Creation of Ultracold ^{87}Rb^{133}Cs Molecules in the Rovibrational Ground State

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We report the creation of a sample of over 1000 ultracold ^{87}Rb^{133}Cs molecules in the lowest rovibrational ground state, from an atomic mixture of ^{87}Rb and ^{133}Cs, by magnetoassociation on an interspecies Feshbach resonance followed by stimulated Raman adiabatic passage (STIRAP). We measure the binding energy of the RbCs molecule to be $hc \times 3811.576(1)$ cm^{-1} and the $|v'' = 0, J'' = 0\rangle$ to $|v'' = 0, J'' = 2\rangle$ splitting to be $h \times 2940.09(6)$ MHz. Stark spectroscopy of the rovibrational ground state yields an electric dipole moment of 1.225(3)(8) D, where the values in parentheses are the statistical and systematic uncertainties, respectively. We can access a space-fixed dipole moment of 0.355(2)(4) D, which is substantially higher than in previous work.

The quest for ultracold samples of trapped polar molecules has attracted considerable attention over the last decade [1,2]. The permanent electric dipole moments of polar molecules give rise to anisotropic, long-range dipole-dipole interactions which can be tuned by applied electric fields [3]. This property, combined with the exquisite control of ultracold systems, offers exciting prospects in the fields of quantum controlled chemistry [4,5], precision measurement [6–8], quantum computation [9], and quantum simulation [10,11].

Direct cooling of molecules into the ultracold regime remains elusive, though recent demonstrations of laser cooling show great promise [12–14]. An alternative approach is to form ultracold molecules indirectly by association of precooled atoms [15,16]. To date, the most successful method has employed magnetoassociation on a Feshbach resonance [15,17] to produce weakly bound molecules which are subsequently transferred to the rovibrational ground state by stimulated Raman adiabatic passage (STIRAP) [18]. Although this technique has been successfully applied in homonuclear Cs$_2$ [19] and triplet ^{87}Rb$_2$ [20], experiments exploring the role of dipole-dipole interactions have been confined to the fermionic ^{40}K^{87}Rb molecule [21]. However, KRb molecules are unstable, as the exchange reaction 2KRb → K$_2$ + Rb$_2$ is exothermic [4]. This leads to significant loss of the molecules [22]. Nevertheless, the confinement of the molecules in a three-dimensional optical lattice [23] eliminates this reaction and has allowed pioneering studies of dipolar spin-exchange interactions [24].

Ground-state RbCs molecules offer an appealing alternative to KRb because both the exchange and trimer formation reactions are endothermic [25,26]. The bosonic ^{87}Rb^{133}Cs molecule also offers a contrast with fermionic ^{40}K^{87}Rb. Moreover, the large predicted electric dipole moment of 1.28 D [27] is easily aligned in the laboratory frame, meaning that only modest electric fields are required to realize significant dipole-dipole interactions. ^{87}Rb^{133}Cs molecules have been formed via magnetoassociation in both Innsbruck [28,29] and Durham [30,31]. The Innsbruck group [32] subsequently performed detailed one- and two-photon molecular spectroscopy and, very recently, reported the transfer of molecules to the rovibrational and hyperfine ground state by STIRAP [33].

In this Letter, we demonstrate STIRAP transfer of ^{87}Rb^{133}Cs molecules from a bound state near dissociation to the rovibrational ground state, producing a sample of over 1000 ground-state molecules. In the process we make detailed measurements of the binding energy and the splitting between the $J'' = 0$ and 2 rotational levels of the vibrational ground state ($v'' = 0$). Stark spectroscopy of both the excited and ground states is presented, leading to a precise measurement of the ground-state permanent electric dipole moment. We demonstrate a space-fixed dipole moment that is substantially larger than in previous work.

