

Formation of ultracold $^{39}\text{K}^{133}\text{Cs}$ Feshbach moleculesCharly Beulenkamp^{1,*}, Krzysztof P. ZamarSKI^{1,*}, Robert C. Bird,² C. Ruth Le Sueur², Jeremy M. Hutson²,
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We report the creation of an ultracold gas of bosonic $^{39}\text{K}^{133}\text{Cs}$ molecules. We first demonstrate a cooling strategy relying on sympathetic cooling of ^{133}Cs to produce an ultracold mixture. From this mixture, weakly bound molecules are formed using a Feshbach resonance at 361.7 G. The molecular gas contains $7.6(10) \times 10^3$ molecules with a lifetime of about 130 ms, limited by two-body decay. We perform Feshbach spectroscopy to observe several new interspecies resonances and characterize the bound state used for magnetoassociation. Finally, we fit the combined results to obtain improved K-Cs interaction potentials. This provides a good starting point for the creation of ultracold samples of ground-state $^{39}\text{K}^{133}\text{Cs}$ molecules.

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I. INTRODUCTION

Magnetic-field-tunable Feshbach resonances [1] are a powerful tool in experiments on ultracold gases. In addition to offering a simple and reliable way to tune atomic collisions, they serve as a bridge between the high level of control in ultracold atomic gases and the rich physics of molecules. Formation of weakly bound molecules via Feshbach resonances [2,3] has led to the observation of molecular Bose-Einstein condensates (BEC) [4–6] and the study of the BEC-BCS crossover [7–9]. Further exploration into ultracold molecule physics was made possible by transferring to deeply bound molecular states by means of stimulated Raman adiabatic passage (STIRAP) [10,11]. Of particular interest in the last decade have been heteronuclear molecules. These have an electric dipole moment in their ground state, giving rise to long-range dipole-dipole interactions with applications in quantum computing [12,13], study of new quantum phases [14] and quantum simulation of spin models [15,16], including t-J models [17] predicted to help explain high-temperature superconductivity [18]. Gases of dipolar molecules have been brought into the degenerate regime, with degenerate Fermi gases created directly from degenerate atomic gases [19–21], and a BEC of bosonic NaCs molecules [22] being realized through evaporative cooling of molecules using microwave shielding of collisions [23].

Ultracold gases of heteronuclear Feshbach molecules have in most cases been created using two different alkali atoms, with over half of the possible combinations realized [24–31]. Notably missing from the list of bialkali molecules is KCs, which offers an electric dipole moment of 1.9 D [32] in its rovibrational ground state and both fermionic and bosonic isotopologues. Previous work has predicted Feshbach resonances for different isotopic combinations of K and Cs [33] and observed a few of the corresponding atom-loss peaks [34], but not achieved molecule formation. Here we report the creation of a trapped ultracold sample of $^{39}\text{K}^{133}\text{Cs}$ Feshbach molecules by magnetoassociation, providing the necessary starting conditions for the creation of ground-state molecules by STIRAP [35]. In Sec. II we describe our experimental procedure for preparing an ultracold mixture. In Sec. III we discuss the creation and purification of Feshbach molecules. We perform Feshbach spectroscopy (Sec. IV) to observe several new resonances at four different thresholds and characterize the bound state used for magnetoassociation by two-photon bound-state spectroscopy and magnetic-moment measurements (Sec. V). Finally, in Sec. VI we fit the combined results to obtain improved K-Cs interaction potentials that provide a detailed understanding of the collision physics.

II. COOLING SEQUENCE

Association of atom pairs to form heteronuclear Feshbach molecules requires the preparation of an ultracold mixture of the two species with at least one species close to degeneracy [36]. The collisional properties of ^{133}Cs and ^{39}K make them more difficult to cool than other alkalis, and they were among the last alkali isotopes to be brought to degeneracy [37,38]. In previous work, we used established techniques to cool the two species sequentially before mixing [34]. This scheme had the advantage of allowing experiments on either single-species BEC or mixtures. It could in principle be used

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to produce a mixture at high enough phase-space density for magnetoassociation, but required long cycle times and significant experimental complexity. To simplify our experimental sequence we explored a cooling strategy similar to those used for RbCs [39] and NaCs [22], where the two species are mixed throughout the cooling process and Cs is cooled sympathetically. These experiments rely on radiofrequency (RF) evaporation of the lighter atom in a magnetic trap to load the mixture into an optical dipole trap. This strategy is not viable for the case of ^{39}K , as its background scattering length of $-35\ a_0$ leads to a Ramsauer-Townsend minimum in the collisional cross section at collision energies around $400\ \mu\text{K}$ [40]. Instead, we use an established technique for preparing ^{39}K BEC [41], with modifications to allow the ^{39}K gas to cool Cs sympathetically. In the following we describe our cooling scheme, and detail how to manage the sympathetic cooling of Cs.

Our setup has been previously described in detail in Ref. [44]. It consists of a steel chamber fed with precooled atoms from separate 2D^+ MOTs [45] of ^{39}K and ^{133}Cs (referred to as K and Cs in the remainder of Secs. II and III). We start our experimental sequence by loading a dual-species MOT with typical atom numbers of 3×10^8 for K and 2×10^7 for Cs. After loading, the MOT is compressed by increasing the magnetic field gradient from $9.5\ \text{G/cm}$ to $21\ \text{G/cm}$ while reducing the repumping intensities. We then shut off the gradient and apply D1-line gray molasses [46] on K and D2-line molasses on Cs to cool to temperatures of $13(1)\ \mu\text{K}$ and $14.2(2)\ \mu\text{K}$, respectively, followed by optical pumping of K to $(f, m_f) = (1, -1)$ [47]. The mixture is then trapped in a quadrupole magnetic trap with an initial magnetic field gradient of $35\ \text{G/cm}$, trapping only atoms in the states $(1, -1)_\text{K}$ and $(3, -3)_\text{Cs}$. The trap is held at this gradient for $100\ \text{ms}$ to ensure spin purity of the trapped gas, followed by a ramp up to $65\ \text{G/cm}$. We then shine a deep optical dipole trap (ODT) (23W , $30\ \mu\text{m}$ waist, $1064\ \text{nm}$) near the center of the magnetic trap. The trap depth for K of $U_\text{K} = 2.1\ \text{mK}$ ensures that typical collision energies overcome the Ramsauer-Townsend minimum [41]. After $2.5\ \text{s}$ of loading, we switch off the magnetic trap. After equilibration the ODT typically contains $1.6(1) \times 10^7$ K atoms and $4.5(1) \times 10^5$ Cs atoms at temperatures of $388(4)\ \mu\text{K}$ and $394(4)\ \mu\text{K}$.

