

Spectroscopy and Ground-State Transfer of Ultracold Bosonic $^{39}\text{K}^{133}\text{Cs}$ Molecules

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We report the creation of ultracold samples of $^{39}\text{K}^{133}\text{Cs}$ molecules in their rovibrational ground state. By investigating potentially suitable excited states using one- and two-photon spectroscopy, we have identified a pathway to the ground state via an exceptionally narrow intermediate state. Using stimulated Raman adiabatic passage, we create trapped samples of up to 3500 molecules at temperatures of 1 μK with one-way efficiencies of 71%. The lifetime of these samples is limited by a near-universal two-body loss process, which could shed new light on similar loss mechanisms in other molecular species. Our results are a step toward establishing an alternative molecular species as a platform for the study of bosonic and fermionic quantum matter with strong dipolar interactions.

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Ultracold dipolar molecules have recently emerged as a versatile experimental quantum physics platform [1–4]. Thanks to their long-range dipolar interactions and an accessible manifold of rotational states, which can be used as a pseudospin degree of freedom, they have found applications in the study of many-body dynamics [2,5–7] and show potential as a tool for quantum computation [3,8–10]. In addition, they can be used to study quantum chemistry [4] and perform precision measurements of fundamental constants [11].

Dipolar molecules can either be cooled directly [12–17], or assembled from ultracold atoms [18–27]. The latter approach has achieved very high phase-space densities, leading to the formation of degenerate Fermi gases [28–30] and Bose-Einstein condensates [31]. In order to take full advantage of their properties, dipolar molecules must be prepared in their rovibrational ground state. Highly efficient transfer to deeply bound molecular states can be achieved using stimulated Raman adiabatic passage (STIRAP) [18,32], which has by now been applied to several dipolar species [19–25,27,33].

Some of the referenced molecules [18,19,24,27] undergo exothermic chemical reactions, which intrinsically limit their lifetimes. The ones that do not, however, also exhibit near-universal two-body losses [20,23,33–36]. In some cases these have been proven to stem from photoexcitation

of two-molecule complexes [37,38], but the resulting models fail to predict the behavior of other systems [35,36]. Creating an ultracold sample of a new molecular species would shed more light on this problem [39]. Regardless of their origin, it has been shown that these losses can be suppressed by applying a high dc electric field and confining the molecules to two dimensions [40] or tuning their interactions [41], or by using circularly polarized microwave radiation [42–45], all of which allow the molecules to be evaporatively cooled.

Among the chemically stable molecules, NaK has so far been the only available example with both fermionic and bosonic isotopologues [22,25]. Another such molecule would be KCs, which has a similar permanent dipole moment (1.9 Debye as opposed to 2.7 Debye [46]), but a much larger mass, resulting in a larger dipole length [47] and a lower microwave intensity required for efficient collisional shielding [48]. In addition, KCs exhibits a much higher dynamic polarizability at 1064 nm [49], which would facilitate experiments in optical lattices under a microscope.

In this Letter, we report on the production of bosonic $^{39}\text{K}^{133}\text{Cs}$ molecules in their rovibrational ground state in a 1064-nm optical trap. Using STIRAP, we create up to 3500 molecules at temperatures around 1 μK , with one-way transfer efficiencies of up to 71%. The intermediate state that we use for this purpose has the narrowest natural linewidth among all excited states used for STIRAP of alkalis thus far, measured to be $\Gamma/2\pi = 80(6)$ kHz, which allows adiabatic transfer at lower Rabi frequencies. The polarizability ratio of the ground-state and weakly bound KCs molecules at 1064 nm is 0.93(4), which prevents the transfer from exciting excessive sample oscillations.

The experiments presented here start with an ultracold sample of weakly bound Feshbach molecules [50]. They

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are created from a mixture of ^{39}K and ^{133}Cs atoms in their hyperfine ground states in a 1064-nm single-beam optical dipole trap. We first sweep the magnetic offset $\vec{B}$ over a broad interspecies Feshbach resonance at $|\vec{B}| = 361.6$ G. This causes a fraction of the atom pairs to enter an s -wave bound state with a total angular momentum projection of $M_F = 4$. Once the resonance is crossed, the remaining free atoms are removed from the trap by a force due to a magnetic-field gradient [51]. After the gradient is switched off, we wait until the field settles around $|\vec{B}| = 343$ G, where the bound state is closed-channel dominated, and where the spectroscopy and STIRAP are performed. At the end of the sequence, $|\vec{B}|$ is swept back up across the resonance to break up the Feshbach molecules and allow the constituent atoms to be detected via absorption imaging.