Details of the apparatus have been described in our studies of dual-species condensates [34,35] and Feshbach spectroscopy [30,31]. In this work we use a nearly degenerate sample of $\sim 2.5 \times 10^5$ ^{87}Rb atoms in the $|f = 1, m_f = 1\rangle$ state and $\sim 2.0 \times 10^5$ ^{133}Cs atoms in the $|3, 3\rangle$ state, confined in the levitated dipole trap illustrated in Fig. 1(a) at a temperature of ~ 300 nK. The near-threshold bound states of ^{87}Rb^{133}Cs relevant to our magnetoassociation sequence are shown in Fig. 1(b). As in [29], these states are labeled as $|n(f_{\text{Rb}}, f_{\text{Cs}})L(m_{f_{\text{Rb}}}, m_{f_{\text{Cs}}})\rangle$, where n is the vibrational label for the particular hyperfine (f_{Rb}, f_{Cs}) manifold,
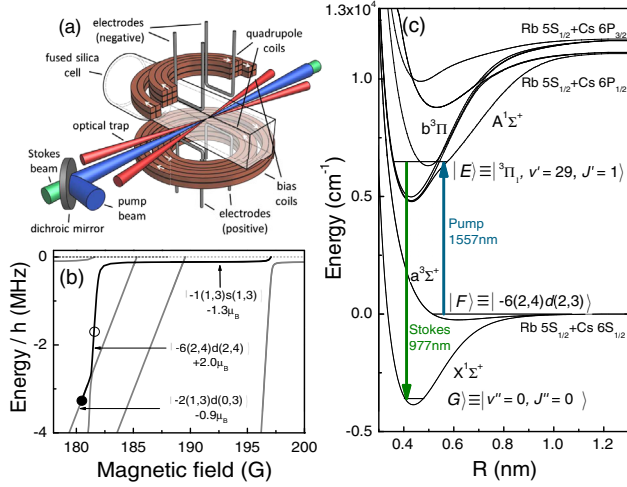
FIG. 1 (color online).   Experimental overview. (a) Simplified diagram of key elements of the apparatus. (b) Near-threshold bound states for ^{87}Rb^{133}Cs and the magnetoassociation path (solid black line). Stern-Gerlach separation is carried out at 180.487(4) G (closed circle) and STIRAP transfer is carried out at 181.624(1) G (open circle). An avoided crossing at ~181.3 G allows transfer between these two states. (c) Potential energy curves for RbCs, indicating the transitions used for STIRAP.
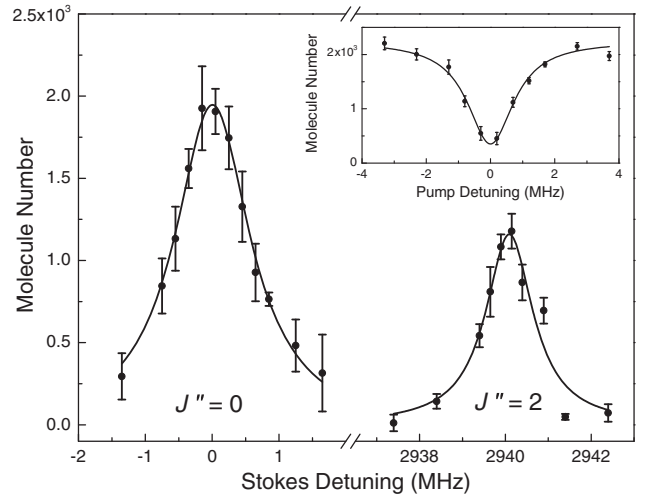

FIG. 2.   Two-photon spectroscopy of the RbCs vibrational ground state. The molecules remain in the initial near-dissociation state when the Stokes light is on resonance with the $J'' = 0$ and $J'' = 2$ rotational states. The solid lines illustrate Lorentzian fits used to determine the resonance positions. Inset: One-photon loss spectrum for the $|^3\Pi_1, v' = 29, J' = 1\rangle$ state. The pump laser is held on resonance with this transition during the two-photon spectroscopy.

counting down from the least-bound state which has $n = -1$, and L is the quantum number for rotation of the two atoms about their center of mass. All states in Fig. 1(b) have $M_{\text{tot}} = 4$, where $M_{\text{tot}} = M_F + M_L$ and $M_F = m_{f_{\text{Rb}}} + m_{f_{\text{Cs}}}$.

To create weakly bound molecules we sweep the magnetic field across a Feshbach resonance at 197.10(3) G to produce weakly bound molecules in the $|-1(1,3)s(1,3)\rangle$ state. These molecules are then transferred to the $|-2(1,3)d(0,3)\rangle$ state at 180.487(4) G following the path shown in Fig. 1(b) and separated from the remaining atoms using the Stern-Gerlach effect [31]. To detect the molecules, we ramp back to a field above the 197.10(3) G resonance to dissociate the molecules to atoms, which are then detected by absorption imaging. We typically create trapped samples of ~2500 molecules in the $|-2(1,3)d(0,3)\rangle$ state with the same temperature as the original atomic sample and a lifetime of 200 ms.

