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Direct Frequency comb control of molecular dynamics

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The advent of the ultrafast frequency comb, emitted from phase stabilized mode-locked laser oscillators, revolutionized the field of precision optical spectroscopy [1], by supplying a 'ruler' of known, equally-spaced reference frequencies. Traditionally, one would measure the exact frequency of a CW laser field by comparing it against this static reference. An alternative approach however, is that of direct frequency comb spectroscopy, where the comb light is directly used as the spectroscopic excitation source [2]. Here, the excitation field is a phase coherent train of short pulses, where selective excitation is achieved by the coherent interference in the excited medium of many such pulses, just like in a generalized Ramsey experiment. The specific quantum transition to be excited is then selected via control over the comb parameters (repetition rate and carrier-envelope phase).

The ability to use the comb to measure quantum transitions precisely indicates that precise control over the quantum dynamics governing these transitions is also possible, as was indeed demonstrated in the for cold Rb atoms in the perturbative regime [3]. Here we describe several schemes [4,5] that make use a coherent train of weak pump-dump pairs of shaped ultrashort pulses to precisely and efficiently control molecular Raman transitions. The use of weak pulses permits an analytic description within the framework of coherent control in the perturbative regime, while coherent accumulation of many pulse pairs enables near unity transfer efficiency with a high spectral selectivity. We specifically consider application of this concept to the formation of stable, deeply bound, ultracold molecules.

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