

Ultracold molecular spectroscopy: toward the narrow-line cooling of molecules

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PAPER

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Abstract

A new spectroscopic method that utilizes microwave transition of ultracold molecules is demonstrated. The narrow spectral linewidth (approximately 100 Hz) was guaranteed by preparing molecules at low temperature, and a high signal-to-noise ratio was achieved by preparing a significant fraction of molecules in the target internal state. The repetition rate of the experiment was approximately 10 Hz, which was only limited by the time needed to load ultracold atoms into the magneto-optical trap. To demonstrate the performance, we investigated the hyperfine structures of the vibrational ground states of the $X^1\Sigma^+$ and $b^3\Pi_0^+$ states of KRb molecules. This technique not only allows us to pursue the narrow-line laser cooling of KRb molecules, but also provided us with essential information for realizing precision spectroscopies e.g., the search for the temporal variation of the electron-to-proton mass ratio.

1. Introduction

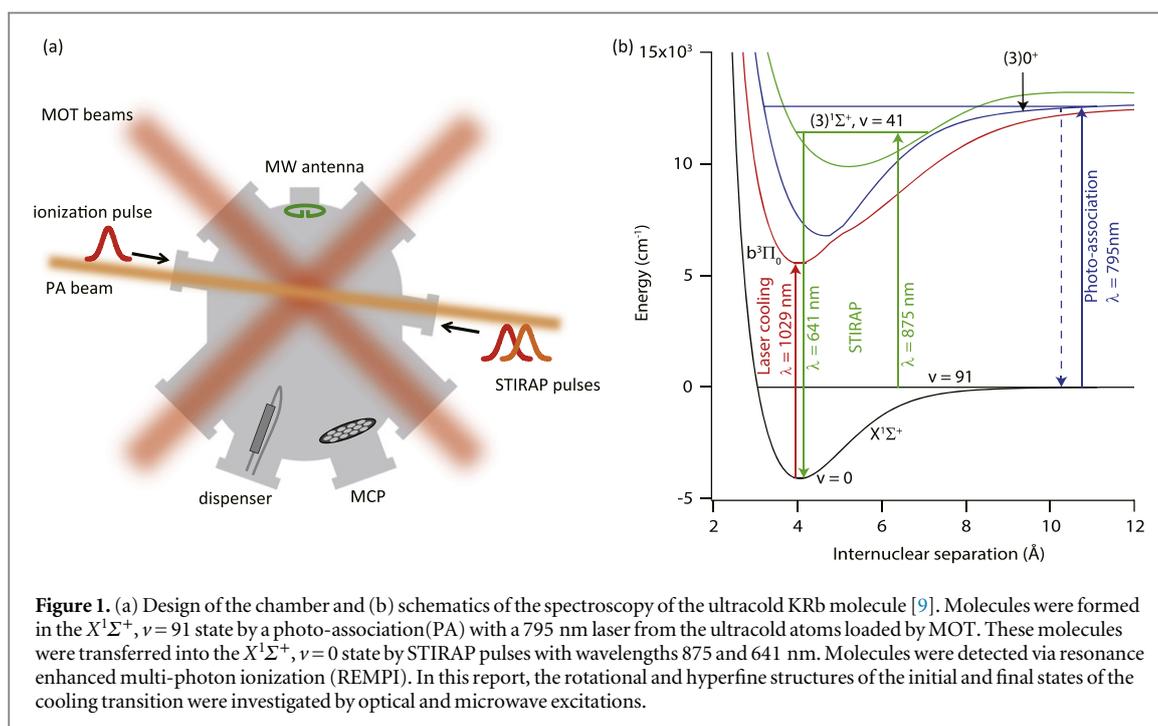
Recently, various methods have been developed for cooling molecules to ultracold temperatures [1–6]. These methods can be characterized by several figures of merit: the temperature, number of molecules, density, and repetition rate of the experiments. For producing a quantum degenerate sample of molecules, the density and temperature are of prime concern. For spectroscopic purposes, the number, repetition rate, and temperature are the key figures of merit.

