

MEASUREMENT OF ROTATIONAL STATE-TO-STATE RELAXATION COEFFICIENTS BY RAMAN-RAMAN DOUBLE RESONANCE. APPLICATION TO SELF-COLLISIONS IN ACETYLENE.

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We have developed a technique for the measurement of state-to-state rotational relaxation rates due to collisions in the gas phase. A single J state of the $v_2 = 1$ vibrational level of acetylene is populated by a Stimulated Raman process. After a variable delay of a few ns we record high resolution ($\sim 0.003 \text{ cm}^{-1}$) spectra of the Q-branch of the $v_2 = 2 \leftarrow v_2 = 1$ transition by a second Stimulated Raman process. The relative intensities of the lines of this Q-branch, recorded as a function of the the initially pumped J level and the number of collisions between the pump and probe stages (delay \times pressure product), allows us to obtain a set of state-to-state energy transfer coefficients by fitting the evolution of the observed populations to that predicted by a master equation with adjustable coefficients. The experimental details and first results for odd-J levels of acetylene at 150 K will be presented.