For STIRAP we use two external-cavity diode lasers, later referred to as pump and Stokes laser. They are stabilized with respect to two separate high-finesse optical cavities using the Pound-Drever-Hall scheme [55]. The cavities have finesse of $2.18(2) \times 10^5$ and $2.22(4) \times 10^5$, respectively, and occupy the same spacer, made of ultralow expansion glass, which reduces the relative phase noise. In order to increase the optical power, the pump laser seeds a fiber amplifier, and the Stokes laser seeds a multimode laser diode. The two beams enter the experimental chamber in a nearly copropagating configuration, perpendicular to the quantization axis, which is set by $\vec{B}$. They are both linearly polarized, so depending on the polarization direction they can address either π or σ^+ and σ^- transitions. Their waists are $83(2)$ and $32(2)$ μm , respectively.

Efficient STIRAP requires the identification of a suitable excited molecular state. This state needs to have sufficient overlap with both the Feshbach and ground states, long enough lifetime to allow adiabatic transfer, and sufficiently well-resolved hyperfine structure to avoid destructive interferences [56,57]. The excited states of the KCs molecule have been characterized experimentally using heat-pipe spectroscopy [58–60], but so far direct molecular excitations have not been observed in the ultracold regime. Figure 1 shows the potential energy curves of KCs calculated based on the heat-pipe results [61]. The lowest two potentials in the first electronically excited state, $A^1\Sigma_0$ and $b^3\Pi_\Omega$, are depicted in their bare form, but in reality they are strongly mixed by spin-orbit coupling, which is why transitions to these states are allowed both from triplet Feshbach states and the singlet ground state. In addition, the $b^3\Pi_\Omega$ potential consists of three sublevels ($b^3\Pi_0$, $b^3\Pi_1$, and $b^3\Pi_2$) with different values of Ω , the projection of the total electronic angular momentum onto the molecular axis. If $\Omega = 0$, the Zeeman effect is greatly reduced, and the hyperfine structure usually becomes difficult to resolve experimentally.

We have performed loss spectroscopy on samples of Feshbach molecules by illuminating them with the

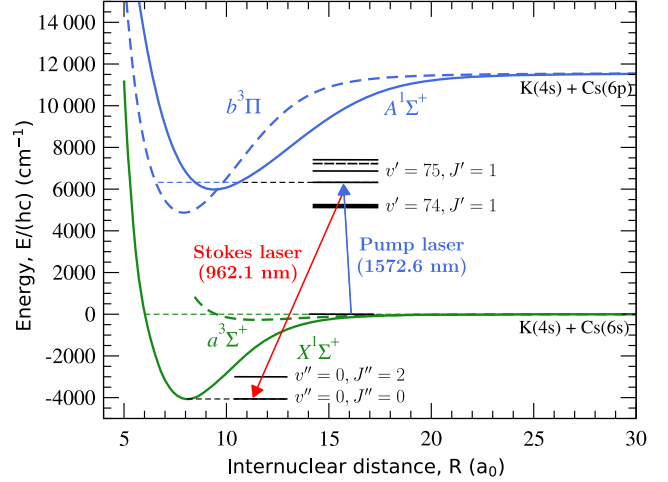


FIG. 1. Calculated potential energy curves of the $^{39}\text{K}^{133}\text{Cs}$ molecule. Black lines show the levels investigated in this Letter (splittings within each potential are not drawn to scale), and the arrows represent the transitions that we use for STIRAP to the rovibrational ground state. Adapted from [61] with permission.

pump laser for 100 μs and measuring their remaining number (Fig. 2). The two groups of loss features that we have identified match the theoretically predicted parameters of two vibrational levels of the spin-orbit coupled $A^1\Sigma_0 - b^3\Pi_\Omega$ potential presented in Table I [62]. In both cases, we can only address the $J' = 1$ angular momentum state ($'$ denotes an electronically excited state), since we are starting from an s -wave Feshbach molecule. As expected, due to its $\Omega = 0$ character, we are unable to resolve any hyperfine sublevels of the $v' = 74$ state [Fig. 2(a)]. This makes it unsuitable for STIRAP. As a consequence, we have focused on the $v' = 75$ state, which has an almost pure $\Omega = 1$ character.