We demonstrated that it is possible to produce KRb molecules in the rovibrational ground state by combining photo-association and stimulated Raman adiabatic passage (STIRAP) [7]. The temperature of the molecules was approximately 100 μ K, and the repetition rate of the experiment was approximately 10 Hz. Furthermore, we proposed a procedure to laser-cool these ground-state molecules to approximately the 1 μ K range by utilizing the narrow inter-combination transition [6]. A lower temperature would be beneficial not only for optical spectroscopy but also for spectroscopy in the microwave frequency, because it will increase the interaction time.

The idea of narrow-line cooling relies on the nearly-diagonal Franck–Condon factors (FCFs) between the ground state and the excited state [6]. Because the FCF is sensitive to the rotational constants, it is important to measure the rotational constants of each state. Here, we developed a method to measure the rotational constants by combining photo-association, STIRAP, and microwave spectroscopy of ultracold molecules. This method has been used for determining the rotational and hyperfine structure of KRb molecules in the $X^1\Sigma^+$, $\nu = 0$ state and $b^3\Pi_0^+$, $\nu = 0$ state. The rotational constant in the $X^1\Sigma^+$, $\nu = 0$ state was determined with 10-digit accuracy.

2. Ultracold molecular spectroscopy method

The experimental scheme for spectroscopy is similar to that described in our previous publications [7, 8] (figure 1). First, ^{87}Rb and ^{41}K atoms were loaded into a dual-species magneto-optical trap (MOT) from the

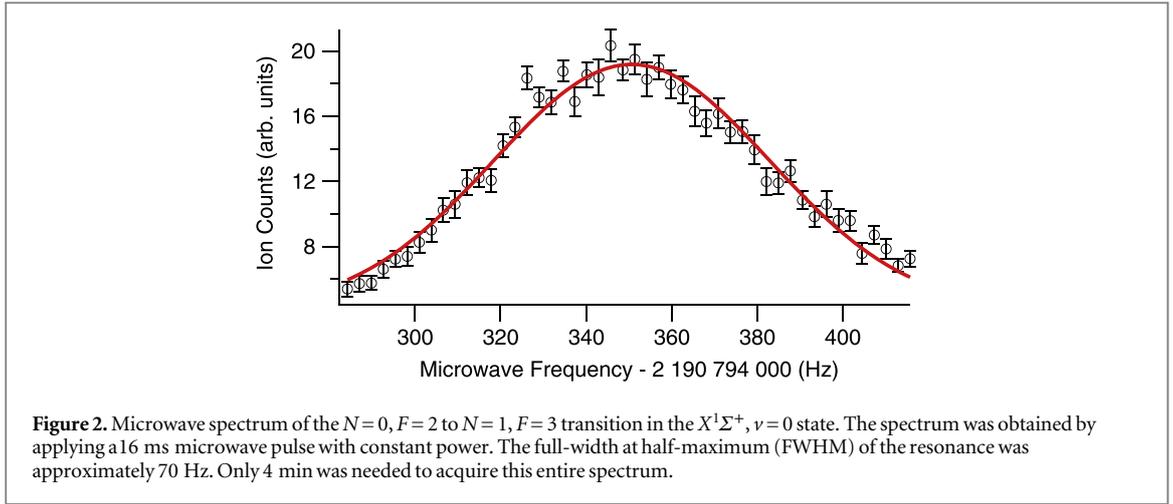


background gas, and compressed further during the compressed-MOT stage. The typical number of atoms, density, and temperature for the Rb (K) atoms were 3×10^7 (5×10^6), 3×10^{10} (9×10^9) cm^{-3} , and 70 (280) μK , respectively. A photo-association laser, which was red-detuned from the D_1 transition of Rb, was kept on during the MOT and compressed-MOT stage and produced loosely bound ultracold molecules in the electronic ground state.