The mixture can now be further cooled by evaporation near an intraspecies Feshbach resonance for K. The interspecies scattering length takes values between $60\ a_0$ and $80\ a_0$ away from interspecies Feshbach resonances, ensuring sufficiently high elastic collision rates for the two species to thermalize. The ratio of trap depths, $U_\text{Cs}/U_\text{K} = 1.94$ at $1064\ \text{nm}$, suppresses direct evaporation of Cs. Cs is therefore sympathetically cooled by K. Since we load Cs in the state $(3, -3)_\text{Cs}$ into a tight trap, we have to consider collisional loss processes, which can inhibit efficient sympathetic cooling. Dipolar relaxation couples the spins of a colliding atom pair to their relative angular momentum, resulting in heating and depolarization. Additionally, the typically large scattering length of Cs results in high three-body recombination rates. Both two-body and three-body loss rates have an upper bound in the unitary limit $ka \gg 1$, where k is the wave number of a colliding pair and a is the scattering length. The limiting of the loss rate in the unitary regime can be seen in loss spectroscopy of the Cs

cloud at different temperatures [Fig. 1(a)]. Loss rates are high near zero crossings of the scattering length, and become more pronounced at lower temperatures [48]. At magnetic fields between $150\ \text{G}$ and $160\ \text{G}$ and temperatures above $10\ \mu\text{K}$, the Cs loss rates remain low enough for efficient sympathetic cooling. The scattering length for K can be tuned using a broad Feshbach resonance at $162\ \text{G}$, making it suitable for the first step of evaporative cooling.

In addition to loss due to Cs-Cs collisions, we observe a one-body decay of the Cs atom number with a rate proportional to the ODT depth. To separate one-body loss from two-body and three-body loss in the Cs cloud, we reduce the density to below $2 \times 10^{11}\ \text{cm}^{-3}$ and set the magnetic field to $150\ \text{G}$. Figure 1(b) shows the decay rate as a function of the trap light intensity averaged over the Cs cloud, expressed in terms of the Cs trap depth. We interpret the trap-depth-dependent decay as depolarization of Cs atoms due to scattering of trap light, followed by exothermic spin-changing collisions. Scattering of trap photons can leave the atom in the same internal state (Rayleigh scattering), or a different one (Raman scattering) [49]. Laser light with an intensity I_L , frequency ω_L and polarization unit vector $\mathbf{u}_L$ induces transitions from an initial state g to a final state g' at a rate that can be expressed in terms of the atomic polarizability as (see Supplemental Material [50]):

$$\Gamma_{g \rightarrow g'} = I_L \frac{\omega_L^3}{3\pi \hbar c^4 \epsilon_0^2} \left| \langle g' | \alpha_{nJ}^s \mathbf{u}_L - i \alpha_{nJ}^v \frac{\mathbf{u}_L \times \hat{\mathbf{J}}}{2J} | g \rangle \right|^2, \quad (1)$$

where $\hat{\mathbf{J}}$ is the total electronic angular momentum operator of the atom, while α_{nJ}^s and α_{nJ}^v are the scalar and vector polarizabilities, respectively. Due to its large fine-structure splitting relative to the trap-light detuning from the D1 and D2 lines, Cs has a significant vector polarizability at $1064\ \text{nm}$ of $\alpha^v = -200\ a_0^3$ [51], which results in a higher Raman scattering rate at this wavelength than for other alkalis. Most Cs atoms that change state end up in levels with $f = 4$, after which they can undergo exothermic two-body collisions with K, leading to atom loss. We compute a corresponding loss rate by assuming that every Raman scattering event results in the loss of the atom and find good agreement between calculated and measured loss rates. The depolarizing effect of the trap light will affect not just evaporative cooling, but also the ODT loading process. The ODT depth must be sufficiently high to load K atoms, but a further increase of trap depth will result in fast decay of loaded Cs atoms, limiting the amount of Cs that can be loaded.

Having characterized the loss processes for Cs, we now proceed with evaporative cooling in the ODT. We start evaporation at $150\ \text{G}$ by reducing the ODT power from $23\ \text{W}$ to $1\ \text{W}$ in $250\ \text{ms}$. We then hold the cloud for $250\ \text{ms}$ at $114\ \text{G}$ to remove spin impurities. At this field, impurities in state $(1, 0)_\text{K}$ (probably originating from spin-relaxation collisions between K atoms during ODT loading) are quickly depleted by three-body recombination due to a Feshbach resonance for the collisions with atoms in $(1, -1)_\text{K}$. Cs spin impurities undergo exothermic spin exchange with $(1, -1)_\text{K}$ atoms, which ejects them from the trap.

