

## 2C-R4WM SPECTROSCOPY OF JET COOLED NO<sub>3</sub>

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We have generated NO<sub>3</sub> from pyrolysis of N<sub>2</sub>O<sub>5</sub> following supersonic free jet expansion, and carried out two color resonant four wave mixing ( 2C-R4WM ) spectroscopy of the  $\tilde{B}^2E' - \tilde{X}^2A'_2$  electronic transition. One laser was fixed to pump NO<sub>3</sub> to a ro-vibronic level of the  $\tilde{B}$  state, and the other laser ( probe ) was scanned across two levels of the  $\tilde{X}^2A'_2$  state lying at 1051 and 1492 cm<sup>-1</sup>, the  $\nu_1$  ( $a'_1$ ) and  $\nu_3$  ( $e'$ ) fundamentals, respectively. The 2C-R4WM spectra have unexpected back-ground signal of NO<sub>3</sub> ( stray signal due to experimental set-up is also detected ) similar to laser induced fluorescence ( LIF ) excitation spectrum of the 0-0 band, although the back-ground signal was not expected in considering the 2C-R4WM scheme. Despite the back-ground interference, we have observed two peaks at 1051.61 and 1055.29 cm<sup>-1</sup> in the  $\nu_1$  region of the spectrum, and the frequencies agree with the two bands, 1051.2 and 1055.3 cm<sup>-1</sup>, of our relatively higher resolution dispersed fluorescence spectrum, the former of which has been assigned to the  $\nu_1$  fundamental. Band width of both peaks,  $\sim 0.2$  cm<sup>-1</sup>, is broader than twice the experimental spectral-resolution, 0.04 cm<sup>-1</sup> ( because this experiment is double resonance spectroscopy ), and the 1051.61 cm<sup>-1</sup> peak is attributed to a  $Q$  branch band head ( a line-like  $Q$  branch ) of the  $\nu_1$  fundamental. The other branches are suspected to be hidden in noise of the back-ground signal. The 1055.29 cm<sup>-1</sup> peak is also attributed to a  $Q$  band head. The  $\tilde{B}^2E'_{\frac{1}{2}} ( J' = \frac{3}{2}, K' = 1 ) - \tilde{X}^2A'_2 ( N'' = 1, K'' = 0 )$  ro-vibronic transition was used as the pump transition. The dump ( probe ) transition to both  $a'_1$  and  $e'$  vibronic levels are then allowed as perpendicular transition. Accordingly, it cannot be determined from present results whether the 1055.29 cm<sup>-1</sup> band is attributed to  $a'_1$  or  $e'$  ( $\nu_3$ ), unfortunately. The 2C-R4WM spectrum of the 1492 cm<sup>-1</sup> band region shows one  $Q$  head at 1499.79 cm<sup>-1</sup>, which is consistent with our dispersed fluorescence spectrum. By considering with the  $\nu_3 + \nu_4 - \nu_4$  hot band<sup>a</sup>, the present results suggest that both 1055.29 and 1499.79 cm<sup>-1</sup> levels are  $a'_1$  level.

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<sup>a</sup>K. Kawaguchi *et al.*, *J. Phys. Chem. A* 117, 13732 (2013) and E. Hirota, *J. Mol. Spectrosc.* 310, 99 (2015).