DYNAMICS OF CH_3F-($ortho$-H_2)n CLUSTERS IN SOLID $para$-H_2 CRYSTAL STUDIED BY PUMP AND PROBE SPECTROSCOPY USING TWO CW-QUATUM CASCADE LASERS

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The ν_3 vibrational band of CH_3F in solid $para$-H_2 is known to have a distinct regular series of lines corresponding to clusters forming CH_3F-($ortho$-$H_2)_n$, with n = 0 to 12 [a]. By using infrared cw-QC laser spectroscopy, we found many satellite lines around the main n-th cluster lines [b] and a photochromic phenomenon among those lines [c]. This time, we studied the dynamics of this reversible process by pump and probe spectroscopy using two cw-QC lasers. As direct absorption spectroscopy using a cw-QC laser provided high resolution and good signal-to-noise ratio spectrum even in rapid frequency scanning of 1 cm^{-1} with the reputation rate of 500Hz, one laser was used for monitoring the spectrum with very small radiation power, while the other was used for pumping a target peak with much higher power. Decrease of the pumped line and increase of the others including satellite lines were monitored in real-time with an oscilloscope and a video camera. The rate equation of three-level model including an intermediate state was successfully applied to explain the temporal behavior of those lines. This kinetic analysis gives us entirely new information for understanding the relation between the spectral lines and the structure of the clusters. For example, the rate constants thus determined suggests that there is another new regular series of the lines among the satellites lines around the n=0 main line.

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[c] A. R. W. McKellar, A. Mizoguchi, and H. Kanamori, Phys. Chem. Chem. Phys. 13, 11587(2011)