Beyond this point, evaporative cooling using the $(3, -3)_\text{Cs}$ state becomes inefficient due to high two-body and three-body

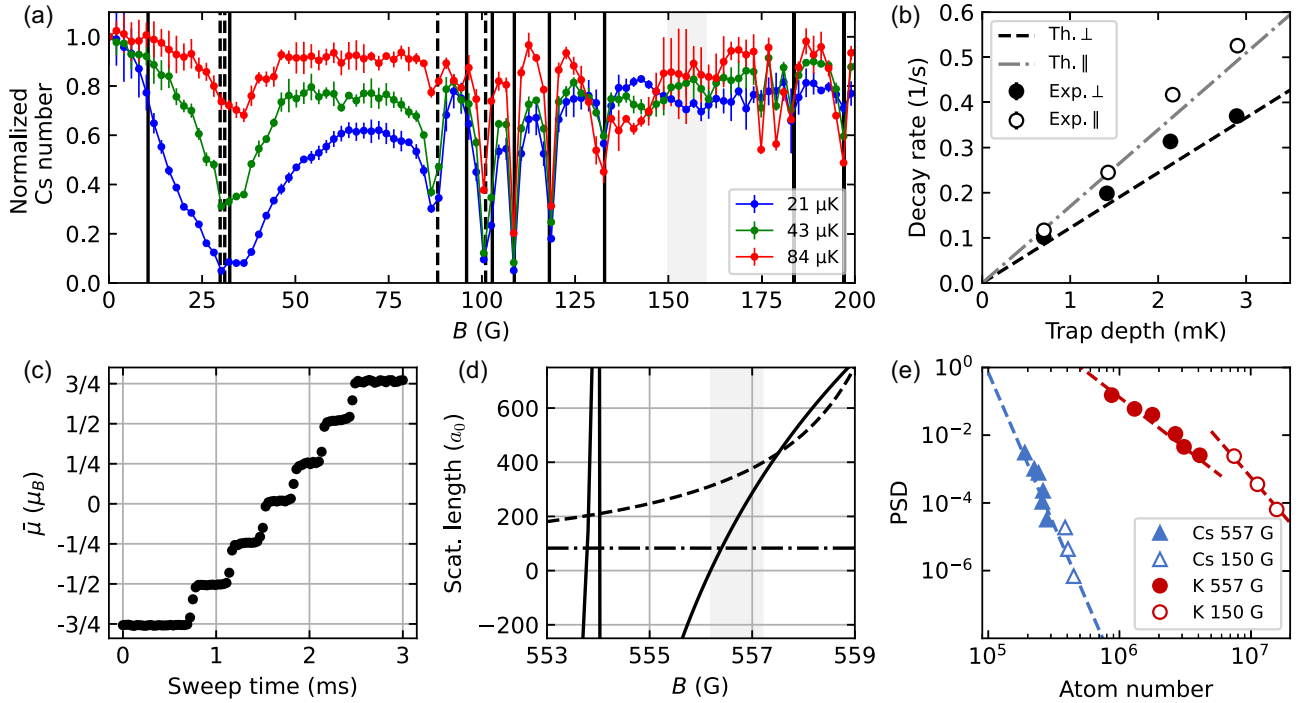


FIG. 1. Cooling of the mixture. (a) Loss spectroscopy of Cs in the state $(3, -3)$. We plot the fraction of remaining Cs atoms after a 1 s hold time at initial peak density $1.5(5) \times 10^{12} \text{ cm}^{-3}$ and temperatures 84 μK (red), 43 μK (green), and 21 μK (blue) as a function of the magnetic field B . Error bars are the standard deviations from three experimental runs. Solid (dashed) vertical lines mark peaks (zero crossings) in the imaginary (real) part of the scattering length [42]. The shaded area marks a magnetic field range over which the K scattering length can be tuned from $50 a_0$ to $300 a_0$ [43]. (b) Cs decay rate as a function of ODT depth for polarizations perpendicular (filled dots) and parallel (hollow dots) to the magnetic bias field. Error bars are smaller than the plot markers. The dashed (dash-dotted) line is the predicted Raman scattering rate for perpendicular (parallel) polarization. (c) Stern-Gerlach measurements of the average magnetic moment of the Cs cloud during the trap-mediated Raman transfer. Error bars correspond to the uncertainty in the cloud position as determined by fitting a Gaussian distribution to the density distribution, and are smaller than the plot markers. (d) Scattering lengths of $(1, -1)_K$ (dashed) [43], $(3, 3)_{\text{Cs}}$ (solid) [42], and $(1, -1)_K + (3, 3)_{\text{Cs}}$ (dash-dotted) [34] near 557 G, as a function of the magnetic field B . The shaded area marks the region where condensates of K and Cs are both miscible and stable. (e) Phase-space density (PSD) as a function of atom number for K (dots) and Cs (triangles) during evaporative cooling in the ODT at 150 G (hollow) and 557 G (filled). The dashed lines indicate evaporation efficiencies of 3 and 5 for K and 9 for Cs.

Cs-Cs loss rates, so we proceed in the channel $(1, -1)_K + (3, 3)_{\text{Cs}}$ at 557 G. As there is no way to couple the states $(3, -3)_{\text{Cs}}$ and $(3, 3)_{\text{Cs}}$ directly, spin transfer requires briefly populating intermediate states. These intermediate states suffer from much higher collisional loss rates, so time spent in them should be minimized. Achieving high Rabi frequencies using RF coupling is complicated by constraints to antenna geometry and RF pickup in surrounding electronics. Instead, we exploit the large vector polarizability of Cs at 1064 nm and drive Raman transitions using our trapping light. We switch on a second beam, labeled RODT, copropagating with the ODT, with orthogonal polarization and a detuning of 41 MHz. By sweeping the magnetic field up by 10 G in 3 ms, we drive a series of adiabatic passages. The spin transfer is shown in Fig. 1(c). We switch off RODT at various times during the magnetic field sweep and perform Stern-Gerlach separation of the Cs cloud. We fit a Gaussian function to the number density of the cloud to determine its center-of-mass position. From the position of the cloud we extract an average magnetic moment $\bar{\mu}$, which can be seen to increase in steps of one quarter of the Bohr magneton μ_B .