The atomic vapor pressure in the chamber was approximately 10^{-9} Torr, and the time constant for MOT loading was approximately 1 s. Thus, one can increase the number of atoms in the MOT by increasing the loading time. However, as we increased the MOT loading time, the number of molecules formed by photo-association was rapidly saturated. We attributed this phenomenon to the saturation of atomic densities during the compressed-MOT stage. We consider that the densities saturated because of light-assisted collisions and radiation trapping. Thus, it was more efficient to keep the MOT loading time approximately 50 ms. One can then repeat the entire experiment in approximately 120 ms, which matches well with the repetition rate of the pulsed laser used for detecting ionized molecules. This high repetition rate of the experiment contrasts well with experiments using evaporative cooling, which typically require more than 10 s.

Photo-association followed by a spontaneous decay is a strong technique for producing loosely bound molecules. The irreversibility of the process is ensured by the spontaneous nature of the decay. However, precisely because of this spontaneous nature of the decay, the obtained molecules were distributed in many (electronic, vibrational, rotational, electron-spin, and nuclear-spin) internal states. To increase the branching ratio to the target state, we used a vibrational state in the $(3)0^+$ potential. We selected a bound state that lies 8.8 cm^{-1} below the Rb D_1 line, whose FCF to the $X^1\Sigma^+, v=91$ was as high as 0.1. The number of molecules per single hyperfine level in the $X^1\Sigma^+, v=91$ state was of the order of 100, and the temperature of the loosely bound molecules was approximately 130 μK . Molecules were detected via resonance enhanced multi-photon ionization (REMPI), i.e., ionization with a ns-pulsed laser followed by detection by micro channel plate (MCP).

In this study, we have performed microwave spectroscopy for ultracold molecules. Because the molecules were not trapped, the interaction time of the spectroscopy was limited by the thermal diffusion of the molecular cloud. Because the mean velocity of the molecules and the diameter of the ionization laser were approximately 0.13 m s^{-1} and 4 mm, respectively, the interaction time was about 10 ms. The only way to improve this interaction time further is to construct a ‘molecular fountain’ because gravity cannot be neglected on such a long time scale.



3. Investigation of the rotational and hyperfine structures of the $X^1\Sigma^+, \nu=0$ and $b^3\Pi_{0^+}, \nu=0$ states toward the laser cooling

Cooling the molecule is a simple solution for increasing the interaction time and making the spectral linewidth narrower. We proposed a narrow-line laser cooling of a KRb molecule using $X^1\Sigma^+, \nu=0 - b^3\Pi_{0^+}, \nu=0$ transition, where the molecules could be cooled into sub-microkelvin [6]. To discuss the prospects of the laser cooling, we must investigate the rotational and hyperfine structure of the cooling transition. In the following, we will report on the experiments and analysis of the rotational and hyperfine structures of the $X^1\Sigma^+, \nu=0$ and $b^3\Pi_{0^+}, \nu=0$ and $\nu=1$ vibrational states.

3.1. Spectroscopy of the rotational and hyperfine structures of the $X^1\Sigma^+, \nu=0$ state

The hyperfine structure of the $X^1\Sigma^+, \nu=0$ state is theoretically predicted in [10]. We have experimentally determined the structure by observing the microwave transition of $N=0$ to $N=1$. Using the sequence described above, the molecules were transferred to the $X^1\Sigma^+, \nu=0, N=0$ state using STIRAP, where the molecules were distributed in $F=0-3$ hyperfine states. The molecules were then transferred into the $N=1$ state by applying a microwave pulse, whose frequency was approximately 2 GHz. Because of the large electric dipole moment of the KRb molecule, minimal microwave power was needed to drive the transition. To detect the molecules in the $N=1$ state selectively, the molecules in the $N=0$ state were optically pumped into other vibrational states. figure 2 shows the typical spectrum of the microwave transition. The interaction time was 16 ms, and the full-width-of-half-maximum (FWHM) of the resonance was approximately 70 Hz. The spectrum was slightly distorted because of the inhomogeneous electric field. All 4 (10) hyperfine states in the $N=0$ ($N=1$) rotational states were observed.

