Controlling the Rotational and Hyperfine State of Ultracold $^{87}\text{Rb}^{133}\text{Cs}$ Molecules

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We demonstrate coherent control of both the rotational and hyperfine state of ultracold, chemically stable $^{87}\text{Rb}^{133}\text{Cs}$ molecules with external microwave fields. We create a sample of $\sim$ 2000 molecules in the lowest hyperfine level of the rovibrational ground state $N = 0$. We measure the transition frequencies to 8 different hyperfine levels of the $N = 1$ state at two magnetic fields $\sim 23$ G apart. We determine accurate values of rotational and hyperfine coupling constants that agree well with previous calculations. We observe Rabi oscillations on each transition, allowing complete population transfer to a selected hyperfine level of $N = 1$. Subsequent application of a second microwave pulse allows transfer of molecules back to a different hyperfine level of $N = 0$.

Ultracold heteronuclear molecules can provide many exciting new avenues of research in the fields of quantum state controlled chemistry, quantum information, quantum simulation, and precision measurement. The large electric dipole moments accessible in such systems allow interactions to be tuned over length scales similar to the spacing between sites in an optical lattice. As such, this is an area of intense research with multiple groups recently reporting the production of dipolar molecules at ultracold temperatures.

Full control of the quantum state has been an invaluable tool in ultracold atom physics; it is therefore highly important to develop similar methods for ultracold molecules which address the complex rotational and hyperfine structure. Such control is at the heart of nearly all proposals for applications of ultracold polar molecules. For example, the rotational states of molecules might be used as pseudo-spins to simulate quantum magnetism. This requires a coherent superposition of opposite-parity states to generate dipolar interactions, which may be probed by microwave spectroscopy. Similarly, hyperfine states in the rotational ground state have been proposed as potentially useful tools in high precision measurements.

In this letter, we report microwave spectroscopy of bosonic $^{87}\text{Rb}^{133}\text{Cs}$ in its ground vibrational state, and coherent state transfer from the absolute rovibrational and hyperfine ground state to a chosen well-defined single hyperfine state in either the first-excited or ground rotational states. We demonstrate the high precision with which we can map out the rotational energy structure of the $^{87}\text{Rb}^{133}\text{Cs}$ molecule in the lowest vibrational state.

We calculate the energy level structure of $^{87}\text{Rb}^{133}\text{Cs}$ in the electronic and vibrational ground state by diagonalizing the relevant Hamiltonian. In the presence of an externally applied magnetic field, this Hamiltonian ($H$) can be decomposed into rotational ($H_{r}$), hyperfine ($H_{hf}$), and Zeeman ($H_{z}$) components.

$$H = H_{r} + H_{hf} + H_{z}. \quad (1)$$

These contributions are given by

$$H_{r} = B_{v}N^{2} - D_{v}N^{2}N^{2}, \quad (2a)$$

$$H_{hf} = \sum_{i=Rb,Cs} V_{i} \cdot Q_{i} + \sum_{i=Rb,Cs} c_{i}N \cdot I_{i} + c_{3}I_{Rb} \cdot T \cdot I_{Cs} + c_{4}I_{Rb} \cdot I_{Cs}, \quad (2b)$$

$$H_{z} = -g_{r}\mu_{N}B \cdot N - \sum_{i=Rb,Cs} g_{i}(1 - \sigma_{i})\mu_{N}I_{i} \cdot B. \quad (2c)$$

The rotational contribution (Eqn. 2a) is defined by the rotational angular momentum of the molecule $N$, and the rotational and centrifugal distortion constants $B_{v}$ and $D_{v}$. The hyperfine contribution (Eqn. 2b) consists of four terms. The first term describes the electric quadrupole interaction with coupling constants ($eqQ_{Rb}$) and ($eqQ_{Cs}$). The second term is the interaction between the nuclear magnetic moments and the magnetic field generated by the rotation of the molecule, with spin-rotation coupling constants $c_{Rb}$ and $c_{Cs}$. The final two
terms represent the tensor and scalar interactions between the nuclear magnetic moments, with tensor and scalar spin-spin coupling constants \( c_3 \) and \( c_4 \) respectively. Finally, the Zeeman contribution (Eqn. 2c) consists of two terms which represent the rotational and nuclear interaction with an externally applied magnetic field. The rotation of the molecule produces a magnetic moment which is characterized by the rotational \( g \)-factor of the molecule \((g_r)\). The nuclear interaction similarly depends on the nuclear \( g \)-factors \((g_{RB}, g_{Cs})\) and nuclear shielding \((\sigma_{RB}, \sigma_{Cs})\) for each species. We do not apply electric fields in this work, which would require the addition of a further Stark contribution to the Hamiltonian and significantly complicate the spectra [28].