Following the spin transfer of Cs, we immediately increase the magnetic field to 557 G to avoid interspecies spin-exchange collisions that can occur below 355 G. We proceed with evaporative cooling at this magnetic field, reducing the ODT power from 1 W to 50 mW in 1000 ms. The interspecies and intraspecies scattering lengths in this magnetic-field region are depicted in Fig. 1(d). A Cs Feshbach resonance at 548 G [42,52] and K resonance at 561 G [43] result in a window 1.5 G wide where both species can be efficiently cooled and condensates are miscible. The interspecies scattering length in this window is approximately $83 a_0$, while the intraspecies scattering length for K is above $350 a_0$ at the Cs zero crossing. As the evaporation progresses and the mixture gets colder, interspecies thermalization slows down while the three-body loss of K speeds up. Evaporation ramps are optimized empirically by maximizing the number of molecules created by magnetoassociation as discussed below.

To judge the quality of our evaporative cooling scheme, we measure the phase-space densities (PSD) as a function of atom numbers, as depicted in Fig. 1(e). The efficiency of

evaporative cooling can be quantified by the parameter $\gamma = -\frac{d \log \rho}{d \log N}$, where ρ is the phase-space density and N is the atom number. For K we find $\gamma \approx 5$ initially, dropping to $\gamma \approx 3$ at 557 G. For Cs we find $\gamma \approx 9$, which indicates efficient sympathetic cooling. At the end of evaporation our mixture consists of $8.7(3) \times 10^5$ K atoms at a PSD of 0.15(2), and $1.9(1) \times 10^5$ Cs atoms at a PSD of 0.0031(4). The clouds remain well overlapped, as the estimated differential gravitational sag of $1.4 \mu\text{m}$ is small compared to the computed $1/e$ cloud radii of $9.1 \mu\text{m}$ and $7.6 \mu\text{m}$ for K and Cs, respectively.

In an unbalanced mixture, the molecule creation efficiency $N_{\text{mol}}/N_{\text{atom}}$ with respect to the minority component depends primarily on the phase-space density of the majority component [36]. Cooling K closer to degeneracy would therefore increase the molecule creation efficiency with respect to Cs. However, evaporative cooling beyond the point described in the previous paragraph becomes very inefficient for Cs, because a tilt of the trap potential due to gravity lowers the trap depth for Cs below that of K. The loss of Cs atoms results in a lower molecule number, negating the gains in molecule creation efficiency from the increased phase-space density of K.

III. MOLECULE FORMATION

We now proceed to the creation of weakly bound molecules. Previous experiments have observed six Feshbach resonances between K atoms in states with $f = 1$ and Cs atoms in the state $(3, 3)_{\text{Cs}}$ [34]. Of these, the most promising for molecule creation is the resonance at 361.7 G in the channel $(1, 1)_{\text{K}} + (3, 3)_{\text{Cs}}$. This resonance has the largest predicted width, and as both species are in their hyperfine ground states, molecular states below this threshold do not have internal decay channels. To avoid heating and atom loss due to the large background scattering length of Cs, we have to minimize the time between the end of evaporative cooling and molecule creation. The magnetic-field changes during molecule formation are depicted schematically in Fig. 2(a). First we jump our magnetic field to 377 G in order to avoid a broad Feshbach resonance at 395 G for atoms in state $(1, 1)_{\text{K}}$. The K atoms are then transferred from $(1, -1)_{\text{K}}$ to $(1, 1)_{\text{K}}$ in 2 ms by two Raman adiabatic passages. We then jump our magnetic field down close to the interspecies Feshbach resonance, before crossing it at a reduced speed of 3 G/s to ensure adiabatic molecule formation, followed by a jump to 345 G. After crossing the resonance we apply a magnetic-field gradient of 45 G/cm, which levitates the Feshbach molecules while overlevitating K and Cs atoms. K atoms are ejected from the trap, while Cs atoms remain trapped. To remove the Cs atoms, we ramp down the ODT power from 50 mW to 8 mW in 3 ms. After 10 ms of magnetic separation, the ODT power is ramped back up to 30 mW in 10 ms. This leaves a purified cloud of $7.6(10) \times 10^3$ Feshbach molecules at a temperature of $0.69(5) \mu\text{K}$ and a PSD of $2.8(8) \times 10^{-4}$. To image the molecules, we dissociate them by ramping the bias field up across the resonance in 2 ms, followed by absorption imaging at zero field of either the K or Cs atoms.

The molecules can be lost by relaxation to deeply bound states in collisions with either atoms or molecules. After molecule formation, and before the unbound atoms are ejected from the trap, molecules can collide with either K or Cs atoms.

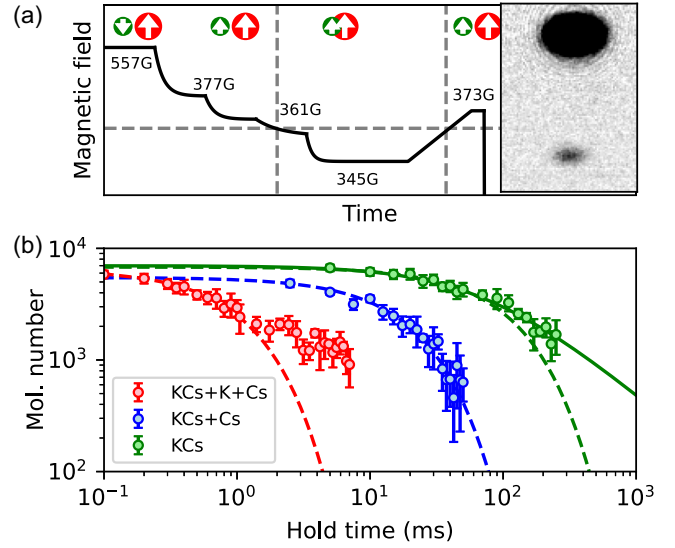


FIG. 2. Molecule creation and lifetimes. (a) Schematic of the magnetic-field changes during molecule creation. The horizontal dashed line marks the pole of the Feshbach resonance. The inset on the right shows a typical Stern-Gerlach absorption image of the molecular cloud (below) and remaining K atoms (above). (b) Molecular lifetimes in the presence of both K and Cs (red), only Cs (blue), and for a purified molecular cloud (green). Dashed lines are exponential fits to short hold times. The solid green line is a fit to a two-body decay curve.

