PROBING COLLISIONAL COOLING DYNAMICS USING STIMULATED EMISSION PUMPING-HOLE FILLING SPECTROSCOPY

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Bis(2-hydroxyphenyl)methane (2HDPM) exhibits two conformations (labeled A and B) in a jet-cooled environment. The stimulated emission pumping (SEP) spectrum of conformer A reveals beautiful Franck-Condon factors up to 1200 cm⁻¹ above the zero point level (ZPL). These SEP transitions were used as a means of initiating studies of collisional cooling over a wide range of well-defined energies. SEP-hole filling (SEP-HF) spectroscopy is a triple resonance technique where cold molecules are pumped to the S₁ state, immediately dumped down to a well defined vibrational level (below the barriers to isomerization) in S₀, and then allowed to collisionally cool back to the ZPL before being probed by a third laser. To obtain the dynamics data, the distance of the SEP lasers from the nozzle orifice was varied as a means of varying the number of collisions (from about 2 to more than 100) with helium between the pump/dump and probe steps. As long as there were a sufficient number of collisions to cool population back to the ZPL, a gain was detected in the signal of the probe laser. However, once there were no longer enough collisions to remove all of the energy supplied to the molecules no gain was observed. The internal energy at which 50% of the population returned to the ZPL was determined and plotted versus the number of collisions, and one from 0-25 collisions with slopes of 13.4, 0.6, and 7.2 cm⁻¹/collision respectively. Possible reasons for these peculiar slopes and the full experimental setup will be discussed.