Editors' Suggestion

Formation of Ultracold Molecules by Merging Optical Tweezers

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We demonstrate the formation of a single RbCs molecule during the merging of two optical tweezers, one containing a single Rb atom and the other a single Cs atom. Both atoms are initially predominantly in the motional ground states of their respective tweezers. We confirm molecule formation and establish the state of the molecule formed by measuring its binding energy. We find that the probability of molecule formation can be controlled by tuning the confinement of the traps during the merging process, in good agreement with coupled-channel calculations. We show that the conversion efficiency from atoms to molecules using this technique is comparable to magnetoassociation.

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Arrays of molecules confined in optical potentials are a powerful platform for quantum science [1]. Full quantum state control of molecules is essential to realise the potential of this system in the domains of quantum simulation [2–8] and quantum information processing [9–13]. Recent progress on trapping molecules in optical tweezers and optical lattices has demonstrated the first steps toward this goal, including single-site readout of individual molecules [14], entanglement of pairs of molecules [15–17], and the assembly of molecules predominantly occupying a single motional level of an optical tweezer [18].

Ultracold molecules may be prepared in optical potentials by following an indirect approach, where the molecules are produced by associating precooled pairs of atoms. This approach benefits from the wealth of techniques developed for the cooling and control of atoms and results in the formation of molecules which inherit the low temperatures of the constituent atoms. The formation of molecules often follows a two-step process where first weakly bound molecules are produced from an atomic sample using magnetoassociation [19,20] and then the weakly bound molecules are transferred to the rovibrational ground state using stimulated Raman adiabatic passage (STIRAP) [21,22]. Magnetoassociation exploits an avoided crossing between atomic and molecular states as a function of magnetic field and has been widely employed to convert atoms trapped in weakly confining optical potentials into molecules.

When the atoms are trapped in the tightly confining potentials of optical tweezers or lattices, only atom pairs in the relative motional ground state of the optical trap can be converted into weakly bound molecules using magnetoassociation [23]. In addition, confinement-related effects arise when the harmonic confinement length approaches the value of the s-wave scattering length. Effects like elastic [24,25] and inelastic [26,27] confinement-induced resonances (CIRs) have been observed experimentally in a number of different systems and dimensionalities [28–32]. These confinement-related effects offer new ways to form molecules. Using pairs of fermions in 1D, inelastic CIRs have been used to form molecules coherently in an optical trap [33]. In addition, molecules have been formed coherently utilizing spin-motion coupling in a strongly focused optical tweezer with large polarization gradients [34].

In contrast, there has been little experimental investigation of the interactions of two particles in separate optical potentials with tunable separation [35,36], despite the existence of theoretical work in this area [37,38]. Stock *et al.* [37] predicted the existence of avoided crossings between molecular and confined-atom states at critical values of the separation of two optical potentials. They termed these trap-induced shape resonances. Figure 1(a) shows the energies of a system with two atoms in separate but overlapping traps as a function of trap separation Δz. At large separation, the energies of the separately confined atom pairs are almost independent of Δz. However, there can also be a molecular state that is weakly bound at $\Delta z = 0$. The energy of this state increases quadratically with Δz due to the tweezer potentials and there is an

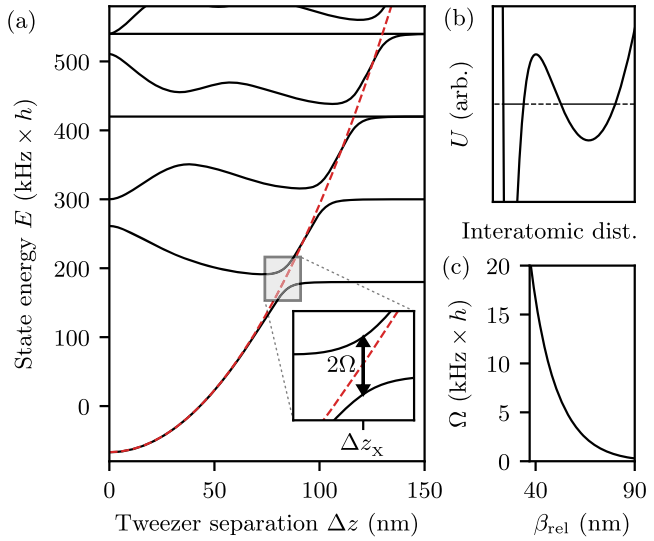

FIG. 1. (a) Energy levels for the Rb + Cs system as a function of separation between two spherically symmetric optical tweezers with confinement length $\beta_{rel} = 40$ nm for relative motion. The red dashed line shows the harmonic trapping experienced by the molecule in the absence of tunneling. An avoided crossing between the molecular and atom-pair states at Δz_X, expanded in the inset, allows mergoassociation. (b) Cartoon of the interaction energy as a function of interatomic distance when $\Delta z = \Delta z_X$. (c) The effective matrix element Ω between the lowest-energy atom-pair and molecular states as a function of β_{rel}.

