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Coherent control of ionic yields after tailored multiphoton excitation in atoms and molecules

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When excited by a pulse sequence generated by sinusoidal spectral phase modulation a resonantly driven atom or molecule may experience the control mechanism SPODS. The Selective Population of Dressed States enables selective and efficient transient as well as final state control by precisely tailoring the phase of the laser field to the induced dynamics and making use of the dynamical Stark effect. The same pulse sequences can be employed to excite atoms or molecules non-resonantly. In several species we could observe a strong modulation of the ionic yield with respect to the relative interpulse optical phase. We used the prototype systems xenon as well as isopropyl alcohol, ethanol and acetone and excited these substances with the shaped femtosecond laser pulses of constant pulse energy centred at 785 nm. The first absorption band of all examined species lies well above 3 eV, it is therefore driven by at least 2 infrared photons, 6 in the case of xenon. Surprisingly, already a simple level approach is sufficient to model the experimental results. We use our simulations to analyze the underlying population dynamics. Our calculations indicate them as a general behaviour of systems undergoing multiphoton excitation followed by resonantly driven Rabi-oscillations among excited electronic states.

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