurement of the temporal evolution of both coherent and thermal lattice motions with sub milli-ångström spatial resolution and on the sub-ps timescale. The damped single-mode breathing motion of 20-nm-thick Al film along the surface normal was recorded as the coherent oscillation of Bragg peak position with a period 6.4 ps, as determined by the 1-D standing wave condition. The lattice temperature (thermal motion) evolution with a time constant of 600 fs was measured by following the associated Bragg peak intensity attenuation. By fitting these data with the differential equation of a harmonic oscillator using a driving term including both lattice and electron heating, we find that the electron pressure contributes more significantly than the lattice heating for the first half cycle of the lattice vibration. These results provid a direct and clear evidence of the non-thermal generation mechanism of coherent acoustic phonons.

Keywords: lattice dynamics, time-resolved diffraction, electron phonon coupling