The nuclear spins in \(^{87}\)Rb\(^{133}\)Cs are \( I_{RB} = 3/2 \) and \( I_{Cs} = 7/2 \). At zero field, the total angular momentum \( F = N + I_{RB} + I_{Cs} \) is conserved. For the rotational ground state \((N = 0)\), the total nuclear spin \( I = I_{RB} + I_{Cs} \) is very nearly conserved, and there are 4 hyperfine states with \( I = 2, 3, 4 \) and 5 with separations determined by \( c_4 \) [27]. For excited rotational states, however, only \( F \) is conserved and \( I \) is a poor quantum number.

An external magnetic field splits each rotational manifold into \((2N + 1)(2I_{RB} + 1)(2I_{Cs} + 1)\) Zeeman-hyperfine sublevels, so there are 32 levels for \( N = 0 \) and 96 levels for \( N = 1 \). Assignment of quantum numbers to the individual hyperfine levels is non-trivial and depends on the magnetic field regime [27]. The field mixes states with different values of \( F \) that share the same total projection \( M_F \). At low field, the levels are still approximately described by \( F \) and \( M_F \) (equivalent to \( I \) and \( M_I \) for \( N = 0 \)). At high field, however, the nuclear spins decouple and the individual projections \( M_N, m_{RB}^I \) and \( m_{Cs}^I \) become nearly good quantum numbers, with \( M_F = M_N + m_{RB}^I + m_{Cs}^I \).

A microwave field induces electric dipole transitions between rotational levels. At low field, all transitions allowed by the selection rules \( \Delta F = 0, \pm 1 \) and \( \Delta M_F = 0, \pm 1 \) have significant intensity. At higher field, however, additional selection rules emerge. If hyperfine couplings are neglected, electric dipole transitions leave the nuclear spin states unchanged \((\Delta m_{RB}^I = \Delta m_{Cs}^I = 0)\) and are allowed only between neighboring rotational states such that \( \Delta N = \pm 1, \Delta M_N = 0, \pm 1 \) for microwave polarizations \( \pi, \sigma^\pm \). In the absence of hyperfine interactions (where \( M_N \) would be a good quantum number) we would be able to drive at most three transitions from any given hyperfine level, as shown in Fig. 1a). Nuclear quadrupole coupling in the \( N = 1 \) state mixes levels with different values of \( M_N, m_{RB}^I \) and \( m_{Cs}^I \), and additional transitions become allowed. The relative strengths of the transitions depend on the magnitude of the component of the destination state that preserves \( m_{RB}^I \) and \( m_{Cs}^I \) [29]. The presence of state mixing allows us to use a multi-photon scheme to move the population to different hyperfine states of the rotational ground state. Fig. 1b) shows an example of the simplest possible variation of the scheme using two microwave photons to change the hyperfine state by \( \Delta M_F = -1 \).

Our experimental apparatus and method for creating ultracold \(^{87}\)Rb\(^{133}\)Cs molecules have been discussed in previous publications [11, 29–34]; we will therefore give only a brief overview here. We begin by using magnetoassociation on a magnetic Feshbach resonance to create weakly bound molecules from an ultracold atomic mixture confined in a crossed-beam optical trap (\( \lambda = 1550 \) nm) [33]. We remove the remaining atoms by means of the Stern-Gerlach effect, leaving a pure sample of trapped molecules. These molecules are then transferred to a single hyperfine state of the rovibational ground-state by stimulated Raman adiabatic passage (STIRAP) [11, 35]. In this work, we create a sample of up to \( \sim 2000 \) \(^{87}\)Rb\(^{133}\)Cs molecules in the lowest hyperfine state (shown in Fig. 2a) at a temperature of 1.17(1) \( \mu \)K and peak density of \( 8.1(8) \times 10^{10} \) cm\(^{-3} \). It is important to note that in order to measure the number of molecules in our experiment we reverse both the STIRAP and magnetoassociation steps and subsequently use absorption imaging to detect the atoms that result from the molecular dissociation. Throughout, therefore, we always measure the number of molecules in the hyperfine state initially populated by STIRAP.