Scanning the frequency of the pump laser in the vicinity of the observed $v' = 75$ state transition has revealed four narrow loss features [Fig. 2(b)]. They are grouped in three energy regions, which can be assigned to the three different projections (M_J) of the angular momentum J' . We fit each of these features to an analytical model of the form [63]

$$N(t) = N_0 \exp[-t\Omega_p^2\Gamma/(\Gamma^2 + 4\Delta_p^2)], \quad (1)$$

where N_0 is the initial molecule number, t is the laser exposure time, Ω_p is the angular Rabi frequency of the pump laser, Γ is the spontaneous decay rate of the excited state, and Δ_p is the detuning of the pump laser from resonance in angular units. From this fit, we extract the natural linewidth $\Gamma/2\pi$ of each level and calculate the corresponding Rabi frequencies using an independent measurement of the on-resonance lifetime [51]. Table II summarizes the results of these calculations: the small variation in $\Gamma/2\pi$ could be explained by weak mixing with other J' levels [57].

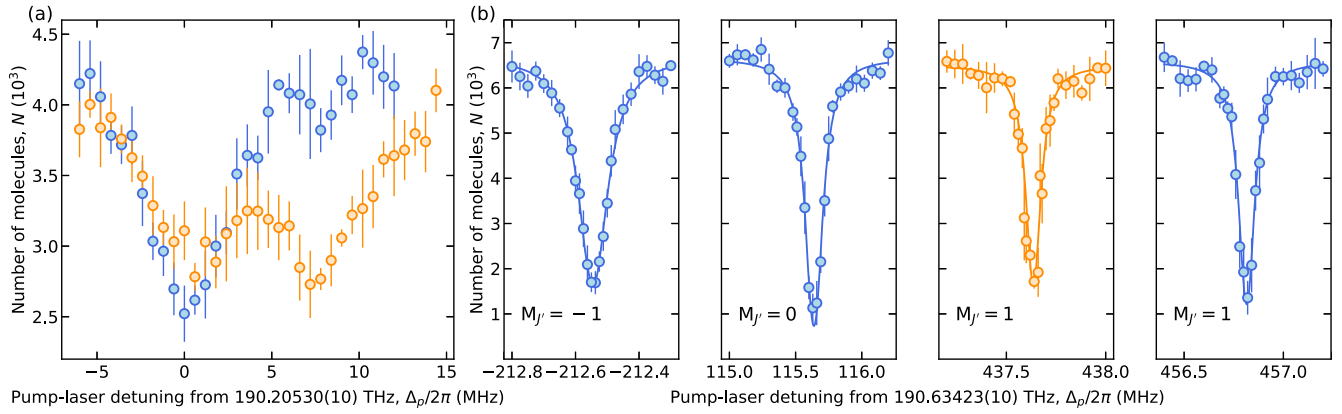


FIG. 2. Loss spectroscopy of the $^{39}\text{K}^{133}\text{Cs}$ excited states. All measurements are performed at $|\vec{B}| = 343.03(6)$ G by exposing the sample to the pump-laser beam for 100 μs , and the laser power is adjusted to achieve a similar on-resonance lifetime for each state. Blue (orange) points are taken with the beam's polarization perpendicular (parallel) to $\vec{B}$. Unless stated otherwise, the error bars in each figure reflect the standard deviation from five experimental runs. (a) $v' = 74$ state. The shapes of the observed loss features suggest that they comprise multiple unresolved hyperfine components. (b) Hyperfine-resolved spectroscopy of the $v' = 75$ state. The solid lines are fits to the analytical model discussed in the text, yielding the linewidths and Rabi frequencies presented in Table I.

A finite admixture of the $A^1\Sigma_0$ potential in the $v' = 75$ state makes transitions to the $X^1\Sigma_0$, $v'' = 0$ manifold allowed ($''$ denotes a singlet electronic ground state). We are therefore able to measure the frequencies of these transitions by shining the Stokes laser while the pump laser is locked on resonance and observing a revival of the molecular population when the two-photon resonance is reached. Among the hyperfine sublevels of $v' = 75$ that we have identified, the strongest coupling to $X^1\Sigma_0$, $v'' = 0$ is exhibited by the $M_{J'} = -1$ state, and the parity selection rule allows transitions from it to both the $J'' = 0$ and $J'' = 2$ levels in the ground state. We have identified two groups of resonances separated by 5.476 GHz [Figs. 3(a) and 3(b)], which is consistent with the splitting expected between $J'' = 0$ and $J'' = 2$ according to the most recent calculation of the KCs rotational constant at equilibrium bond length of $B_e = h \times 914.014(21)$ MHz [64]. Within the $J'' = 0$ state [Fig. 3(a)], we are able to detect only one peak at 311.59709(5) THz, using a beam polarized