We add a delay between molecule creation and magnetic-gradient switch-on and measure the decay of the molecular sample [Fig. 2(b)]. During the first ms, we measure a fast decay with a $1/e$ time of $0.96(4)$ ms. This fast decay leads to the loss of a significant fraction of the molecules on the timescale in which K can be ejected from the trap. To confirm this, we switch off the trap directly after magnetoassociation, which reduces atom density. We then observe typical molecule numbers of 11×10^3 , corresponding to molecule creation efficiencies of 1.4%(6%) on K(Cs). With K atoms removed but Cs still kept in the trap, the molecule lifetime is increased to $18(2)$ ms. This is long enough to allow our magnetic field to stabilize for STIRAP transfer to the ground state, and thus allows the creation of a mixture of Cs atoms and KCs ground-state molecules. Such a mixture is chemically stable under collisions [53], and may possess Feshbach resonances [54]. When both K and Cs are removed, the molecular decay is well described by a two-body decay curve $N(t) = N_0/(1 + \Gamma_2 \bar{n}_0 t)$ with a loss coefficient of $\Gamma_2 = 5.6(12) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$, where $\bar{n}_0$ is the mean initial density. The corresponding $1/e$ lifetime is about 130 ms.

IV. RESONANCE CHARACTERIZATION

The creation of our ultracold mixture and molecules makes it possible to collect new data on the scattering and bound-state properties of $^{39}\text{K}^{133}\text{Cs}$ and refit the interaction potentials. Previous Feshbach spectroscopy on a $^{39}\text{K} + ^{133}\text{Cs}$ mixture was carried out by Gröbner *et al.* [34]. They observed the positions of six resonances in three different collision channels. They used these to fit interaction potentials based

TABLE I. Overview of newly observed loss peaks observed in Feshbach spectroscopy and comparison with Feshbach resonance positions calculated with the interaction potentials of Ref. [34].

Threshold	$(f, m_f)_\text{K} + (f, m_f)_\text{Cs}$	B_loss^exp (G)	$B_\text{res}^\text{calc, 2017}$
b+b	$(1, 0)_\text{K} + (3, 2)_\text{Cs}$	315.551(2)	319.35
c+a	$(1, -1)_\text{K} + (3, 3)_\text{Cs}$	463.251(5)	none
c+a	$(1, -1)_\text{K} + (3, 3)_\text{Cs}$	466.230(5)	467.70
b+c	$(1, 0)_\text{K} + (3, 1)_\text{Cs}$	299.562(4)	304.62
d+a	$(2, -2)_\text{K} + (3, 3)_\text{Cs}$	478.028(4)	478.85
d+b	$(2, -2)_\text{K} + (3, 2)_\text{Cs}$	464.579(3)	466.01

on the singlet and triplet potential curves of Ferber *et al.* [55]. Gröbner *et al.* found that the resonances they observed were insufficient to determine both singlet and triplet potentials and could be fitted satisfactorily by modifying only the triplet potential.

To understand and overcome this limitation, we carry out coupled-channel calculations on the bound states responsible for potentially observable resonances in 14 different incoming channels with $M_F = m_{f,\text{K}} + m_{f,\text{Cs}} \geq 0$. These calculations use the bound package [56,57] with the interaction potentials of Gröbner *et al.* [34]. The results are shown in Supplemental Material [50]. In addition to the bound-state energies, we calculate the singlet fraction for each bound state at a magnetic field well removed from any avoided crossing that would perturb its character. The calculated singlet fractions range from 0.088–0.479, but those for the resonances observed by Gröbner *et al.* cover the much smaller range from 0.360–0.479. This explains why the singlet and triplet potentials could not be modified independently in Ref. [34].

In the present work, we use these calculations to identify resonances with a wide range of singlet fractions that have sufficient strength to be observed by atom loss spectroscopy on a nondegenerate mixture. After cooling the ultracold mixture, we drive Raman adiabatic passages to prepare K and Cs in the target spin state and hold the mixture for 50–100 ms, after which we measure the number of remaining Cs atoms. In the vicinity of a Feshbach resonance, the mixture will undergo enhanced loss due to three-body recombination. For each resonance, we fit a Lorentzian function to obtain the position of maximal loss.

When discussing resonances at multiple thresholds, explicit quantum numbers become cumbersome. In the remaining sections, we therefore specify atomic states with Roman sequence letters in order of increasing energy. A threshold is identified with a pair of letters, first for K and then for Cs.

The newly observed loss features are shown in Table I and compared with resonance positions calculated using the interaction potentials of Gröbner *et al.* [34]. There are discrepancies of up to 5 G in some cases, particularly for the resonances at the b + c and b + b thresholds, which are the ones for which the corresponding bound states have the lowest singlet fractions (0.135 and 0.191, respectively). This justifies refitting the model to optimize the singlet and triplet potentials simultaneously.