avoided crossing with the lowest confined-atom state at a critical separation Δz_X. The strength of the avoided crossing depends on the height and width of the barrier between the atomic and molecular wells, as shown in Fig. 1(b); it is greatest when the bound state is close to threshold, corresponding to a large positive value of the s-wave scattering length a_s, and when the confinement length β_{rel} for relative motion of the atoms is comparable to a_s [39]. This avoided crossing offers an unexplored path to the formation of molecules by merging together two optical potentials. This was not observed in previous demonstrations of molecule formation in lattices [15,40,41] and tweezers [34,42], probably because $a_s \ll \beta_{rel}$ for the systems investigated.

In this Letter, we report the observation of molecule formation through merging of two optical tweezers, one containing a single Rb atom and the other a single Cs atom. Guided by coupled-channel calculations, we elucidate the experimental conditions that are necessary for molecule formation by merging of the optical potentials, a process referred to here as *mergo*association. We demonstrate mergoassociation by using optical spectroscopy to measure the binding energy of the molecular state occupied after molecule formation. The interaction potential between Rb and Cs atoms is accurately known [43] and comparison of our measurements with coupled-channel calculations of the near-threshold bound states allows us to identify the molecular state and understand the molecule formation process. We explore the tunability of the molecule formation probability by controlling the confinement strength during merging and compare the molecule formation probability to that obtained using magnetoassociation. Finally, we confirm that mergoassociation can be performed at low magnetic fields, without any magnetic field ramps, and detect the formation of molecules using microwave spectroscopy. This demonstrates the utility of mergoassociation in systems that do not possess Feshbach resonances suitable for magnetoassociation.

Our experiments begin by preparing single ^{87}Rb and ^{133}Cs atoms in hyperfine states $(f_{Rb} = 1, m_{Rb} = 1)$ and $(f_{Cs} = 3, m_{Cs} = 3)$ in the motional ground states of spatially separated, species-specific optical tweezers [44,45]. These tweezers are subsequently brought together in order to form a molecule. For the tightest traps we use ($\beta_{rel} \simeq 37$ nm), the trap frequencies of the separated tweezers in the direction of merging are $\simeq$140 kHz for Rb and $\simeq$100 kHz for Cs. The Rb + Cs atom-pair state $(1,1) + (3,3)$ has a near-threshold bound state with binding energy 110 ± 2 kHz × h at magnetic fields far from any Feshbach resonance; this corresponds to an interspecies background scattering length $a_s = 645(60)a_0$ [approx. 34.1(3) nm] [43]. The binding energy is comparable to the energy spacing of the harmonic levels in the tweezers and therefore within the regime where Stock *et al.* [37] predicted the existence of strong avoided crossings.

We have carried out coupled-channel calculations of the energy levels for pairs of atoms in separated tweezers, using the methods described in Supplemental Material [45]. Our calculations treat the individual tweezers as spherical and harmonic and neglect coupling between motions in the relative and center-of-mass coordinates. We represent the atom-atom interaction with a point-contact potential chosen to reproduce a_s. The resulting energies for $\beta_{rel} = 40$ nm are shown in Fig. 1(a). For each value of β_{rel}, we locate Δz_X and characterize the strength of the avoided crossing in terms of an effective matrix element Ω; this gives the strengths shown in Fig. 1(c). The value of Δz_X is approximately $\beta_{rel}\sqrt{3 + \beta_{rel}^2/a_s^2}$, so the crossing occurs at larger interatomic separations for weaker confinement. This leads to a reduction in tunneling through the barrier and in the strength of the avoided crossing.

If the avoided crossing is sufficiently strong, it may be traversed adiabatically with a slow enough change in Δz. This leads to the conversion of an atom pair in the ground state of relative motion into a molecular bound state. We calculate the probability of traversing the avoided crossing adiabatically using Landau-Zener theory [45].

