Nonlinear resonant Auger spectra and transient x-ray absorption spectra in CO using an x-ray pump-control scheme

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Synopsis In this work we propose nonlinear femtosecond x-ray pump-probe spectroscopy to study the vibrational dynamics of a core-excited molecular state and discuss numerical results in CO. A femtosecond pump resonantly excites the carbon core-excited 1s-1π* state of the CO molecule. A second strong probe (control) pulse is applied at variable delay and is resonantly coupled to a valence excited state of the molecule (Fig. 1). The strong nonlinear coupling of the control pulse induces Rabi flopping between the two electronic states. During this process, a vibrational wave packet in the core-excited state is created, which can be effectively manipulated by changing the time delay between pump and control pulses. We present an analysis of the resonant Auger electron spectrum (Fig. 2) and the transient absorption or emission spectrum (Fig. 3) on the pump transition and discuss their information content for reconstruction of the vibrational wave packet [1].

FIG. 1. Potential-energy curves for the four involved electronic states and the schematics of the present pump-probe scheme. The optical nonlinear interaction is caused by the strong probe (control) pulse on the transition between states R and F.

FIG. 2. The resonant Auger spectra for different Δt for a strong control pulse of ω2 = 277.6 eV and Ω2 = 0.05 a.u.

Fig. 3. X-ray transient absorption or emission spectra of CO for the pump field around 287.4 eV. Shown is a comparison for weak and strong control pulses. The absorption or emission peaks for the case of Δt = −2 fs are labeled as vertical dashed lines.

References

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