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Title:Adsorbate-localized versus substrate-mediated excitation mechanisms for generation of coherent Cs-Cu stretching vibration at Cu(111)
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Abstract: Coherent Cs–Cu stretching vibration at a Cu(111) surface covered with a full monolayer of Cs is observed by using time-resolved second harmonic generation spectroscopy, and its generation mechanisms and dynamics are simulated theoretically. While the irradiation with ultrafast pulses at both 400 and 800 nm generate the coherent Cs–Cu stretching vibration at a frequency of 1.8 THz (60 cm–1), they lead to two distinctively different features: the initial phase and the pump fluence dependence of the initial amplitude of coherent oscillation. At 400 nm excitation, the coherent oscillation is nearly cosine-like with respect to the pump pulse and the initial amplitude increases linearly with pump fluence. In contrast, at 800 nm excitation, the coherent oscillation is sine-like and the amplitude is saturated at high fluence. These features are successfully simulated by assuming that the coherent vibration is generated by two different electronic transitions: substrate d-band excitation at 400 nm and the quasi-resonant excitation between adsorbate-localized bands at 800 nm, i.e., possibly from an alkali-induced quantum well state to an unoccupied state originating in Cs 5d bands or the third image potential state.