V. BOUND-STATE CHARACTERIZATION

To provide additional input for refitting the model, we characterize the bound state corresponding to the Feshbach resonance at 361.7 G by measuring its binding energy and magnetic moment. Our binding energy measurements start from the ultracold mixture at the end of evaporative cooling. We first transfer Cs to $(3, 2)_\text{Cs}$ and then measure the free-bound transition frequency by two-photon spectroscopy. When the two-photon detuning matches the sum of Zeeman splitting, binding energy, and relative kinetic energy of a colliding atom pair, the atomic pair is coupled into the weakly bound molecular state. Transfer to the molecular state shows up as atom loss, as molecules are quickly lost through atom-dimer relaxation or laser light absorption.

To extend the characterization to magnetic fields further below the Feshbach resonance, we measure the magnetic moment of our purified molecular sample μ_mol as a function of the magnetic field. After ramping the magnetic bias and gradient fields to their target values, the ODT power is reduced to 5 mW. When the magnetic-field gradient matches the levitation gradient $B'_\text{lev} = m_{\text{KCs}}g/\mu_\text{mol}$, molecules remain trapped. We plot the results of the binding-energy and magnetic-moment measurements in Fig. 3(a) and Fig. 3(b), respectively.

VI. FITTING POTENTIAL CURVES

The interaction potential curves $V_S(R)$ of Refs. [55] and [34] are made up of three segments. For each electronic state (singlet $X^1\Sigma^+$, $S = 0$, or triplet $a^3\Sigma^+$, $S = 1$), the central segment $V_S^\text{mid}(R)$ (around the potential minimum) is represented as a high-order power series in a transformed internuclear distance R . This is supplemented by a long-range extrapolation based on dispersion coefficients and a short-range extrapolation based on a simple inverse-power expansion. The central segment was fitted to extensive electronic spectra from Fourier-transform spectroscopy in Ref. [55]. The details of the potential functions are given in Refs. [55] and [34].

The physical quantities that principally determine the near-threshold bound states and the positions of Feshbach resonances are the singlet and triplet scattering lengths, a_s and a_t , and the leading dispersion coefficient C_6 . The singlet and triplet scattering lengths depend on the entire potentials $V_S(R)$. In the present work, we wish to adjust a_s and a_t to reproduce the observed ultracold properties, while retaining the fit to the Fourier-transform spectra as much as possible. We do this by retaining the power-series expansion coefficients for $V_S^\text{mid}(R)$ from Ref. [55], while adjusting the short-range extrapolations. The adjustments we make have only small effects on the energies of deeply bound vibrational states.

An important aspect of fitting interaction potentials is to ensure that parts of the potential that are not well determined by the experiments are constrained to have physically reasonable behavior. Previous work on potential curves for the alkali dimers has used short-range extrapolations of the form

$$V_S^\text{SR}(R) = A'_S + B'_S/R^{N_S} \quad \text{for } R < R_S^\text{SR}. \quad (2)$$

A'_S and B'_S are usually chosen to match the value and derivative of $V_S^\text{mid}(R)$ at R_S^SR . However, the values of the arbitrary powers N_S have ranged from 1.8 [58] to 12 [59], producing

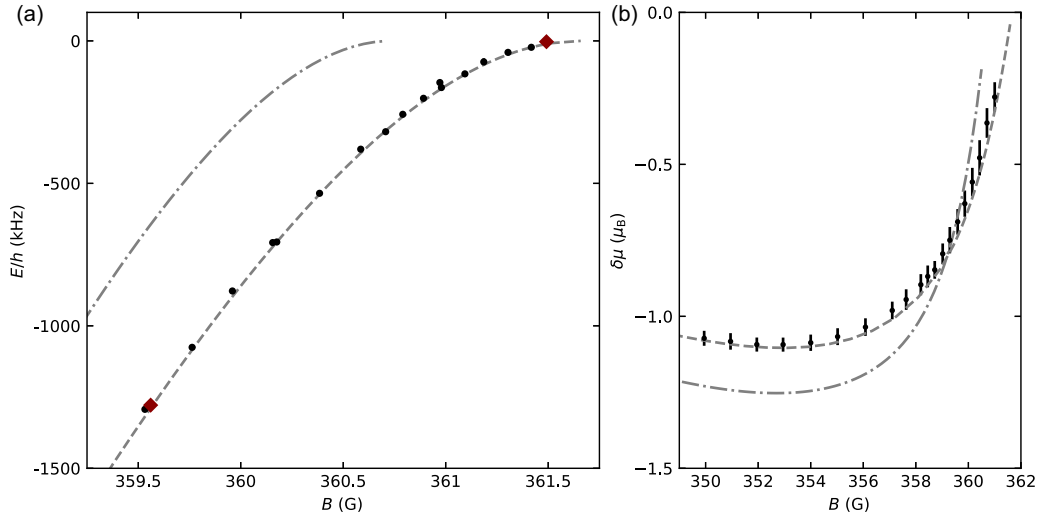


FIG. 3. Measurements to characterize the bound state that causes the Feshbach resonance at 361.7 G in the lowest channel. Coupled-channel calculations are plotted for the potentials from Gröbner *et al.* [34] (dash-dotted) and our refitted potentials (dashed). (a) Binding energies obtained through two-photon spectroscopy as a function of magnetic field B . Error bars are smaller than plot markers. The two data points used for refitting the potentials are marked with red diamonds. (b) Differential magnetic moment of the molecules with respect to an atom pair at the $(1, 1)_K + (3, 3)_{Cs}$ threshold as a function of magnetic field B .

unphysical variations in the hardness of the repulsive wall between systems. In the present work, we replace Eq. (2) with

$$V_S^{\text{SR}}(R) = A_S + B_S \exp(-\alpha_S R) \quad \text{for } R < R_S^{\text{SR}}. \quad (3)$$

Ihm *et al.* [60] have shown that, for a variety of systems, the repulsive part of the interaction between two atoms is well represented by an exponent $\alpha \approx 0.85\beta$, where β is the average of $\sqrt{8E_I(m_e/\hbar^2)}$ for the two atoms and E_I is the ionization energy. We have found that this approach gives a good representation of the short-range potentials for pairs of alkali-metal atoms [61]. For KCs, $0.85\beta = 1.767 \text{ \AA}^{-1}$. A_S and B_S are again chosen to match the value and derivative of the midrange potential at R_S^{SR} . In fitting with this functional form for KCs, we choose values of $R_0^{\text{SR}} = 3.5 \text{ \AA}$ and $R_1^{\text{SR}} = 5.25 \text{ \AA}$ that allow sufficient variations of scattering length for small variations of α_S around 0.85β . We then vary α_S , choosing A_S and B_S for each α_S to match the value and derivative of $V_S^{\text{mid}}(R)$ at R_S^{SR} .