We transfer the molecules from the weakly bound state $|F\rangle$ to the ground state $|G\rangle$ by coupling them via a level $|E\rangle$ of the coupled $A^1\Sigma^+ + b^3\Pi$ manifold. This requires two lasers, labeled the pump ($\lambda = 1557$ nm) and Stokes ($\lambda = 977$ nm) lasers in Fig. 1(c). These are referenced to an optical cavity [36]. Up to 16 mW of light at each wavelength can be focused to a ~35 μm waist at the molecule sample. High transfer efficiency requires a high value of Ω^2/γ for both transitions, where Ω is the Rabi frequency and γ is the natural linewidth. Debatin et al. have identified several states suitable for STIRAP [32]. To locate the states, we pulse 20 μW of pump light, polarized parallel to the magnetic field, on the molecules in the $|F\rangle$ state for 750 μs. We observe the molecule loss as a function of

optical frequency, as shown in the inset to Fig. 2, and locate the center with a Lorentzian fit. In total we have detected seven electronically excited states and their numerous hyperfine sublevels spanning a 1.8-THz range. We focus on the transition to the lowest hyperfine state of the odd-parity (e) component of the $|^3\Pi_1, v' = 29, J' = 1\rangle$ level, which has well-separated hyperfine states and high Ω_P^2/γ [38].

To determine the pump Rabi frequency Ω_P, we set the pump laser on resonance, vary the pulse time t, and fit the fraction of remaining molecules N/N_0 to $N/N_0 = \exp{(-\Omega_P^2 t/\gamma)}$. We observe stronger coupling to the $|^3\Pi_1, v' = 29, J' = 1\rangle$ state from $|-6(2,4)d(2,4)\rangle$ than from $|-2(1,3)d(0,3)\rangle$. Unfortunately, the former has a positive magnetic moment and cannot be magnetically levitated in our current setup. We therefore increase the depth of the optical trap to 12.7 μK, ramp off the magnetic field gradient to transfer to an all-optical trap, and adjust the bias field to transfer the molecules into $|F\rangle \equiv |-6(2,4)d(2,4)\rangle$ at 181.624(1) G [open circle, Fig. 1(b)]. In this trap we observe a pump Rabi frequency of $2\pi \times 0.18(1)$ MHz at maximum power for the transition from $|F\rangle$ to $|E\rangle \equiv |^3\Pi_1, v' = 29, J' = 1\rangle$. An unlevitated trap is advantageous for STIRAP as it removes the variable Zeeman shift across the cloud. However, the transfer to the deeper trap heats the molecular cloud to 1.5(2) μK and we observe a shorter lifetime of 23(2) ms in state $|F\rangle$.

We find the Stokes transition by setting the pump laser on resonance with an increased power of 40 μW, simultaneously pulsing on 16 mW of Stokes light polarized

perpendicular to the magnetic field and scanning the optical frequency. At two-photon resonance, a dark state forms and the state $|F\rangle$ is not excited to the lossy $|E\rangle$ state. This is observed as an increase in the $|F\rangle$ state population if $\Omega_S \gg \Omega_P$, as seen in Fig. 2. We observe both the $J'' = 0$ and $J'' = 2$ levels of the electronic and vibrational ground state, separated by $h \times 2940.09(6)$ MHz. To our knowledge this is the most accurate direct measurement of this splitting. Neglecting centrifugal distortion, this implies a rotational constant $B_0 = 0.0163452(3)$ cm^{-1}, which is consistent with the theoretical prediction of $0.0163(4)$ cm^{-1} [39]. We measure absolute frequencies of 192 572.09 (2) GHz and 306 830.49(2) GHz for the pump and Stokes transitions respectively. This implies a zero-field binding energy of $hc \times 3811.576(1)$ cm^{-1} for the $J'' = 0$ state, relative to the degeneracy-weighted hyperfine centers. This is consistent with the latest theoretical values [39] and experimental measurements [32].

