

# Parallel and Coupled Perpendicular Transitions of RbCs 640 nm System

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Alkali metal diatomic molecules have been broadly investigated, both experimentally and theoretically, and have attracted much attention from various fields such as molecular spectroscopy, photodissociation dynamics, and photoassociation spectroscopy. Particularly, recent rapid progress in experiments with ultracold molecules has mainly focused on the heteronuclear alkali dimers since they can be applied to qubit generators for quantum computation.<sup>(1)</sup> RbCs is one of the heteronuclear alkali dimers. Recently, the low-lying strongly coupled  $1^1\Pi - 2^3\Sigma^+ - 1^3\Pi$  system of RbCs was employed for producing ultracold  $X^1\Sigma^+$  RbCs molecules.<sup>(2)</sup>

We report a newly identified parallel transition of  $2^3\Pi_0 \leftarrow X^1\Sigma^+$  and coupled perpendicular transitions of  $2^1\Pi$ ,  $2^3\Pi_1$ , and  $3^3\Sigma_1^+ \leftarrow X^1\Sigma^+$  observed by mass-resolved resonance enhanced two-photon ionization (RE2PI) in a cold molecular beam of RbCs.<sup>(3)</sup> Very complex vibronic structures have been observed in our RE2PI spectrum near 640 nm. The  $2^3\Pi_0$  state, however, shows a very regular vibronic structure, indicating the absence of significant perturbation. Rotationally-resolved spectra of the  $2^3\Pi_0 \leftarrow X^1\Sigma^+$  transitions were obtained. By fitting the observed term values of these parallel bands using the Dunham-type expression of the rovibronic term values, we have determined the molecular constants (electronic term value  $T_e = 15348.2928(96) \text{ cm}^{-1}$ , harmonic vibrational frequency  $\omega_e = 30.5175(26) \text{ cm}^{-1}$ , and rotational constant  $B_e = 0.012984(15) \text{ cm}^{-1}$ ) and constructed the Rydberg-Klein-Rees (RKR) potential energy curve of the  $2^3\Pi_0$  state.

The origin of the complex vibronic structures has been attributed to strong spin-orbit interactions among the  $\Omega = 1$  states. In the lower energy spectral region where the onsets of the  $2^3\Pi_1$  and  $3^3\Sigma_1^+ \leftarrow X^1\Sigma^+$  transitions were observed, we have identified the electronic symmetry and the vibrational quantum numbers of the upper vibronic states for the observed perpendicular bands (see Region I of Fig. 1). For the  $2^3\Pi_1$  state which shows regular vibronic structures for the vibrational levels  $v' \leq 4$ , we could determine the electronic term value and the harmonic vibrational frequency ( $T_e = 15437.34(14) \text{ cm}^{-1}$  and  $\omega_e = 31.726(96) \text{ cm}^{-1}$ ). The diagonal spin-orbit interaction constant of the  $2^3\Pi$  state ( $89 \text{ cm}^{-1}$ ) has been estimated from the observed splitting of the electronic term values of the  $\Omega = 0$  and 1 components. The observed intensity distribution of the  $3^3\Sigma_1^+ \leftarrow X^1\Sigma^+$  transitions implies that the equilibrium internuclear distance ( $R_e$ ) of the upper  $3^3\Sigma_1^+$  state is very close to that of the lower  $X^1\Sigma^+$  state. This allowed us to assign the lowest observed vibronic band of the  $3^3\Sigma_1^+ \leftarrow X^1\Sigma^+$  transitions to the  $v' = 0 \leftarrow v'' = 0$  origin band. The theoretically predicted Franck-Condon factors of the  $2^3\Pi_1$  and  $3^3\Sigma_1^+ \leftarrow X^1\Sigma^+$  transitions were in very good agreement with the observed vibronic-band intensity distributions. In the higher spectral region, we could hardly recognize the regular vibronic structures other than that of the  $2^3\Pi_0 \leftarrow X^1\Sigma^+$  transition (see Region II of Fig. 1). The vibronic energy level shifts of the  $2^1\Pi$  and  $2^3\Pi_1$  states

were clearly identified. In order to interpret the irregular vibronic structures observed in Region II, the ab initio calculation of the spin-orbit matrix elements and the deperturbation analysis using the theoretical parameters are in progress.

