Multichannel interactions of two atoms in an optical tweezer

J.D. Hood,1, 2, 3 ∗Y. Yu,2, 1, 3 Y.-W. Lin,1, 2, 3 J.T. Zhang,2, 1, 3
K. Wang,2, 1, 3 L.R. Liu,2, 1, 3 B. Gao,4 and K.-K. Ni1, 2, 3,

1Department of Chemistry and Chemical Biology, Harvard University, Cambridge, Massachusetts, 02138, USA
2Department of Physics, Harvard University, Cambridge, Massachusetts, 02138, USA
3Harvard-MIT Center for Ultracold Atoms, Cambridge, Massachusetts, 02138, USA

(Dated: July 26, 2019)

The multichannel Na-Cs interactions are characterized by a series of measurements using two atoms in an optical tweezer, along with a multichannel quantum defect theory (MQDT). The triplet and singlet scattering lengths are measured by performing Raman spectroscopy of the Na-Cs motional states and least-bound molecular state in the tweezer. Magnetic Feshbach resonances are observed for only two atoms at fields which agree well with the MQDT. Our methodology, which promotes the idea of an effective theory of interaction, can be a key step towards the understanding and the description of more complex interactions. The tweezer-based measurements in particular will be an important tool for atom-molecule and molecule-molecule interactions, where high densities are experimentally challenging and where the interactions can be dominated by intra-species processes.

Tuning interactions in ultracold gases of atoms and molecules via Feshbach resonances or optical lattices has enabled studies of many rich quantum phenomena such as the BEC-BCS crossover [1], superfluid-to-Mott insulator transitions [2], and supersolidity [3–5]. Feshbach resonances have also been utilized to associate loosely-bound molecules, which has been an important step for creating ultracold ro-vibrational ground state molecules [6–10]. A key prerequisite for these experiments is an understanding of the underlying two-body and few-body interactions. Although the origins of these interactions are complex molecular potentials, in the low-temperature regime effective theories can describe the interactions with no reliance on the short-range potentials. For example, the multichannel quantum defect theory (MQDT) can provide an efficient description of atom-atom interactions in all spin channels and partial waves using as few as three parameters: the triplet and singlet scattering lengths, and the van der Waals C₆ coefficient [11].

Probing two- to few-body interactions in cold atoms has traditionally been performed by scattering experiments in bulk gases [12] or by spectroscopy in optical lattices [13, 14]. More recently, single atoms trapped in optical tweezers have become widely pursued as a versatile experimental platform for studying few and many-body physics through bottom-up scaling [15–23]. An optical tweezer with only two atoms is a pristine environment for studying ultracold collisions [24–26] or for producing two-particle entanglement [19, 27]. Optical tweezers are now even being used with molecules, where they can either assemble molecules from single ultracold atoms [25, 28] or be used to directly trap single ultracold molecules [29].

The natural questions arise: would measurements performed on two particles in an optical tweezer be sufficient to fully characterize two-body interaction, including the identification of Feshbach resonances? And, in the fu-
ture, can such a platform offer a way to investigate interactions of more complex composite particles? Here we demonstrate a combined experimental and theoretical effort to fully characterize multichannel atom-atom interactions and to search for Feshbach resonances using two atoms in the same optical tweezer: one $^{23}$Na and one $^{133}$Cs. Despite previous characterization of the Na-Cs potential using Fourier-transform spectroscopy with hot atoms [30], the near-threshold NaCs ground-state potential and Feshbach resonances have not been directly probed.

Taking advantage of the exquisitely prepared initial conditions in the optical tweezer, we probe the collisions of exactly two particles in fully-controlled quantum states without any contribution from multi-body effects or intra-species processes. Our work combines Raman spectroscopy of trap motional states and molecular levels with MQDT to extract the Na-Cs singlet and triplet scattering lengths. Furthermore, we observe the first Na-Cs Feshbach resonances, and the corresponding magnetic fields agree to within 1% of our effective theory.

Our tweezer-based scheme can be extended to more complex interactions, such as atom-molecule or molecule-molecule interactions [31–33]. The sub-micron optical tweezer creates a high effective density for collisions of around $\rho \approx 10^{14}$ cm$^{-3}$, which is useful for species where achieving high densities is otherwise experimentally challenging.

We perform our experiment with a single Na and a single Cs atom in the motional ground state of the same optical tweezer, as schematically shown in Fig. 1(a). Details of the setup are reported in Ref. [28]. Initially, a single Na and Cs atom are loaded stochastically into separate tweezers from a magneto-optical trap with a combined probability of $\approx 35\%$ [25]. The final results, however, can be post-selected to guarantee both atoms are initially present with high confidence. The single atoms are then cooled simultaneously to their 3D motional ground state [28]. The resulting mean separation distance is 112 nm, which gives an effective density of $\rho = 2 \times 10^{14}$ cm$^{-3}$.

The strategy for extracting the triplet and singlet scattering lengths is outlined here and described in more detail in the following paragraphs. The triplet scattering length is directly related to the binding energy of the triplet least-bound molecular state by a single channel quantum defect theory (QDT) [37–39], which we measure with two-photon spectroscopy. We also measure the shifts of the Na-Cs motional states in the tweezer due to the interactions in various spin combinations, which can be related to scattering lengths through a numerical calculation of two atoms in a harmonic potential. Finally, we use MQDT [11, 40–43] (which accounts for hyperfine interaction) to extract the singlet scattering length consistent with the measured scattering lengths for the different hyperfine spins.

