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Coherent excitation and control of surface phonons

YOSHIYASU MATSUMOTO, Institute for Molecular Science

The excitation and control of nuclear wavepackets using tailored laser pulses have attracted a lot of interest recently and being realized mainly in gas-phase molecules. In contrast, there have been little studies on the coherent excitation and control for adsorbates particularly on metal surfaces. This is because dephasing is substantially rapid on metal surfaces due to efficient couplings between adsorbates and metals. Recently, we have demonstrated the time-domain observation of nuclear wavepacket dynamics of monolayer adsorbate by femtosecond time-resolved second harmonic generation (TRSHG). When metal surfaces covered with alkali metal atoms are irradiated by ultrafast laser pulses, coherent surface phonon modes are excited. The formation and dissipation processes of coherent surface phonons are probed by time-resolved second harmonic generation. SHG signal intensities are enhanced by alkali atoms adsorption by various resonant transitions in the adsorbate-substrate system. However, not all resonant electronic transitions lead to the generation of coherent stretching vibrations of alkali atoms. The measurements of TRSHG traces as a function of the excitation photon energy at a fixed alkali coverage indicate that resonant transitions between adsorbate-induced surface states is responsible for the coherent vibrational motions. By carefully examining the Cs coverage dependence of the TRSHG waveform, we found that TR-SHG traces show beating structures. This indicates that the oscillatory TR-SHG traces are contributed by at least two kinds of coherent surface phonon modes: the Cs-Pt stretching mode (2.3 THz) and the Rayleigh phonon mode (2.6 or 2.9 THz, depending on the Cs coverage). We used fs pulse trains with the repetition frequencies of 2.0 - 2.9 THz that are synthesized by using a spatial-light modulator as an excitation source for the coherent phonons. By tuning the pulse train frequency, we succeed in the selective excitation of a coherent phonon mode.