COLLISION INDUCED ELECTRONIC TRANSITIONS (CIET) IN NESTED SINGLET EXCITED STATES OF N_2: A TWO-COLOR PUMP-PROBE REMPI STUDY[a]

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Collision induced electronic transitions (CIET) occur between two or more different electronic states that lie energetically close to one another. If these electronic states are associated with different vibrational anharmonicities, dipole moments or polarizabilities, then near-resonance conditions can occur between some of the vibronic levels of these nested electronic states in different surroundings. Well-known examples of such processes include radiative and non-radiative relaxation among the triplet states of CO.
In addition, CIET plays a very important role in the processes that occur in earth's upper atmosphere, where airglow and aurora occur through excitation of atomic and molecular nitrogen and oxygen species by electrons from solar wind, photons from the sun or atom recombination.
In order to get accurate rate constants for CIET occurring in N_2 between the excited singlet states $a^1\Pi_g$ and $a'\,^1\Sigma_u^-$, we have carried out two-color pump-probe REMPI experiments. The pump laser is used to populate initially v = 1 level of the $a^1\Pi_g$ state through a two-photon excitation. The probe laser ionizes N_2 from the time-evolved electronic states through a one-photon resonance to highly excited singlet states of N_2. Our results show that direct collisional relaxation from v = 1 to v = 0 of the $a^1\Pi_g$ state is far less efficient (around 10%) than CIET to generate v = 1 of the $a'\,^1\Sigma_u^-$ state. Further details of our investigations and their relevance to atmospheric processes will be presented and discussed.

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