perpendicularly to $\vec{B}$. In $J'' = 2$ [Fig. 3(b)], which we investigated with the beam polarized parallel to $\vec{B}$, we are able to resolve three hyperfine levels, which is not enough to unambiguously assign their quantum numbers. The ground-state rotational constant extracted from this measurement is $B_0 = h \times 912.68(18)$ MHz, which is reasonably close to the predicted B_e . The uncertainty is limited by our lack of knowledge of the hyperfine quantum numbers [51].

In order to extract the strength of the detected transition to $J'' = 0$, we have performed an electromagnetically induced transparency [65] measurement by keeping the Stokes laser locked on resonance and scanning the frequency of the pump laser [Fig. 3(c)]. We fit the data to a numerical master-equation model that takes into account the excited-state linewidth, as well as a finite decoherence

TABLE I. Comparison of the predicted and measured properties of two vibrational states of the $A^1\Sigma_0 - b^3\Pi_\Omega$ potential of the $^{39}\text{K}^{133}\text{Cs}$ molecule [62]. Here, v' is the vibrational quantum number, $A^1\Sigma_0$, $b^3\Pi_0$ and $b^3\Pi_1$ are the contributions of these three electronic states, TDM is the predicted transition dipole moment in units of $10^{-4}ea_0$, and E_{th} and E_{exp} are the predicted and measured transition energies from the initial Feshbach state at $|\vec{B}| = 343$ G in units of $h \times \text{THz}$. The angular momentum J' is equal to 1 for both states.

v'	$A^1\Sigma_0$	$b^3\Pi_0$	$b^3\Pi_1$	TDM	E_{th}	E_{exp}
74	0.824	0.176	$<10^{-5}$	3.96	190.2053	190.20530(10)
75	$<10^{-5}$	$<10^{-5}$	1	4.92	190.6342	190.63423(10)

TABLE II. Properties of the observed hyperfine levels of the $A^1\Sigma_0 - b^3\Pi_\Omega$, $v' = 75$ state. Here, p is the pump laser polarization with respect to the quantization axis, $M_{J'}$ is the projection of the angular momentum J' , TDM_{th} and TDM_{exp} are the predicted and measured transition dipole moments in units of $10^{-4}ea_0$, $\Omega_0/2\pi$ is the measured intensity-normalized Rabi frequency in units of $\text{kHz} \times \sqrt{I/\text{mW cm}^{-2}}$, and $\Gamma/2\pi$ is the measured natural linewidth in units of kHz. The values of $\Omega_0/2\pi$ measured for polarization perpendicular to $\vec{B}$ have been multiplied by 2 to obtain the value expected for the optimal (circular) polarization.

p	$M_{J'}$	TDM_{th}	TDM_{exp}	$\Omega_0/2\pi$	$\Gamma/2\pi$
$\parallel$	1	4.92	2.8(2)	0.31(2)	65(6)
$\perp$	1	4.92	4.3(4)	0.48(4)	65(8)
$\perp$	0	4.92	3.0(2)	0.34(3)	96(9)
$\perp$	-1	4.92	5.8(4)	0.65(4)	80(6)