Transfer to the ground state via STIRAP relies on a dark state $|D\rangle$ that is an eigenstate of the system on two-photon resonance. This is composed of a superposition of the $|F\rangle$ and $|G\rangle$ states, $|D\rangle = \cos\theta |F\rangle + \sin\theta |G\rangle$, where the mixing angle θ is defined by $\tan\theta = \Omega_P/\Omega_S$. Transfer from state $|F\rangle$ to $|G\rangle$ (and back) is then achieved by an adiabatic change in the mixing angle, using the pulse sequence shown in the right inset to Fig. 3. The Stokes beam is initially turned on to 7 mW for 20 μs. With $\Omega_S \neq 0$ and $\Omega_P = 0$, $|D\rangle$ is equivalent to the starting state $|F\rangle$. The Stokes beam is then ramped down in 10 μs while the pump beam is ramped up to 16 mW. This adiabatically transfers the population to the ground state $|G\rangle$. We cannot detect the ground state directly, so after a 20-μs hold we reverse the process to transfer back to the initial state, allowing measurement of the square of the one-way efficiency. The maximum efficiency is achieved with both lasers on resonance, as shown in the left inset to Fig. 3. We map out the transfer by truncating the pulse sequence and recording the molecules remaining in the state $|F\rangle$, as shown in Fig. 3 for the on-resonance case.

The polarizations of the pump and Stokes beams drive $\Delta M_{\text{tot}} = 0$ and ± 1 transitions, respectively. The state $|F\rangle$ has $M_{\text{tot}} = 4$, so that we can reach states with $M_{\text{tot}} = 3$ or $M_{\text{tot}} = 5$. Takekoshi *et al.* [33] have shown that the transition to the $M_{\text{tot}} = 5$ hyperfine state has the strongest coupling from $|E\rangle$, with very little population transferred into the $M_{\text{tot}} = 3$ states. Furthermore, theory shows that the $I'' = 5$, $M_{\text{tot}} = 5$ state, where I'' is the total molecular nuclear spin, is the lowest hyperfine state at magnetic fields above about 90 G [40]. We observe only one state (left inset to Fig. 3) which we therefore conclude is the $M_{\text{tot}} = 5$ absolute ground state.

We model the transfer by numerically integrating the Lindblad master equation [36]. The maximum pump Rabi frequency of $2\pi \times 0.18(1)$ MHz is taken from one-photon measurements (see above) and both detunings are set to
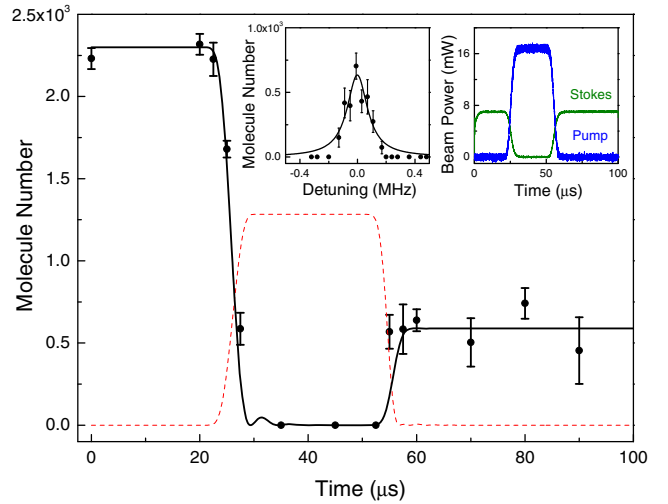

FIG. 3 (color online). The number of molecules remaining in the Feshbach state $|F\rangle$ when both lasers are switched off during the STIRAP sequence. The black solid and red dashed lines show the Feshbach and ground-state populations obtained from the Lindblad model described in the text. Left inset: The final population of Feshbach molecules as a function of Stokes detuning. Right inset: The pump and Stokes beam powers during the STIRAP pulse sequence.

zero. The Stokes Rabi frequency is fitted as a free parameter and thus estimated as $2\pi \times 0.21(1)$ MHz. The results indicate a one-way transfer efficiency of 50% and the model shows that we produce ~ 1250 ground-state molecules (red dashed line in Fig. 3). The efficiency is currently limited by the Rabi frequency achieved for the pump transition. We note that a slower transfer does not increase the efficiency, because of increased laser frequency noise over longer time scales.

The permanent electric dipole moment of a polar molecule is the key quantity of interest for many applications. Without an externally applied electric field, the averaged electric dipole moment in the laboratory frame is zero. Turning on an electric field couples states of opposite parity and hence polarizes the molecules in the direction of the field. In the experiment, we apply the necessary electric field with an array of four electrodes positioned outside the fused silica cell [shown in Fig. 1(a)] [36]. We first measure the dc Stark shift of the pump transition as a function of the applied electric field. The result is shown in the upper inset in Fig. 4; the initially linear response is interrupted by an apparent avoided crossing with higher-lying hyperfine states. By following the avoided crossing, we can then measure the relative shift between the $|E\rangle$ and $|G\rangle$ states (the Stokes shift). As the electric dipole moment of the state $|F\rangle$ is negligible due to the large interatomic separation, the difference between the pump and Stokes shifts yields the dc Stark shift of the rovibrational ground state (shown in Fig. 4).

