

MAR08-2007-020352

Abstract for an Invited Paper
for the MAR08 Meeting of
the American Physical Society

Quantum control spectroscopy with multipulses

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The manipulation of molecular vibrations by laser light has been always considered as a very promising means to control chemical reaction. The coherently controlled time-dependent superposition of vibrational states may represent motion along a reaction coordinate and therefore allows for a high degree of selectivity. Pulse shapes for manipulating vibrations can be predicted to be trains of pulses with temporal spacing between the sub-pulses equal to an integer of the vibrational phase. If the manipulation of molecular vibrations with pulse trains is expected to be one of the important mechanisms on the long standing aim of mode selective chemistry, it is necessary to understand its application limits. In this work, the interaction of pulse trains with matter is discussed under the light of time-resolved nonlinear experiments and density matrix simulations. Emphasis is given to the role of electronic coherence between excited and ground-state, to the excited state population relaxation time and to the electronic resonance. In particular the lifetime of the excited state poses a challenge for the coherent control with multipulses and, thus, for the mode filtering capability in the excited state. This is investigated by applying a shaped femtosecond excitation pulse to different molecules in solution and probing the response by transient absorption, nonlinear Raman and DFWM spectroscopy. Finally, the effect of the phase of sinusoidal modulation on the envelope of the multipulse sequence and its consequences on pump-probe spectroscopy is discussed, particularly near zero delay between pump and probe pulses.