Our apparatus is equipped with two omnidirectional \( \lambda/4 \) antennas placed close to the outside of the fused silica cell. The polarization from each is roughly linear at the position of the molecules. They are oriented perpendicular to each other and aligned with respect to the direction of the static magnetic field such that one preferentially drives transitions with \( \Delta M_F = 0 \) and the other drives those with \( \Delta M_F = \pm 1 \). Each antenna is con-

![FIG. 1. (Color online) Electric dipole transitions in \(^{87}\)Rb\(^{133}\)Cs between the \( N = 0 \) and \( N = 1 \) rotational levels in the vibrational ground state. (a) First-order allowed electric-dipole transitions keep the hyperfine state projections from the atomic nuclear angular momentum unchanged \((\Delta m_{RB}^I = \Delta m_{Cs}^I = 0)\). In the absence of hyperfine interactions we would therefore expect to be able to drive 3 transitions with \( \Delta M_F = \Delta M_N = 0, \pm 1 \) for microwave polarizations \( \pi, \sigma^\pm \). (b) Scheme for changing nuclear angular momentum in the rotational ground state. Interactions involving the nuclear electric quadrupole moments of \(^{87}\)Rb and \(^{133}\)Cs mix quantum states with different nuclear spin quantum numbers \( m_{Nd}^I \) and \( m_{Fs}^I \). We use a two-photon pulse sequence to transfer up to a mixed excited quantum state in which both the initial and desired values of \( M_N \) are present.

![diagram](image-url)
frequencies from two magnetic fields at which spectroscopy is performed in this work. (b) Comparison of experimentally measured transition state in \( N = 0 \) and \( N = 1 \) states. The initial state in \( N = 0 \) is highlighted as the bold red line. The 10 states that are accessible from this initial state in \( N = 1 \) are shown as bold blue (\( M_F = 4 \)), red (\( M_F = 5 \)), and green (\( M_F = 6 \)) lines. The vertical dotted lines mark the two magnetic fields at which spectroscopy is performed in this work. (b) Comparison of experimentally measured transition frequencies from \( |N = 0, M_F = 5\rangle \) to \( |N = 1, M_F = 4, 5, 6\rangle \) with the fitted theory. Dashed lines indicate transitions that are weakly allowed but we have not observed. Error bars are not visible at this scale (see Table I). (c–j) Spectra of all the transitions found in this work at a magnetic field of \( \sim 181.5 \) G. The vertical lines shows the position of each transition given by fit to the results. The widths of all of the features are Fourier-transform limited. The pulse duration used is less than a \( \pi \) pulse for each transition. The specific pulse durations are (c) 12 \( \mu \)s, (d) 150 \( \mu \)s, (e) 100 \( \mu \)s, (f) 400 \( \mu \)s, (g) 60 \( \mu \)s, (h) 50 \( \mu \)s, (i) 400 \( \mu \)s, (j) 200 \( \mu \)s.

![Fig. 2](image)

**TABLE I.** Microwave transitions found in \( ^{87}\text{Rb}^{133}\text{Cs} \) from \( |v = 0, N = 0\rangle \) to \( |v = 0, N = 1\rangle \). All transitions start from the spin-stretched \( M_F = +5 \) hyperfine level of the rotational ground state. Each transition is labeled by the \( M_F \) quantum number of the destination hyperfine level in the first-excited rotational state.

