JET-COOLED CAVITY RING-DOWN SPECTROSCOPY OF THE $\tilde{A}^2E''$-$\tilde{X}^2A_2'$ VIBRONIC TRANSITION OF NO_3

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The three energetically lowest electronic states ($\tilde{X}\ ^2A_2'$, $\tilde{A}\ ^2E''$, and $\tilde{B}\ ^2E'$) of NO_3 are strongly coupled by vibronic interactions and have been treated in considerable detail theoretically.[b] Corresponding experimental characterization of the interaction is much less detailed. Previous experimental results primarily consist of IR measurements of vibrational transitions in the ground state.[cd] In addition, the electronically forbidden $\tilde{A}$-$\tilde{X}$ transition has been observed in ambient temperature CRDS studies.[ef] A slit-jet nozzle with a high voltage pulsed discharge has been applied to produce the NO_3 radical by dissociating the N-O bond of N_2O_5, and the jet-cooled NO_3 CRDS absorption spectrum has been successfully observed with a high-resolution laser source ($\Delta\nu\ \approx$250MHz, intrinsic resolution considering the instrumental linewidth and the residual Doppler broadening in the jet). The 4_0^1 band (parallel band) shows complex rotational structure which is presently being analyzed. The 2_0^1 band has also been measured as an example of a perpendicular band. Besides the ν_2 and ν_4 vibronic bands, the vibronically forbidden origin band (0_0^0 band) has been recorded under the same experimental conditions. The weakly observed $\tilde{A}$-$\tilde{X}$ origin band is likely either a magnetic dipole or an electric quardrupole transition.

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