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Ultracold molecule formation using shaped femtosecond pulses

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We present experiments on the formation of ultracold molecules by femtosecond laser pulses in a pump-probe scheme [1]. Previous experiments [2,3] have only demonstrated the dissociation of molecules by femtosecond pulses, whereas now active photoassociation is observed. A shaped pump pulse excites a collision pair of laser cooled rubidium atoms to a bound molecular state below the $5s5p_{1/2}$ asymptote, from where the molecule is transferred to the molecular ionic state by a probe pulse a few picoseconds later. A femtosecond pulse shaper is used to apply a sharp low pass filter to the pump pulse spectrum with a cutoff frequency a few wavenumbers below the atomic D1 resonance. The photoassociation signal shows oscillatory dynamics between the electronic molecular states coupled by the pump pulse, resulting from the pump pulse's high electric field strength and its specific spectral shape close to the molecular dissociation limit. Applying linear chirps to the pump pulse results in a higher photoassociation efficiency for positive than for negative chirps, indicating the propagation of a molecular wavepacket in the excited state potential [4]. Simulations of the pump pulse excitation have been performed by numerically solving the time dependent Schrödinger equation using a mapped-fourier-grid-hamiltonian algorithm and are in excellent agreement with the experimental results.

References:
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