INVESTIGATION OF L-UNCOUPLING AND Λ-DOUBLING IN THE RYDBERG STATES OF THE SODIUM DIMER

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The phenomenon of electronic orbital angular momentum L uncoupled from its internuclear axis has been observed in the sodium dimer using high resolution cw optical-optical double resonance spectroscopy. When L-uncoupling occurs, the degeneracy of Λ-doubling is removed. In our experiment, the intermediate $B\,^1\Pi_u$ state of Na_2 is excited from the thermally populated ground $X\,^1\Sigma_g^+$ state by a single line Ar^+ laser. Then, a single-mode dye laser is used to probe the Rydberg states from the intermediate state. The signals are detected by monitoring the UV fluorescence from the triplet *gerade* states back to the $a\,^3\Sigma_u^+$ state via collision energy transfer. Under our experimental resolution, the splitting of L-uncoupling and Λ-doubling in both the $5\,^1\Delta_g$, $5\,^1\Pi_g$ states of Na_2 can be measured.Total of 136 rovibronic levels with e/f parities have been assigned to the $5\,^1\Delta_g$ state. The Λ-splitting constants deduced from these data are : q_0=$0.376(90)\times10^{-4}\,\mathrm{cm}^{-1}$, q_v=$0.114(6)\times10^{-4}\,\mathrm{cm}^{-1}$, μ= $0.76(33)\times10^{-8}\,\mathrm{cm}^{-1}$. In general, the Λ-splitting of the Δ states is considerably smaller than that of the Π states. However, the first order splitting constants q_0 and q_v reported here are larger than those in the $B\,^1\Pi_u$ state. This is due to the L-uncoupling of the Rydberg states. Total of 230 rovibrational levels are assigned to the Na_2 $5\,^1\Pi_g$ state, and the following are its correspondent Λ-splitting constants : q_0=$0.1758320898(0.017)\times10^{-3}\,\mathrm{cm}^{-1}$, q_v=$-0.7124318162(0.071)\times10^{-5}\,\mathrm{cm}^{-1}$, μ=$-0.9608208561(0.8)\times10^{-8}\,\mathrm{cm}^{-1}$. The splitting of Λ-doubling increases quadratically with the rotational quantum number J and weakly depends on the vibrational quantum number v. These splitting constants are much larger than those in the Na_2 $B\,^1\Pi_u$ state. This indicates that the splitting of Λ-doubling in the $5\,^1\Pi_g$ state is affected by both the perturbations by adjacent Σ states and the L-uncoupling.
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