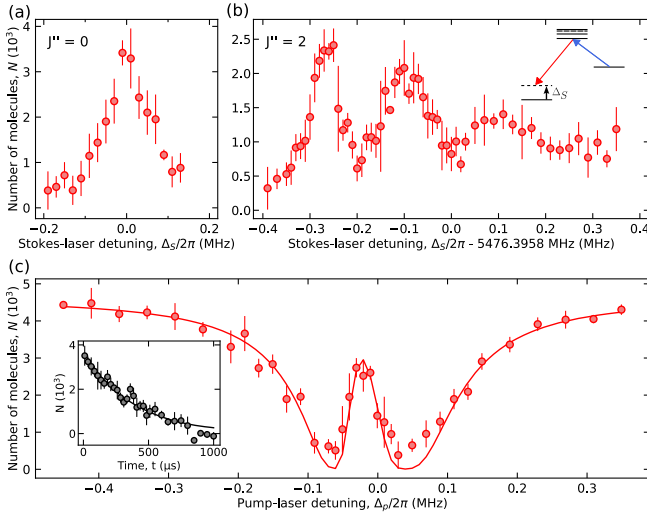


FIG. 3. Two-photon spectroscopy of the $^{39}\text{K}^{133}\text{Cs}$ ground states. Each measurement is taken by exposing the sample to both laser beams for 100 μ s, with the pump beam tuned to the $v' = 75$, $M_{J'} = -1$ state. (a) Frequency scan of the Stokes beam, polarized perpendicularly to $\vec{B}$, near the $v'' = 0$, $J'' = 0$ state. Zero detuning corresponds to 311.59709(5) THz. (b) Frequency scan of the Stokes beam, polarized parallel to $\vec{B}$, near the $v'' = 0$, $J'' = 2$ state. We are able to resolve three unidentified hyperfine levels. (c) Dark resonance spectrum obtained by keeping the Stokes laser on resonance with the $v'' = 0$, $J'' = 0$ state and scanning the pump-laser frequency. The solid line is a fit on the basis of a numerical master-equation model. Zero detuning is defined by the transition to the excited state and is extracted from the fit. The inset shows the number of molecules as a function of laser exposure time on two-photon resonance, where the solid line is a fit based on the model used in the main figure, but with Rabi frequencies and detunings fixed. The error bars in the inset reflect the standard deviation from three experimental runs.

rate between the initial and final states [51]. The fit yields a normalized Rabi frequency of $\Omega_0/2\pi = 0.52(5)$ kHz $\times \sqrt{I/\text{mW cm}^{-2}}$ (assuming the optimal, circular polarization), which gives a transition dipole moment of $4.7(4) \cdot 10^{-4} e a_0$ (the predicted value is $4.75 \cdot 10^{-4} e a_0$ [62]). From a parallel measurement of the lifetime of molecules on the two-photon resonance [inset in Fig. 3(c)] we extract an effective decoherence timescale of 25(2) μ s.

Having identified the necessary transitions, we proceed to transfer the molecules to the rovibrational ground state using STIRAP. The pulse sequence used [Fig. 4(a)], usually referred to as “counterintuitive,” requires the Stokes laser to be switched on before the pump laser. The transfer to the ground state takes place as the pump laser power is being ramped up while the Stokes laser power is being ramped down, which causes the state of the molecules to adiabatically change character from the Feshbach state to the ground state. This process takes around 10 μ s and is then reversed to allow the detection of remaining molecules. Figure 4(b) shows the evolution of the observed number of

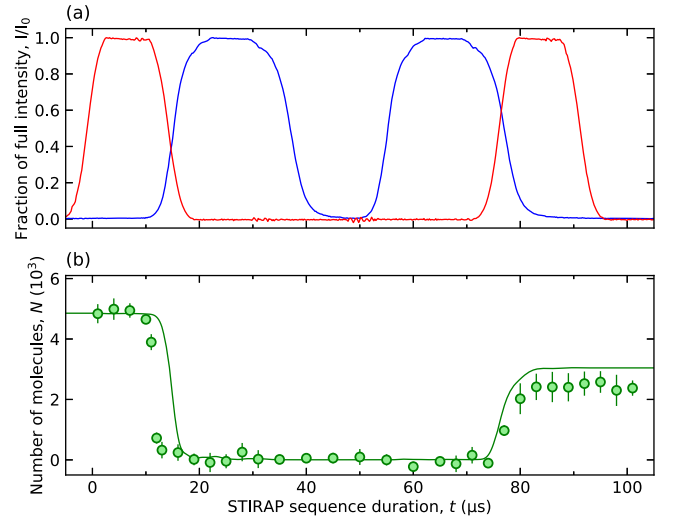


FIG. 4. STIRAP sequence. (a) Intensities of the pump beam (blue) and the Stokes beam (red) as a function of time, recorded by photodiodes and averaged over nine experimental runs. (b) Number of Feshbach molecules observed at different points of the pulse sequence by abruptly switching both beams off. The solid line is the result of a master-equation simulation, in which the only free parameter is the initial number of molecules (see text for details).

molecules as a typical STIRAP pulse sequence proceeds. The peak Rabi frequencies used were $\Omega_p/2\pi = 342(19)$ kHz for the pump beam and $\Omega_S/2\pi = 307(16)$ kHz for the Stokes beam. The ratio of the final and initial numbers allows us to estimate the one-way transfer efficiency to be around 71%. The typical temperature of the molecules is around 1 μ K at this stage and we do not observe any significant heating caused by the STIRAP pulses.