| \( M_F \) | \( B \) (G) | \( f_{\text{the}} \) (kHz) | \( f_{\text{Exp.}} \) (kHz) | \( \Delta f \) (kHz) |
|---|---|---|---|---|
| +5 | 181.507(2) | 980 231.07 | 980 233(2) | -2(2) |
| | 204.436(2) | 980 235.14 | 980 237(1) | -2(1) |
| +4 | 181.484(1) | 980 277.96 | 980 278.9(2) | -0.9(2) |
| | 204.397(2) | 980 292.08 | 980 291.0(2) | 1.1(2) |
| +4 | 181.487(1) | 980 320.47 | 980 320.4(2) | 0.1(2) |
| | 204.397(2) | 980 331.83 | 980 331.8(3) | 0.0(3) |
| +6 | 181.541(2) | 980 384.98 | 980 384.97(6) | 0.01(6) |
| | 204.38(1) | 980 384.87 | 980 384.90(5) | 0.03(5) |
| +5 | 181.507(2) | 980 433.97 | 980 444.8(7) | -0.8(7) |
| | 204.436(2) | 980 458.35 | 980 457.2(8) | 1.1(3) |
| +5 | 181.507(2) | 980 546.75 | 980 546.9(7) | -0.2(7) |
| | 204.436(2) | 980 572.86 | 980 573.5(6) | 0.6(6) |
| +4 | 181.487(1) | 980 661.35 | 980 661.15(6) | 0.20(6) |
| | 204.397(2) | 980 694.22 | 980 694.35(5) | -0.13(5) |
| +4 | 181.487(1) | 980 758.64 | 980 758.6(1) | 0.01(1) |
| | 204.397(2) | 980 810.62 | 980 810.8(3) | 0.2(3) |

TABLE I. Microwave transitions found in \( ^{87}\text{Rb}^{133}\text{Cs} \) from \( |v = 0, N = 0\rangle \) to \( |v = 0, N = 1\rangle \). All transitions start from the spin-stretched \( M_F = +5 \) hyperfine level of the rotational ground state. Each transition is labeled by the \( M_F \) quantum number of the destination hyperfine level in the first-excited rotational state.

nected to a separate signal generator, which is frequency referenced to an external 10 MHz GPS reference. Fast \((\sim 100\) ns) switches are used to generate microwave pulses of well-defined duration (typically 1 \( \mu \)s - 500 \( \mu \)s).

The large dipole moment of the molecule (1.225 D [11]) makes it easy to drive fast Rabi oscillations between neighboring rotational states. To perform the spectroscopy, therefore, we pulse on the microwave field for a time \((t_{\text{pulse}})\) which is less than the duration of a \( \pi \) pulse for the relevant transition \((< t_\pi)\). We then observe the transition as an apparent loss of molecules as they are transferred into the first-excited rotational state. To avoid ac Stark shifts of the transition centers, the optical trap is switched off throughout the spectroscopy; the transition frequencies are thus measured in free space. We find that the widths of all of the features we measure are Fourier-transform limited, i.e. the width is proportional to \( 1/t_{\text{pulse}} \). We therefore iteratively reduce the power to get slower Rabi oscillations and allow longer pulse durations. We also note that radically different \( t_{\text{pulse}} \) are required for each transition depending on the transition strength and antenna used. We carry out the spectroscopy at two different magnetic fields \( \sim 23 \) G apart; the magnetic field is calibrated using the microwave transition frequency between the \(|f = 3, m_f = +3\rangle\) and \(|f = 4, m_f = +4\rangle\) states of Cs.

With the population initially in the lowest hyperfine level \((M_F = 5)\) of the rovibrational ground state, we expect to find a maximum of 10 transitions to the first-excited rotational state \(|N = 1, M_F = 4, 5, 6\rangle\). We are able to observe 8 of these transitions, at the frequencies given in Table I. A complete set of spectra at a magnetic field of \( \sim 181.5 \) G is also shown in Fig. 2(c–j). Calculations of the expected intensities of the two unseen transitions show that the relative transition probability is \( \sim 10^{-4} \) lower than for those we do observe.

