RAPID-ADIABATIC-PASSAGE CONTROL OF RO-VIBRATIONAL POPULATIONS IN POLYATOMIC MOLECULES

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We present a simple method for control of ro-vibrational populations in polyatomic molecules in the presence of inhomogeneous electric fields [1]. Cooling and trapping of heavy polar polyatomic molecules has become one of the frontier goals in high-resolution molecular spectroscopy, especially in the context of parity violation measurement in chiral compounds [2]. A key step toward reaching this goal would be development of a robust and efficient protocol for control of populations of ro-vibrational states in polyatomic, often floppy molecules. Here we demonstrate a modification of the stark-chirped rapid-adiabatic-passage technique (SCRAP) [3], designed for achieving high levels of control of ro-vibrational populations over a selected region in space. The new method employs inhomogeneous electric fields to generate space- and time-controlled Stark-shifts of energy levels in molecules. Adiabatic passage between ro-vibrational states is enabled by the pump pulse, which raises the value of the Rabi frequency. This Stark-chirped population transfer can be used in manipulation of population differences between high-field-seeking and low-field-seeking states of molecules in the Stark decelerator [4]. Appropriate timing of voltages on electric rods located along the decelerator combined with a single pump laser renders our method as potentially more efficient than traditional Stark decelerator techniques. Simulations for NH₃ show significant improvement in effectiveness of cooling, with respect to the standard ‘moving-potential’ method [5]. At the same time a high phase-space acceptance of the molecular packet is maintained.