According to [10, 11], the energy level structure of $X^1\Sigma^+, \nu=0$ state can be derived using the following Hamiltonian, which is the sum of contributions from rotation (H_{rot}), hyperfine (H_{hf}), the Stark effect (H_S), and the Zeeman effect (H_Z):

$$H = H_{\text{rot}} + H_{\text{hf}} + H_S + H_Z. \quad (1)$$

Each component can be expressed as follows

$$\begin{aligned} H_{\text{rot}} &= BN^2, \\ H_{\text{hf}} &= \sum_{i=\text{K,Rb}} V_i \cdot Q_i + \sum_{i=\text{K,Rb}} c_i N \cdot I_i + c_3 I_K \cdot T \cdot I_{\text{Rb}} + c_4 I_K \cdot I_{\text{Rb}}, \\ H_S &= -\boldsymbol{\mu} \cdot \mathbf{E}, \\ H_Z &= -g_r \mu_N N \cdot \mathbf{B} - \sum_{i=\text{K,Rb}} g_i \mu_N I_i \cdot \mathbf{B} (1 - \sigma_i). \end{aligned}$$

The rotational Hamiltonian (H_{rot}) describes the rotational energy with the rotational constant B . The hyperfine Hamiltonian (H_{hf}) comprises of four terms. The first term describes the electronic quadrupole interaction with coupling constants (eQq_{K} and (eQq_{Rb}) , where q_{K} (q_{Rb}) is the electric field gradient at the nuclear of the K(Rb) atom and eQ_{K} (eQ_{Rb}) is its quadrupole moment. The second term describes the interaction between the rotational angular momentum and the nuclear spins with the coupling constants c_{K} and c_{Rb} . The

Table 1. Hyperfine and rotational constants of $X^1\Sigma^+$, $\nu = 0$. The observed spectra were analyzed using the Hamiltonian shown in equation (1) (see text). All the energies are given in Hertz (Hz).

	Other works	This work
$(eQq)_K$	-2.98×10^5 ([10])	$-3.35206(6) \times 10^5$
$(eQq)_{Rb}$	-1.52×10^6 ([10])	$-1.415413(3) \times 10^6$
c_K	10.4 ([10])	10.4(6)
c_{Rb}	413.1 ([10])	465.7(3)
c_3	21.3 ([10])	22.3(3)
c_4	896.2 ([10])	926.6(3)
B	$1.09539(2) \times 10^9$ ([12])	$1.09537730535(30) \times 10^9$

third and fourth terms describes the tensor and scalar coupling between the nuclear spins with coupling constants c_3 and c_4 , respectively.

The Stark Hamiltonian (H_S) describes the interaction between the molecular electric dipole moment (μ) and the external electric field (E). The value of μ was experimentally measured in [1]. In our experiment, an electric field of $0.19(6) \text{ V m}^{-1}$ was applied from the MCP during the interaction time, which resulted in a Stark shift of the order of 100 mHz, which was included in our analysis.

The Zeeman Hamiltonian (H_Z) describes the interaction between the molecular magnetic moment and the external magnetic field (B). The first and second term describe the magnetic moments caused by the rotation and nuclear spins, respectively, where g_r, g_K, g_{Rb} are the g factors for the rotation and nuclear spins of K and Rb atoms, respectively. σ_K and σ_{Rb} in the second term are the shielding tensors for K and Rb, respectively, which give small corrections to the Zeeman shift. The values of the g factors and σ s for the KRb molecule are predicted in [10]. The magnetic field during the interaction time was approximately $0.1 \mu\text{T}$. This magnetic field causes Zeeman splitting of the order of 1 Hz, which was smaller than our resolution limit. In the analysis, we assume equal occupations between all the magnetic sub-levels.

The hyperfine and rotational constants obtained by analyzing the experimental data are listed in table 1 with theoretical predictions from other publications. Although error bars for the theoretical predictions are missing, the overall agreement is quite convincing. This precise value of the rotational constant should be a great help for the precise determination of FCFs.