Figure 2(a) shows the experimental sequence used to probe and dissociate molecules. A single Rb atom and a single Cs atom are prepared in species-specific tweezers which are merged together at magnetic field B_{merge}. The field is then ramped down to B_{spec}. Atom pairs can either be

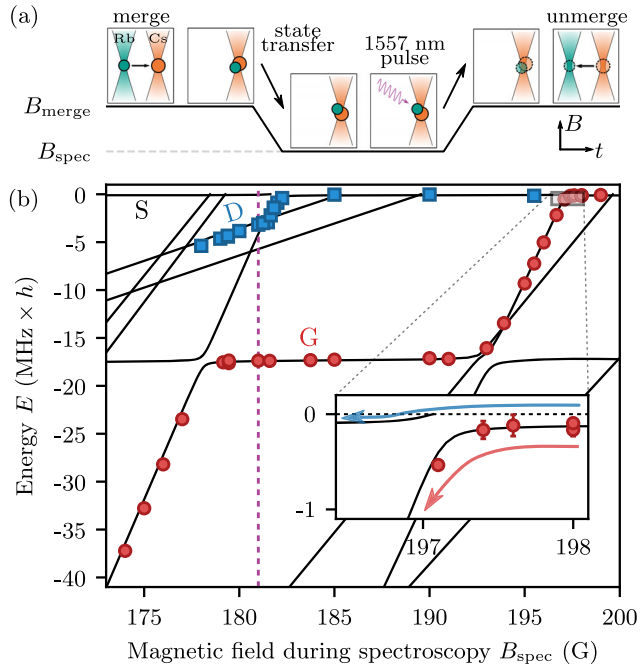

FIG. 2. (a) Sequence for molecule formation and detection. (b) The energy of weakly bound RbCs molecules (relative to threshold) as a function of magnetic field, B_{spec}. The points show the measured energies of molecules produced when merging the traps at 205 G (red circles) and below 197 G (blue squares). The purple dashed line indicates the field used for spectroscopy in Fig. 3 from states D and G. Black lines show state energies calculated from the RbCs molecular potential of Ref. [43]. The inset shows the avoided crossing below the resonance near 197 G and highlights the paths for mergoassociation (red arrow) and magnetoassociation (blue arrow) [61].

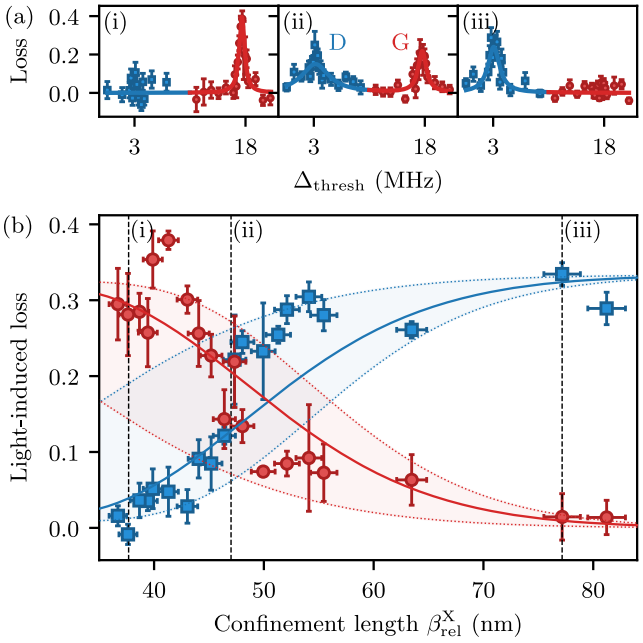

FIG. 3. (a) Spectroscopic identification of molecules formed by mergoassociation (red circles) and magnetoassociation (blue squares) for confinement lengths β^X_{rel} at the avoided crossing of (i) 37.7(8), (ii) 47(1), and (iii) 77(2) nm. Light-induced loss is measured as a function of the detuning Δ_{thresh} from the transition between the atomic threshold and $(^3\Pi_1, v' = 29, J' = 1)$ at 181 G. (b) The probability of light-induced loss of molecules formed by mergoassociation (red circles) and magnetoassociation (blue squares) as a function of β^X_{rel}. The theory curves show the calculated Landau-Zener probabilities scaled to match the light-induced loss. The shaded regions indicate the experimental uncertainty in the merging speed.

mergoassociated during the merging step if the tweezer confinement is sufficiently strong or be magnetoassociated if the magnetic field ramp crosses a Feshbach resonance. A spectroscopy pulse of light at 1557 nm is applied before the field ramps are reversed and the traps are unmerged. Then, fluorescence imaging of the atoms is performed to determine the trap occupancy. When the spectroscopy light is resonant with a molecular transition, loss to other molecular states results in no atoms being reimaged [57–60]. We identify the molecular states that have been populated during a sequence by comparing to coupled-channel calculations using the RbCs interaction potential of Takekoshi *et al.* [43,45].