We fit the Stark shift by calculating the matrix solution of the rigid-rotor Stark Hamiltonian in the laboratory frame
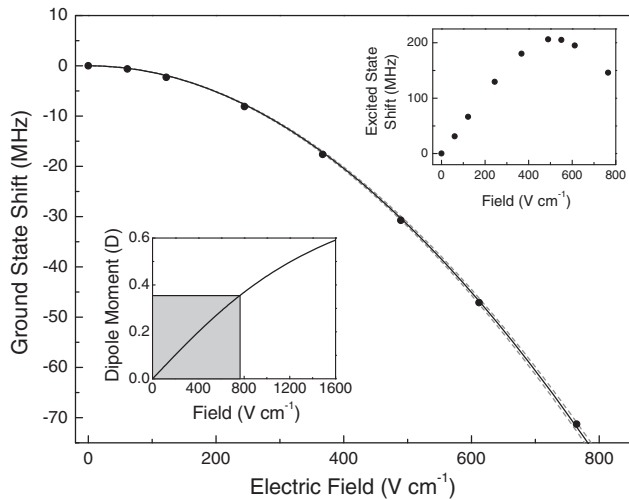
FIG. 4. Stark shift of the rovibrational ground state. The solid black line shows the curve fitted to our results from which we extract a permanent electric dipole moment in the molecular frame of 1.225(3) (8) D. The dotted gray lines indicate the upper and lower bounds due to the systematic error in the electric field calculation. Upper inset: Stark shift of the $|^3\Pi_1, v' = 29, J' = 1\rangle$ excited state used for Stark spectroscopy and STIRAP. The behavior is initially linear with a gradient of approximately 500 kHz/(V cm^{-1}) up to a field of ~400 V cm^{-1}. Lower inset: Ground-state electric dipole moment in the laboratory frame as a function of electric field. The gray region indicates the range of electric dipole moments currently accessible in the experiment.

[36]. We find a permanent electric dipole moment in the rovibrational ground state of 1.225(3)(8) D. The first uncertainty is statistical and the second is systematic, arising from the uncertainty in the electric field [36]. Takekoshi *et al.* recently reported a measured value of 1.17(2)(4) D [33], which agrees with our measurement within their uncertainty. The lower inset in Fig. 4 shows the fitted dc Stark shift converted into the equivalent electric dipole moment in the laboratory frame, and the gray region indicates the dipole moment range currently accessible in the experiment. The maximum laboratory-frame dipole moment we can access is 0.355(2)(4) D at an electric field of 765 V cm^{-1}.

Our results complement those reported recently by Takekoshi *et al.* [33]. The two experiments produce a similar number of molecules in the rovibrational ground state, even though Takekoshi *et al.* demonstrate higher STIRAP efficiencies of 90%. A key difference between the two experiments lies in the trap geometry; our work uses a simple 3D optical trap, whereas Ref. [33] uses a lattice of 2D pancake-shaped traps. A key feature of our work is that we can apply a larger electric field. This allows measurement of the ground-state dipole moment with smaller uncertainties and the realization of larger laboratory-frame electric dipole moments than in [33].

In conclusion, we have presented high-precision spectroscopy of the ground state of ^{87}Rb^{133}Cs molecules and

demonstrated STIRAP transfer to create a sample of over 1000 molecules in the rovibrational ground state. The binding energy of this state is $hc \times 3811.576(1)$ cm^{-1} and the splitting between the $J'' = 0$ and 2 rotational levels of the vibrational ground state is $h \times 2940.09(6)$ MHz. We have used dc Stark spectroscopy to make a precise measurement of the ground-state permanent electric dipole moment as 1.225(3)(8) D, and we demonstrate that laboratory-frame dipole moments up to 0.355(2)(4) D are accessible in our experiment. We believe that this is the largest dipole moment in the laboratory frame accessible in any ultracold molecule experiment to date. For comparison, in ^{87}Rb^{133}Cs, Takekoshi *et al.* access laboratory-frame dipole moments of ~0.03 D [33], while in KRb, Ni *et al.* report values up to 0.22 D [22]. This brings the possibility of observing strong dipolar interactions in a stable ultracold molecular gas within reach.

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