3.2. Spectroscopy of the rotational and hyperfine structures of the $b^3\Pi_{0^+}$ state

To date, there have been neither theoretical nor experimental studies on the hyperfine structures of the $b^3\Pi_{0^+}$ state. However, a detailed study on its hyperfine structure is mandatory to achieve narrow-line cooling of KRb molecules. We used the same Hamiltonian as that used in equation (1), because the projection of the total electronic angular momentum along the internuclear axis (i.e., Ω) is zero. The study of the $b^3\Pi_{0^+}$ state was performed optically. First, molecules were transferred to the $X^1\Sigma^+$, $\nu = 0$, $N = 0$ state using a combination of photo-association and STIRAP. Then, the molecules were excited into the $b^3\Pi_{0^+}$ state by a resonant laser (1029 nm) (figure 1), whose frequency was stabilized with a high-finesse optical cavity [13]. The spectrum was obtained by scanning the frequency of the excitation laser and counting the number of molecules decayed to other vibrational levels of the $X^1\Sigma^+$ state.

Figure 3(a) shows the spectrum of the $X^1\Sigma^+$, $\nu = 0$, $N = 0$ to $b^3\Pi_{0^+}$, $\nu = 0$, $N = 1$ transition. The spectral linewidth was determined by the Doppler broadening effect (approximately 200 kHz), which was much wider than the natural linewidth (approximately 5 kHz). To increase the signal-to-noise ratio, we broadened the linewidth of the excitation laser to a few hundred kilohertz by applying a deep phase modulation using an electro-optic modulator. What was observed in the spectrum are the hyperfine structure of the $b^3\Pi_{0^+}$ state, because the hyperfine splitting of the $X^1\Sigma^+$, $\nu = 0$, $N = 0$, $F = 0-3$ (several kHz) was too small to resolve. For the assignment of each signal, we performed a double resonance spectroscopy, where the microwave and the excitation laser were applied simultaneously.

Figure 3(b) shows the level structure of the $b^3\Pi_{0^+}$, $\nu = 1$, $N = 0$ state. The molecules in the $X^1\Sigma^+$, $\nu = 0$, $N = 0$ state cannot be directly excited into the $b^3\Pi_{0^+}$, $\nu = 1$, $N = 0$ state because of the selection rule on N . Therefore, the molecules in the $X^1\Sigma^+$, $\nu = 0$, $N = 0$ state were transferred into the $N = 1$ states by a microwave transition and then excited into the $b^3\Pi_{0^+}$, $\nu = 1$, $N = 0$ state, using a light pulse with higher intensity to drive the weak transition. The spectrum of the $b^3\Pi_{0^+}$, $\nu = 0$, $N = 0$ state was also obtained in a similar manner.

The rotational and hyperfine constants of $b^3\Pi_{0^+}$, $\nu = 0$ and $\nu = 1$ state are summarized in table 2. These values play an essential role in estimating the FCF between the $X^1\Sigma^+$, $\nu = 0$ and $b^3\Pi_{0^+}$, $\nu = 0$ state [6].