Figure 2(b) shows the binding energies of RbCs molecules formed by mergoassociation, measured using the optical spectroscopy [45]. Molecules formed when merging the traps at $B_{merge} = 205$ G follow the path indicated by red circles when the field is ramped to B_{spec}. Entry into the near-threshold bound state by mergoassociation above the Feshbach resonance at 197.08(2) G allows us to approach this resonance from the molecular side, as shown in the inset, and subsequently to occupy states not accessible in magnetoassociation experiments from this starting field [59,61]. By mergoassociating with B_{merge} below this resonance we instead follow the path indicated by blue square points as the field is ramped. For the purposes of this investigation, we utilise the difference in energy between the states G and D at $B_{spec} = 181$ G (purple dashed line) to distinguish between molecules that have followed these two paths.

Figure 3 shows how the probability of mergoassociation depends on the tweezer confinement length β^X_{rel} at the avoided crossing [45]. For these measurements, the traps are merged at an average speed 1.4 μm/ms. However, the speed $(d\Delta z/dt)_X$ at the avoided crossing depends strongly on the alignment of the two tweezers; simulations of the combined potential predict that $(d\Delta z/dt)_X = 0.9^{+2.7}_{-0.4}$ μm/ms [45]. During the merging step, the applied magnetic field is $B_{merge} = 205$ G. Following merging, the movable tweezer is ramped off before the magnetic field is jumped down to 199 G and then ramped down to 196.8 G in 3 ms; this final step magnetoassociates any remaining atom pairs in the relative motional ground state with an

expected conversion efficiency greater than 99%. The intensity of the remaining tweezer is reduced substantially to prevent loss of the molecules due to photon scattering [45]. The magnetic field is then ramped to $B_{\mathrm{spec}} = 181$ G in 3 ms where it is held for 4 ms during which a 2 ms spectroscopy pulse is applied. The association routine is then reversed to dissociate any remaining molecules. The frequency of the spectroscopy laser at which we observe light-induced loss allows us to determine whether the molecule occupies state G or D and hence whether it was formed by mergoassociation or magnetoassociation. Figures 3(a)(i)–(iii) show optical spectra for strong, intermediate, and weak confinement during merging. For strong confinement we observe high occupation of state G as the majority of atom pairs in the motional ground state are mergoassociated. As the confinement is reduced, fewer atom pairs are mergoassociated, resulting in high occupation of state D.

Figure 3(b) shows the probability of excitation by the spectroscopy light as a function of $\beta_{\mathrm{rel}}^{\mathrm{X}}$ during merging of the traps. The spectroscopy laser saturates the transition from either G or D to determine the occupied molecular state. We clearly observe a change in the probability of mergoassociation from high to low as the confinement is reduced. The peak probability of the mergoassociation data points (red circles) and the magnetoassociation data points (blue squares) indicates that the efficiency of the two techniques is similar. The red (blue) curve shows the calculated Landau-Zener probability P_{LZ} [45] of mergoassociation (no mergoassociation) for our experimental parameters, scaled to that of the light-induced loss. The shaded regions show the uncertainty in P_{LZ} arising from the uncertainty in $(d\Delta z/dt)_{\mathrm{X}}$. The experimental results are in good agreement with our theoretical model, with the observed crossover point in β_{rel} within 10% of the theoretical prediction. The agreement is surprisingly good in view of the approximations made in the model, particularly the assumption that the tweezers are spherically symmetric. In reality, our tweezers have an aspect ratio of 2:5 between the confinement lengths along the axes of approach and tweezer-light propagation. Both magnetoassociation and mergoassociation rely on preparation of the initial atom pair in the correct internal state and the ground state of relative motion. This state preparation is the current limit to our association efficiency [45].

Finally, we verify that mergoassociation can be performed at low magnetic fields, without any magnetic field ramps. Mergoassociation is still possible in this regime, because it relies only on the presence of a bound state near threshold, which for RbCs exists over a large range of magnetic field. The experimental sequence is similar to the one described earlier, but the magnetic field is held constant at the field applied during cooling of the atoms, $B = 4.78$ G, for the entirety of the molecule formation and detection portion of the sequence. A microwave pulse of frequency ~6.84 GHz is applied for 89 μs in place of the pulse of light at 1557 nm. This pulse length approximates a π pulse for the Rb atom and for the RbCs molecule in the least-bound state S [45]. Following the unmerging of the traps, state-sensitive detection is performed by using resonant "pushout" pulses to eject any Rb atoms in the state $f = 2$ and any Cs atoms in the state $f = 4$ prior to imaging [62].