We fit our model to the experimental spectra by minimizing the sum of the squared quotients between each residual and the uncertainty of the line. We fit the rotational constant, nuclear quadrupole constants and
TABLE II. Constants involved in the molecular Hamiltonian for $^{87}$Rb$^{133}$Cs. Parameters not varied in the least-squares fit are taken from the literature. The majority of the fixed terms are calculated using density-functional theory (DFT) \cite{27}, with the exception of the centrifugal distortion constant $D_{\nu}$, which is obtained from laser-induced fluorescence combined with Fourier transform spectroscopy (LIF-FTS) \cite{36}.

| Constant | Value (MHz) | Ref. |
|----------|-------------|------|
| $B_{c}$ | 490.155(5) | \cite{36} |
| $D_{c}$ | 490.173 994(45) | This Work |
| $(eQq)_{Rb}$ | -872 kHz | \cite{27} |
| $(eQq)_{Cs}$ | -809.29(1.13) kHz | This Work |
| $c_{Rb}$ | 29.4 Hz | \cite{27} |
| $c_{Cs}$ | 196.8 Hz | \cite{27} |
| $c_3$ | 192.4 Hz | \cite{27} |
| $c_4$ | 17.3 kHz | \cite{27} |
| $g_{Rb}$ | 0.0062 | \cite{27} |
| $g_{Cs}$ | 0.7331(12) | This Work |

scalar nuclear spin-spin constant. The nuclear $g$-factors and shielding coefficients are multiplied together in the Hamiltonian so it is not possible to separate them, and we therefore fit the shielded $g$-factors $g_{Rb} \cdot (1 - \sigma_{Rb})$ and $g_{Cs} \cdot (1 - \sigma_{Cs})$. The resulting values, along with the values of parameters held fixed at theoretical values, are given in Table II.

The fitted hyperfine parameters in Table II are all within 10% of the values predicted from DFT calculations \cite{27}, except for $(eQq)_{Cs}$, which is about 15% larger than calculated. This helps to calibrate the probable accuracy of the calculations for other alkali-metal dimers. The fitted value $c_4 = 19.0(1)$ kHz removes one of the two largest sources of error in our recent determination of the binding energy $D_0$ of $^{87}$Rb$^{133}$Cs in its rovibrational ground state \cite{34}.

We begin by transferring the molecules to an $M_F = 5$ state. Microwaves with $\sigma^+$ polarization drive transitions to $|N,M,F=1,\mu=+1\rangle$. By varying the duration of the microwave pulse, we observe Rabi oscillations to determine the duration of a $\pi$-pulse allowing complete population transfer to the excited rotational state. (c) Two-photon transfer to change the hyperfine level populated in the rovibrational ground state. Once the population has been transferred to the $N = 1$ excited state, we introduce a second microwave pulse with different frequency and polarization from the first. This drives the population back down to a different hyperfine level of the rovibrational ground state than that initially populated.
second microwave field with a different polarization and frequency. We choose to use $\pi$-polarized microwaves to transfer the molecules to the higher-energy of the two $M_F = 4$ levels of $N = 0$ (transition frequency = 980119.14 kHz). At this field, the composition of this final level is $0.947 \pm 4, 0, 1/2, 7/2 \pm 0.321 \pm 4, 0, 3/4, 5/2$ in the uncoupled basis $|N, M_N, m_R, m_L, m_I\rangle$. We observe Rabi oscillations on the second transition by pulsing on the $\sigma^+$-polarized microwaves in between two $\pi$-pulses on the $\sigma^-$-polarized microwave transition as shown in Fig. [3(c)]. Coherent transfer is achieved with a Rabi frequency of $2\pi \times 29.2(3)$ kHz.

In summary, we have performed high-precision microwave spectroscopy of ultracold $^{87}$Rb$^{133}$Cs molecules in the vibrational ground state, and have accurately determined the hyperfine coupling constants for the molecule. Our results confirm that the hyperfine coupling constants calculated by Aldegunde et al. [27] are generally accurate to within $\pm 10\%$, calibrating the probable accuracy of the calculations for other alkali-metal dimers. The resulting understanding of the hyperfine structure enables full control of the quantum state, as illustrated by our demonstration of coherent transfer to a chosen hyperfine state in either the first-excited or ground rotational state. Such complete control is essential for many proposed applications of ultracold polar molecules, and opens the door to a range of exciting future experimental directions, including studies of quantum magnetism [14,15] and novel many-body phenomena [5,40].

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