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Collisional and Radiative Effects in Transient sub-Doppler Hole Burning: Double Resonance Measurements in CN

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We report transient hole-burning and saturation recovery measurements in the CN radical with MHz frequency resolution and 20 ns time resolution. Narrow velocity groups of individual hyperfine levels of selected rotational states in CN ($X\ ^2\Sigma^+$) are depleted and excited ($A^2\Pi_i$) with a saturation laser and probed by a counterpropagating, frequency modulated probe beam. Recent work in our lab has used this method to measure and characterize the hyperfine splittings for a set of rotational, fine structure, and parity components of CN ($A\ ^2\Pi_i$, v=1). Extending this work, we report time and frequency dependence of the saturation signals following abrupt switching of the CW saturation beam on and off with an electro-optic amplitude modulator. Recovery of the unsaturated absorption following the turnoff of the saturation beam follows pressure-dependent kinetics, driven by collisions with the undissociated NCCN precursor with a rate coefficient of 2 x 10^{-9} cm^3 s^{-1} $molec^{-1}$. Similar recovery kinetics are observed for two-level saturation resonances, where the signal observed is a combination of *X*- and *A*-state kinetics, as well as for three-level crossover resonances, which can be chosen to probe selectively the hole-filling in the *X* state or the decay of velocity-selected *A* state radicals. The observed recovery rates are 8-10 times faster than the estimated rotationally inelastic contribution. The observed recovery rates are likely dominated by velocity-changing collisions in both X and A states, occurring with similar rates, despite the large difference in the properties of these electronic states. Transient signal risetimes following the turning on of the saturation pulse are consistent with the expected Rabi frequency. At lower pressures (~50 mTorr) and higher beam power (~200 mW), we can observe multiple Rabi cycles before collisions disrupt the coherent excitation and the transient signal reaches a steady state.

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