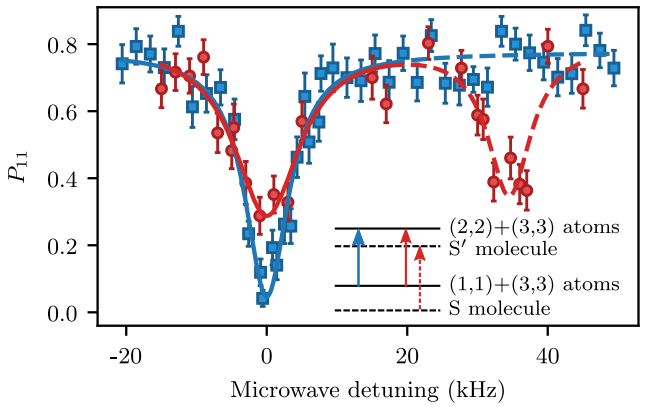

FIG. 4. Microwave spectroscopy of RbCs molecules produced by mergoassociation at $B = 4.78$ G. The probability P_{11} of detecting both a Rb and Cs atom at the end of the sequence is shown as a function of the microwave detuning from the transition $(1,1) \rightarrow (2,2)$ in atomic Rb for strong (red circles) and weak (blue squares) confinement during merging: $\beta_{\mathrm{rel}}^{\mathrm{X}} = 39.4(9)$ and 55(1) nm, respectively. When a molecule is formed, we observe the molecular transition S → S′ at detuning 35(2) kHz. The inset shows the energy-level structure of the relevant states with atomic (molecular) states indicated with solid (dashed) lines.

The results of the microwave spectroscopy are shown in Fig. 4. The blue squares show the results for weak confinement during merging, where the probability of mergoassociation is low and we expect to prepare an atom pair. We observe only a single feature in the probability P_{11} of observing a Rb and Cs atom at the end of the sequence; this is the feature corresponding to the hyperfine transition $(1,1) \rightarrow (2,2)$ in atomic Rb. In contrast, when the tweezers are merged with stronger confinement, as shown by the red circles, we mergoassociate the atom pair to form a molecule. Consequently, we observe an additional feature in P_{11}, detuned by 35(2) kHz; this corresponds to the molecular transition S → S′ illustrated in the inset. Both features are fitted with Lorentzians as shown by solid (dashed) lines for the atomic (molecular) transition. Using the RbCs interaction potential fitted in Ref. [43], we calculate the binding energy of state S′ at 4.78 G to be 80 kHz × h. This value is smaller than that of S (122 kHz × h) and the calculated difference in binding energy (42 kHz × h) is in reasonably good agreement with the experimental measurement. The atom pair is prepared in the required hyperfine states in 78(1)% of experimental runs leading to the offset of P_{11} from unity in Fig. 4. The relative depths of the features in Fig. 4 indicate that 46(8)% of these atom pairs are converted into molecules.

In summary, we have created a trap-induced avoided crossing between atomic and molecular states and used it to create molecules during the merging of pairs of optical tweezers. The efficiency of molecule formation depends on the strength of the avoided crossing, which critically depends on the confinement length for relative motion. The avoided crossing is strongest when there is a bound state near threshold and the confinement length is comparable to the scattering length. This situation is realized for Rb + Cs at a large range of magnetic fields. We have demonstrated that the efficiency of molecule formation by mergoassociation is comparable to that of magnetoassociation in this system.

This work demonstrates a new technique for the formation of molecules in systems with large interspecies interactions. It will be effective even in systems that do not possess Feshbach resonances suitable for magnetoassociation [63]. It would be interesting to test this technique using transport in an optical lattice, where the tighter confinement achievable should allow efficient conversion of atom pairs into molecules for systems with moderate positive scattering lengths. This would also open up applications in neutral-atom quantum computing by using the trap-induced avoided crossing for high-fidelity two-qubit quantum logic operations [64]. Further, our observations have important ramifications for collision measurements using tweezer-confined particles and for the study of hybrid atom-ion systems [65,66].

This work also demonstrates the first production of RbCs molecules in optical tweezers. These weakly bound molecules have sufficient lifetimes to be transferred to the rovibrational ground state using STIRAP, as has been previously demonstrated for weakly confined samples of RbCs [58–60]. The production of ground-state molecules using these techniques prepares the molecules predominantly in the lowest motional state of the trap, which will allow the implementation of high-fidelity entangling gates between molecules [10,11].

The data presented in this Letter are available from [67].

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