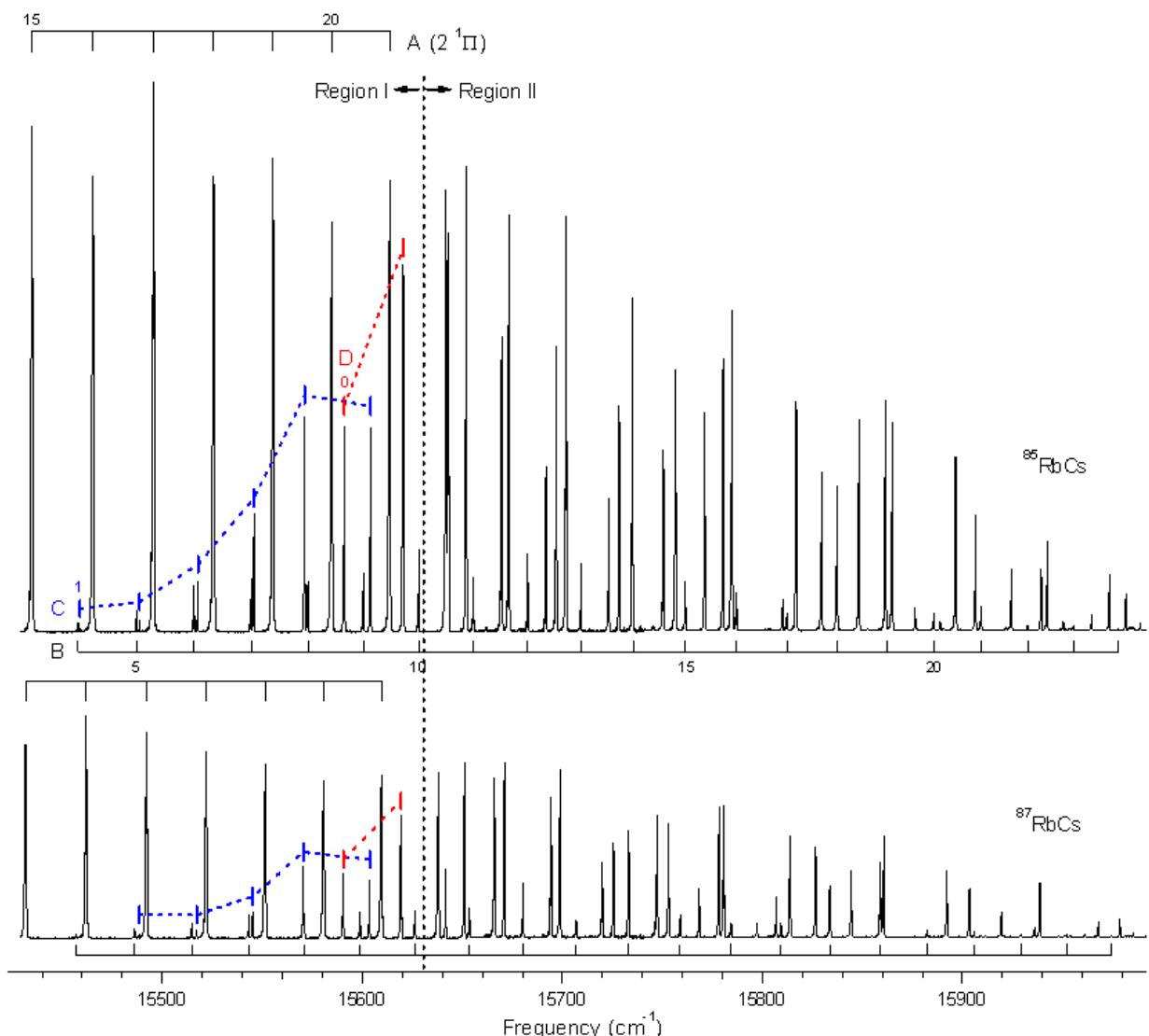


Fig. 1. Low-resolution RE2PI spectra of  $^{85}\text{RbCs}$  and  $^{87}\text{RbCs}$  between 15420 and 15990  $\text{cm}^{-1}$  obtained by expanding Ar carrier gas at 760 Torr. A ( $2 \ 1\Pi \leftarrow X \ 1\Sigma^+$ ), B ( $2 \ 3\Pi_0 \leftarrow X \ 1\Sigma^+$ ), C ( $2 \ 3\Pi_1 \leftarrow X \ 1\Sigma^+$ ), and D ( $3 \ 3\Sigma_1^+ \leftarrow X \ 1\Sigma^+$ ) progressions and their vibrational quantum numbers are indicated.

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