Five of the six new resonances observed here are due to s -wave bound states crossing threshold. The exception is the feature near 463 G at the $c + a$ threshold, which cannot be explained as an s -wave feature and is considered further below. We carry out least-squares fits of α_0 and α_1 to these five resonances, together with the six resonances observed by Gröbner *et al.* [34]. We also include two data points to encapsulate the measurements of binding energies E_b described above: these are the fields at which E_b/h is 10.7 and 1281.5 kHz. Including both these ensures that both the position of the bound state and its curvature close to threshold are accurately reproduced; the curvature is a direct reflection of the width of the resonance, which is sensitive to $a_s - a_t$.

Our least-squares fits use the interactive fitting package I-NOLLS [62], which allows us to include multiple assignments of observables and adjust them manually as the fit proceeds. During the fitting process, all the properties are calculated

using the FIELD package [56,57]. The fitted potential parameters are given in Table II, including the derived quantities A_S and B_S of Eq. (3). The quality of fit to the observables is given in Table III. The resonance positions are fitted to within 1 G, except for the one at 599.32 G, whose interpretation is complicated by an avoided crossing very close to threshold. This level of agreement is reasonable in view of the complicated relationship between the positions of resonances and loss peaks [63].

It is conceptually useful to interpret the fitted potentials in terms of the resulting singlet and triplet scattering lengths, which are $a_s = -29.2(8) a_0$ and $a_t = 77.7(2) a_0$, respectively. The value of a_t for the current potential is outside the error bounds of the value $a_t = 74.88(9) a_0$ obtained in Ref. [34]. This apparent discrepancy arises because a_s and a_t are correlated. The effect of this correlation on uncertainties was implicitly neglected in Ref. [34], where a_s was fixed at the value from Ref. [55]. Our ability to determine a_s and a_t independently here comes both from the inclusion of resonances

TABLE II. The parameters of the fitted short-range potential of Eq. (3). The quantities in parentheses are standard errors including parameter correlation. All digits quoted are needed to reproduce the observables accurately in calculations. The derived parameters A_S and B_S are included for convenience. The complete potential curves are obtained by combining these with the mid-range and long-range potentials of Ref. [55].

Parameter	Singlet	Triplet	Unit
R_S^{SR}	3.5	5.25	$\text{\AA}$
α_S	1.70342(235)	1.726(15)	$\text{\AA}^{-1}$
A_S/hc	-5402.8994	-388.7871	cm^{-1}
B_S/hc	1.224302×10^6	2.266474×10^6	cm^{-1}
a_s	-29.2(8)	77.7(2)	a_0

TABLE III. Quality of fit to resonance positions and bound-state energies, together with the singlet fractions of the bound states with vibrational quantum number $n = -2$ that mostly determine the resonance positions. The quantities listed as “Unc.” determine the relative weights used in the fits; they are composite quantities based on physical arguments including model dependence, not experimental uncertainties.

Threshold	$B_{\text{loss}}^{\text{exp}}$ (G)	Unc. (G)	$B_{\text{res}}^{\text{calc}}$ (G)	Singlet fraction
a+a	442.59	0.4	443.45	0.482
b+a	513.12	0.3	513.77	0.478
b+a	419.30	0.3	420.24	0.360
b+b	315.57	0.2	315.77	0.190
c+a	466.24	0.2	465.61	$n = -1$
c+a	491.50	0.4	492.24	0.394
c+a	599.32	0.3	597.78	0.479
b+c	299.57	0.2	299.87	0.135
d+a	478.05	0.2	478.07	0.306
d+b	464.60	0.2	464.62	0.239
E_b/h (kHz)	B^{exp} (G)	Unc. (G)	B^{calc} (G)	
10.7	361.4995	0.03	361.4901	
1281.5	359.5660	0.02	359.5668	

with a wider range of singlet fractions than in Ref. [34] and from including precise measurements of binding energies. As mentioned above, the curvature of the binding energy near threshold is sensitive to $a_s - a_t$; this is significantly larger for the current potential than for the potential of Ref. [34], and it may be seen in Fig. 3 that both the curvature near threshold and the magnetic moment well below threshold are much better represented by the current potential.

We have used the new interaction potentials to calculate bound-state and scattering properties of $^{39}\text{K} + \text{Cs}$. The scattering calculations used the molscat package [57,64]. We obtained resonance positions, widths and background scattering lengths [65] for all the broad s -wave resonances that occur at the lowest threshold for each $M_F \geq 0$ at fields below 600 G. They are listed in the Supplemental Material [50]. In addition, Fig. 4 shows the near-threshold bound states below the lowest threshold between 310 and 370 G, including d -wave states (with $L = 2$). The s -wave state that produces the resonance near 361.7 G has character $[n(f, m_f)_{\text{K}}(f, m_f)_{\text{Cs}}, L] = [-2(2, 2)_{\text{K}}(3, 2)_{\text{Cs}}, s]$ at depths less than 30 MHz below threshold, but has a broad avoided crossing with $[-1(1, 1)_{\text{K}}(3, 3)_{\text{Cs}}, s]$ below that. It undergoes narrow avoided crossings at 323.17 and 329.04 G with d -wave states. As described in Supplemental Material [50], we observe losses of molecules in magnetic field ramps across 329.21(2) G, which we interpret as nonadiabatic processes due to the d -wave state. This confirms that the present potential works well for d -wave as well as s -wave states. There is an additional, shallower, d -wave state that crosses threshold near 358 G and has a strong avoided crossing with another d -wave state that appears at the lower right of the figure. The improved potentials can be used to compute accurate bound-state wave functions. With the addition of excited-state wave functions, we will be able to construct a model for STIRAP intensities [66] and identify optimal initial states for transfer to the hyperfine ground state.