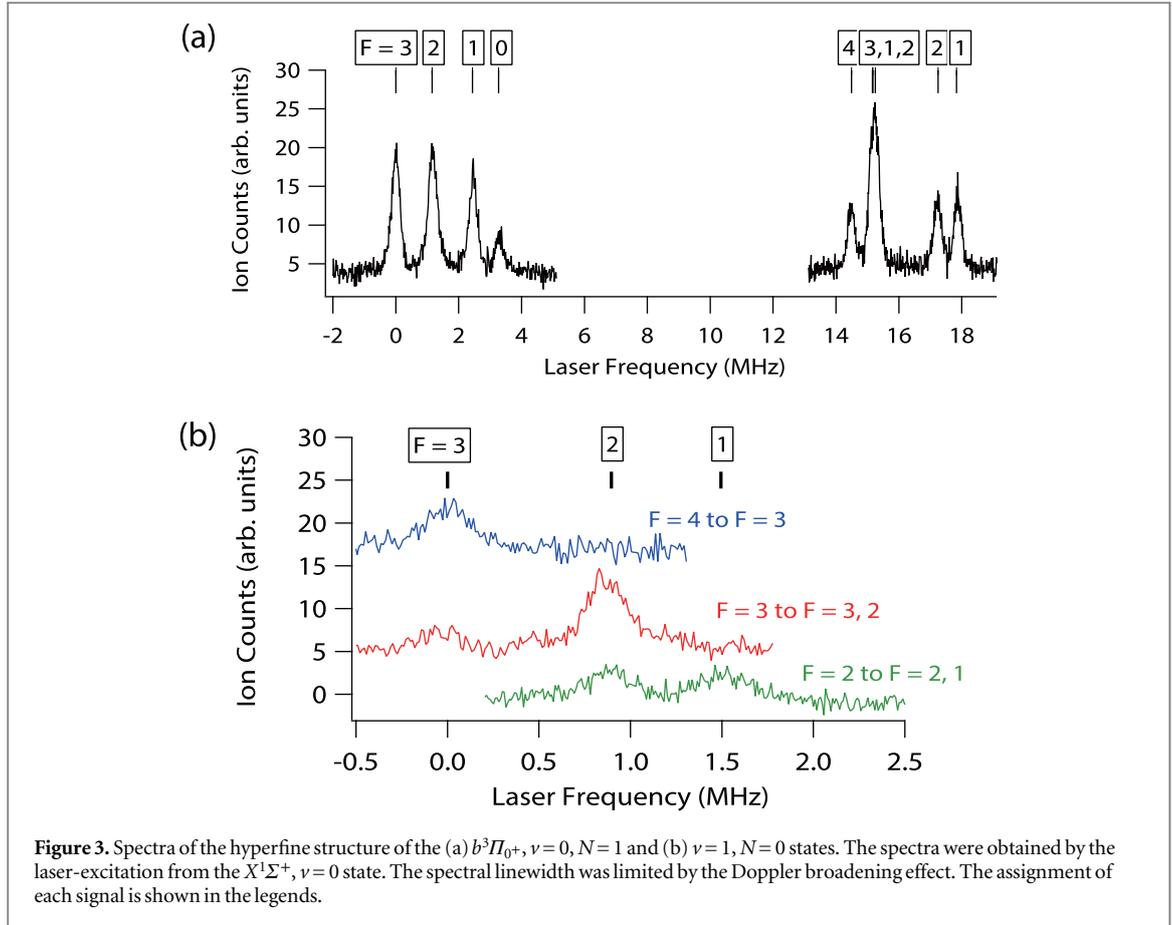
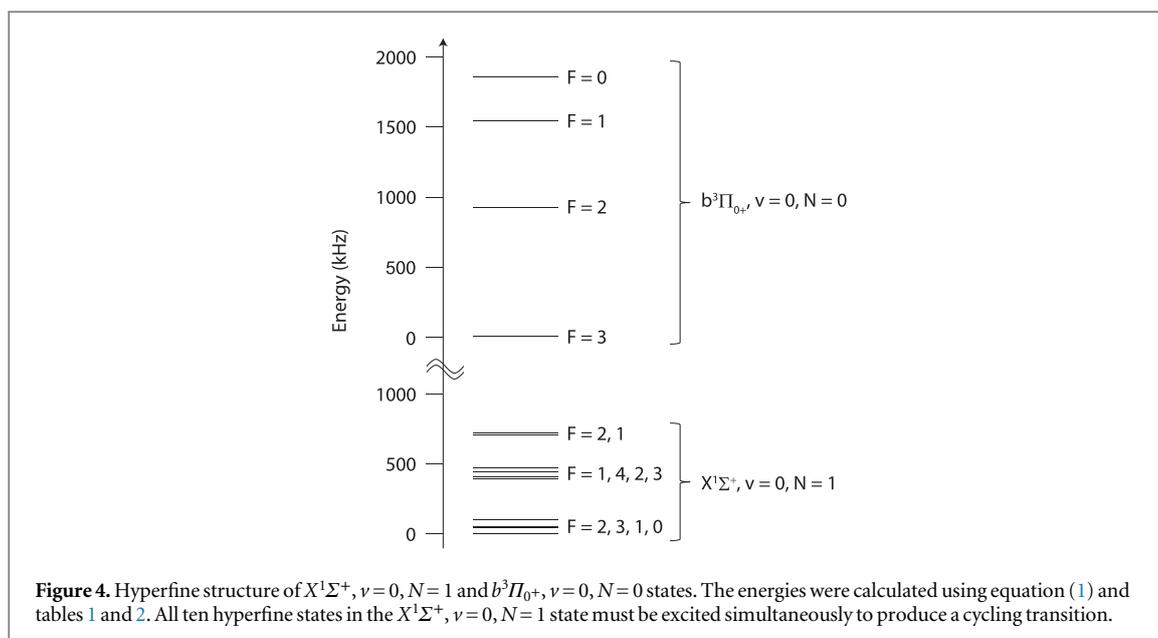


