Fourier transform spectroscopy and deperturbation analysis of the spin-orbit coupled $A^1\Sigma^+$ and $b^3\Pi$ states in RbCs

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We present an extended study of the strongly spin-orbit coupled singlet $A^1\Sigma^+$ and triplet $b^3\Pi$ states ($A-b$ complex for short) of the RbCs molecule, which provide an efficient optical path to transfer ultracold molecules to their rovibrational ground state, as demonstrated in [1]. Earlier several Fourier-transform (FT) studies were performed to obtain the term values of the $A-b$ complex in RbCs, and to describe this complicated system by deperturbation model based on four $A^1\Sigma^+-b^3\Pi_{\Omega=0,1,2}$ coupled-channel (CC) approach [2,3]. However the achieved description of data have not reached the experimental accuracy. The goal of the present work was to obtain systematic term values data in the extended range of energy and quantum numbers of rovibronic levels, and to perform the CC deperturbation analysis allowing to reproduce the data with experimental accuracy. In the experiment the $A-b \rightarrow X$ and $(4)^1\Sigma^+ \rightarrow A-b$ laser induced fluorescence (LIF) FT spectra were recorded. RbCs molecules were produced in a heat pipe at 310 °C. A number of diode lasers covering the wavelength range from 830 nm to 1050 nm were used to excite directly the $A-b$ complex. In a different scheme, a single mode dye laser (Coherent 699-21) was used to excite the $(4)^1\Sigma^+$ state with subsequent observation of LIF signal to the $A-b$ complex. The latter scheme allowed us to observe 'dark' levels of the triplet $b^3\Pi$ state far below the minimum of the singlet $A$ state. Overall 8730 rovibronic term values of $A^1\Sigma^+$ and $b^3\Pi$ states of $^{85}\text{Rb}^{133}\text{Cs}$ and $^{87}\text{Rb}^{133}\text{Cs}$ isotopologues were determined with an uncertainty of 0.01 cm$^{-1}$ in the energy range [9012, 14087] covering rotational quantum numbers $J \in [6, 324]$.

An energy-based deperturbation analysis performed in the framework of the four $A^1\Sigma^+-b^3\Pi_{\Omega=0,1,2}$ coupled-channels approach reproduces 97% experimental term values of both isotopologues with a standard deviation of 0.0036 cm$^{-1}$. The reliability of the deperturbed mass-invariant potentials and spin-orbit coupling functions of the interacting $A^1\Sigma^+$ and $b^3\Pi$ states is additionally proved by a good reproduction of the $A-b \rightarrow X$ and $(4)^1\Sigma^+ \rightarrow A-b$ relative intensity distributions.

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References