To improve our understanding of the transfer efficiency, we perform a master-equation simulation using the same model as for the fit in Fig. 3(c) [solid line in Fig. 4(b)], in which the only free parameter is now the initial number of molecules, and the decoherence rate is assumed to be independent of the laser power. According to the results, increasing the available Stokes-laser power from the current 28 to 100 mW would be enough to reach efficiencies of 90% [51]. Further improvement would most likely require the use of one of the filtering techniques [66,67] to suppress the effect of laser noise [68].

One of the most important properties of a trapped sample of ground-state molecules is its lifetime. Before measuring it, in order to make sure that the molecules do not spread out along the optical trap, we add a perpendicular beam of a larger waist, which results in trap frequencies of 10.6(1), 168(4), 168(4) Hz. We measure a polarizability of 78(11) Hz/(W/cm²) at 1064 nm in the ground state, which is consistent with the prediction of 77.83 Hz/(W/cm²) [49], and is only a factor of 0.93(4) smaller than for the Feshbach molecules [51]. The temperature under these conditions is 830(89) nK, giving a peak

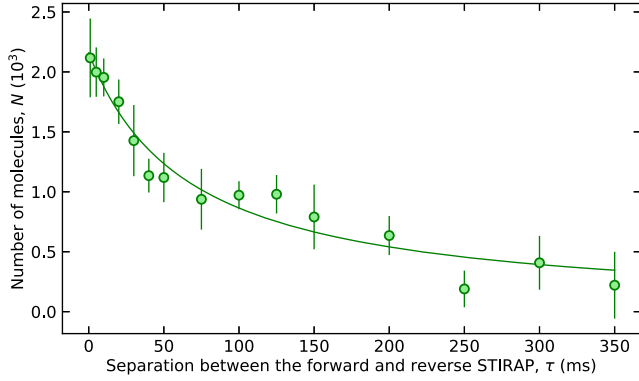


FIG. 5. Loss of $^{39}\text{K}^{133}\text{Cs}$ molecules from a crossed optical trap. The initial peak density is $5.4(15)_{\text{sys}}(8)_{\text{stat}} \times 10^{10} \text{ cm}^{-3}$. The solid line is a fit to a two-body decay function, yielding a loss coefficient of $8(2)_{\text{sys}}(2)_{\text{stat}} \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$.

density of $5.4(15)_{\text{sys}}(8)_{\text{stat}} \times 10^{10} \text{ cm}^{-3}$. We then vary the separation τ between the forward and reverse STIRAP, and measure the final number of molecules (Fig. 5). Because of a significant drift of the magnetic field in our experimental chamber, we have to adjust the relative detuning of the STIRAP lasers for each pulse separation [51]. Assuming that the decay is purely two-body, we extract a loss coefficient of $8(2)_{\text{sys}}(2)_{\text{stat}} \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ [51], which is close to the predicted universal loss rate of $2.8 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ [69]. Such fast two-body losses could be caused by a photo-induced loss mechanism [34,37] or by hyperfine-changing collisions [20], but evaporation from the trap may also be contributing here.

In conclusion, we have characterized two distinct excited states of the $^{39}\text{K}^{133}\text{Cs}$ molecule and the transitions from one of them to the rovibrational ground state. Using STIRAP, we have prepared 3500 ground-state molecules at a temperature of $1 \mu\text{K}$. Thanks to the exceptionally narrow linewidth of the excited state that we have used for this transfer [$\Gamma/2\pi = 80(6) \text{ kHz}$], we have been able to achieve efficiencies up to 71% with lower Rabi frequencies than in other experiments. The identified states are a good starting point for further spectroscopic study of KCs, which would allow a more accurate calculation of its hyperfine parameters. Similarly to other chemically stable molecules, KCs undergoes two-body collisional losses, so it could provide new insights into the details of this process. In the future, we are planning to test existing shielding techniques [40,41,45,48,70] on ultracold KCs samples, which would make them a new tool for the study of bosonic dipolar quantum matter, in particular under a microscope, with the possibility of a direct comparison to fermions.

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Data availability—The data that support the findings of this article are openly available [71].

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