Table 2. Rotational and hyperfine constants of $b^3\Pi_{0^+}, \nu=0$ and $\nu=1$. The observed spectra were analyzed using the Hamiltonian shown in equation (1). All the energies are given in kilohertz (kHz).

	$b^3\Pi_{0^+}, \nu=0$	$b^3\Pi_{0^+}, \nu=1$
$(eQq)_K$	1719(70)	1767(79)
$(eQq)_{Rb}$	-41121(28)	-39210(34)
c_K	46(7)	49(10)
c_{Rb}	1429(4)	1410(5)
c_3	313(4)	297(5)
c_4	-307(2)	-307(2)
B	1118393(3)	1116245(3)

3.3. Toward the laser cooling of KRb molecules

For the laser cooling of the molecules, the cooling transition should be closed for the vibrational, rotational, and hyperfine states. We reported that the FCFs of the $X^1\Sigma^+ - b^3\Pi_{0^+}$ transition are nearly diagonal, and thus one can form the cycling transition for the vibrational states [6]. For the rotational states, one can use the $X^1\Sigma^+, \nu=0, N=1$ to $b^3\Pi_{0^+}, \nu=0, N=0$ transition. The hyperfine structure of the $X^1\Sigma^+, \nu=0, N=1$ and $b^3\Pi_{0^+}, \nu=0, N=0$ states are shown in figure 4. The $X^1\Sigma^+, \nu=0, N=1$ state splits into 10 hyperfine states, where the energy splitting of the nearest states is larger than the natural linewidth of the $X^1\Sigma^+$ to $b^3\Pi_{0^+}$ transition. The $b^3\Pi_{0^+}, \nu=0, N=0$ state splits into four hyperfine states with a larger separation than that of the $X^1\Sigma^+, \nu=0, N=1$ state. To make a cycling transition, all of the hyperfine states in the $X^1\Sigma^+, \nu=0, N=1$ state should be excited simultaneously. The magnetic sublevels of $F=4, |m_F|=4$ become dark states under the π -polarization cooling laser. These dark states could be eliminated by applying a magnetic field of the order of 0.1 mT.



4. Summary

We developed a new type of spectroscopy using ultracold molecules. A narrow linewidth was achieved by preparing molecules at low temperature, and STIRAP transfer was used to prepare molecules in specific internal states. Using this method, we determined the rotational constant of $X^1\Sigma^+, v=0$ with 100 mHz accuracy. We also determined the energy level structure of the $b^3\Pi_{0^+}$ state. To further reduce the linewidth of the resonance, we discussed the prospect of narrow-line laser cooling. Our method could be applied to other precision spectroscopies, such as in the investigation of the temporal variation of the electron-to-proton mass ratio [14].

Acknowledgments

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