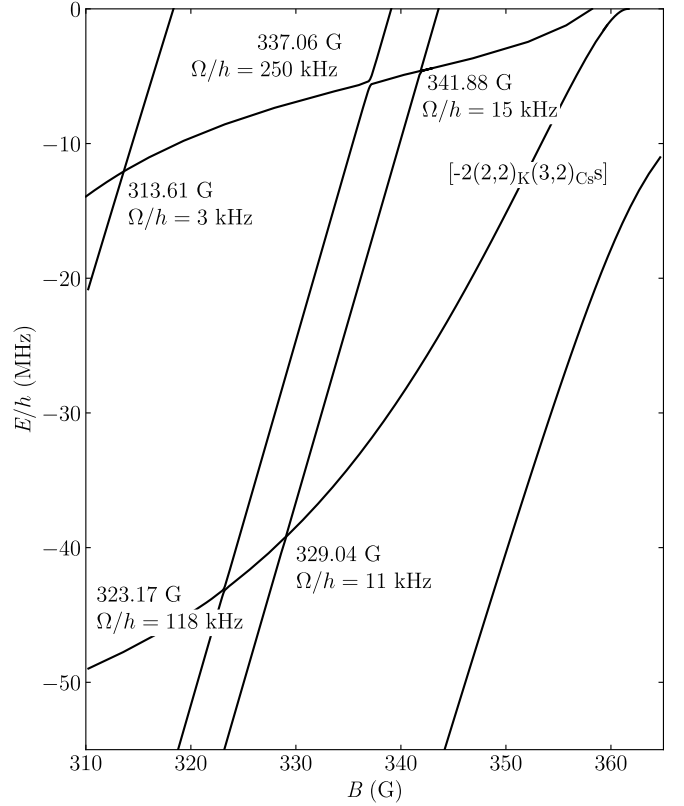


FIG. 4. Bound states of ^{39}KCs with $M_F = 4$ as a function of magnetic field B , shown relative to the lowest threshold. Avoided crossings are labeled with the field and the effective coupling matrix element Ω between the states. The state that is observed experimentally is labeled with its quantum numbers $[n(f, m_f)_{\text{K}}(f, m_f)_{\text{Cs}}, L]$.

We made an attempt to determine the dispersion coefficient C_6 from the present experimental data. This is potentially possible because most of the resonances observed here are due to bound states with vibrational quantum number $n = -2$ with respect to threshold, while the resonance near 466 G at the $c + a$ threshold is due to a state with $n = -1$. The spacing between states with different values of n depends sensitively on C_6 [67]. However, by contrast with RbCs [26] and NaCs [68], we were unable to determine a value of C_6 for KCs that differs significantly from the theoretical value [69]. Reducing the uncertainty in C_6 would require more extensive spectroscopic data or measurements of resonances involving states with a wider range of vibrational quantum numbers.

The interaction potentials do not provide a good explanation of the experimental loss feature observed at 463.27 G at the $c + a$ threshold. It cannot be due to an s -wave resonance, and even the nearest d -wave resonances lies at 459.1(3) G on the present fitted potential. The loss peak might arise from a mechanism other than a Feshbach resonance, such as a laser-driven process [70].

VII. CONCLUSIONS

We have demonstrated a method for producing ultracold mixtures of ^{39}K and ^{133}Cs . The method relies on sympathetic cooling of Cs by ^{39}K to reach sufficiently high phase-space

densities for molecule creation. With minor modifications, our technique should be able to produce similar mixtures of ^{41}K and ^{133}Cs . Further improvements to the cooling scheme may allow the creation of overlapping BECs. The high phase-space density of BECs can be exploited to form molecules at high filling fractions from dual Mott insulators in an optical lattice [71,72].

From our ultracold mixture, we can produce 7.6×10^3 ^{39}K ^{133}Cs Feshbach molecules by magnetoassociation using a Feshbach resonance at 361.7 G. Our purified molecular samples have lifetimes of about 130 ms, limited by inelastic two-body collisions between molecules. They provide an excellent starting point for the creation of ultracold ground-state ^{39}K ^{133}Cs molecules using STIRAP transfer [35].

We have used coupled-channel calculations to analyze the Feshbach resonances used to fit interaction potentials in previous work. We found that the bound states responsible for the resonances span a quite limited range of singlet fractions. Guided by coupled-channel calculations, we have measured the positions of resonances with a much wider range of singlet fractions. In addition, we have performed measurements of binding energies for the bound state corresponding to the 361.7 G resonance. We have used the measurements on bound states and resonance properties to determine new interaction potentials for $\text{K} + \text{Cs}$. The additional measurements in the present paper made it possible to fit the singlet and triplet interaction potentials independently and to obtain well-determined singlet and triplet scattering lengths. The new potentials give a very satisfactory picture of the ultra-

cold properties of $^{39}\text{K} + \text{Cs}$. They could in future be used to make predictions for other isotopic combinations, using coupled-channel calculations with appropriate atomic masses and hyperfine coupling constants.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [79].

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