When both atoms are in the same trap, they interact through their molecular potential $V(r_1 - r_2)$, which consists of short-range molecular forces and a long-range van der Waals potential with a lowest-order term $-C_6/r^6$, where $r$ is the relative coordinate. In the low-energy limit where the de Broglie wavelengths are much larger than the molecular potential, their interaction can be well-modeled by a Fermi pseudo-potential consisting of the scattering length $a$ and a regularized $\delta$-function, $V(r_1 - r_2) = \frac{2\hbar^2}{\mu a} \delta(3)(r_1 - r_2) \frac{\delta}{\delta \beta} r$ [44], where $\mu$ is the reduced mass. The validity of the pseudo-potential is characterized by the ratio of the van der Waals length $\beta_0 = (2\hbar C_6/\mu^2)^{1/4}$ to the relative harmonic oscillator lengths $\beta_R = \sqrt{\hbar/\mu \omega_R}$ [45, 46], where $\omega_R$ is the relative trapping frequency. In our experiment, these are $\beta_0 \approx 6$ nm, $\beta_R, \text{Radial} \approx 66$ nm for the radial axes, and $\beta_R, \text{Axial} \approx 158$ nm for the axial axis.

The two-body interaction shifts the motional trapping energies of the atoms in the tweezer, which we calculate as a function of the scattering length in Fig. 1(b) and probe experimentally in Fig. 2. In the simplified case where the atoms have the same trapping frequencies, as in the case of identical atoms, the Hamiltonian is separable into center-of-mass (CM) and relative coordinates. For this separable case, analytical results exist for a regularized $\delta$-function in a spherically [44] and cylindrically [47] symmetric harmonic trap. However, due to the different masses and ground state polarizabilities at the tweezer wavelength of 976 nm, Cs trapping frequencies are 19% larger than Na trapping frequencies, which are $(\omega_{Na,r}, \omega_{Na,y}, \omega_{Na,z}) = 2\pi \times (109, 118, 20)$ kHz. We therefore calculate the shifted frequencies using the analytic solutions for the separable, cylindrically symmetric Hamiltonian, and then diagonalize a matrix containing the remaining non-separable [48] and anisotropic terms of the full Hamiltonian. The details are described in the SM [49].

In Fig. 1(b), the calculated Na-Cs motional trapping energies are plotted as a function of the scattering length $a$. The motional ground state $\Delta E_g$ (blue) agrees with first-order perturbation theory (dashed) for small scattering lengths, which is given by

$$\Delta E_g \approx a \left( \frac{2\hbar^2}{\mu \sqrt{\pi} \beta_{\text{eff}}} \right),$$

where $\beta_{\text{eff}}$ is the 3-axis effective oscillator length defined in the SM [49]. For positive (negative) scattering lengths, the motional ground state shifts to higher (lower) frequencies, corresponding to a repulsive (attractive) potential. For the excited motional states (black), states with an odd relative axial quantum number $m_R$ have no shift because the relative wavefunction is zero at the $\delta$-
function, and therefore are non-interacting, while states with even \( m_R \) are interacting.

Experimentally, we measure the interaction shifts by resonantly flipping the ground hyperfine spin of one but not the other atom and then comparing it to the bare hyperfine splitting measured in the absence of the other atom, as shown in Fig. 2(a). Atoms are initially prepared in stretched state spin combinations that are stable against hyperfine changing spin collisions and denoted by \( |↑_{Cs}⟩ \) \( |F=4,m_f=4⟩_{Cs} \), \( |↓_{Cs}⟩ \) \( |F=3,m_f=3⟩_{Cs} \), \( |↑_{Na}⟩ \) \( |F=2,m_f=2⟩_{Na} \), and \( |↓_{Na}⟩ \) \( |F=1,m_f=1⟩_{Na} \). The spin flip of one atom is driven by an optical Raman pulse with co-propagating beams at a bias magnetic field of 8.8 gauss.

For the case of flipping a Cs spin in Fig. 2(b), several interaction-shifted peaks are observed by comparing \( |↑_{Na}↑_{Cs}⟩ \rightarrow |↑_{Na↓_{Cs}}⟩ \) (blue solid) to the one-atom case \( |↑_{Cs}⟩ \rightarrow |↓_{Cs}⟩ \) (orange). We identify the largest peak as the shifted ground motional state \( |m_R,m_{CM}=0⟩ \). The shift gives the difference of the interaction shifts of the initial and final spin configurations, \( Δν_1 = (ΔE_g(|↑_{Na}↑_{Cs}⟩) − ΔE_g(|↑_{Na↓_{Cs}}⟩))/ℏ = −32.1(2) kHz \). The smaller peaks at positive frequency shifts correspond to the motional excited states \( |2,0⟩ \) and \( |4,0⟩ \), which can be populated because they have some overlap with the initial state due to the wavefunction modification by the strong interactions. The peak near zero frequency corresponds to the initial Na and Cs population that is not prepared in the motional ground state or an interacting state. The fitted height 0.46 of the \( |0,0⟩ \) peak serves as a lower bound for the relative motional ground state population.

Similarly, for the case of flipping a Na spin \( |↑_{Na}↑_{Cs}⟩ \rightarrow |↓_{Na↓_{Cs}}⟩ \) in Fig. 2(c), we observe several interaction shifted peaks corresponding to the motional states of \( |0,0⟩ \), \( |2,0⟩ \), and \( |0,2⟩ \), and a non-shifted peak as the initial non-interacting population.

Because interaction shifts only give the difference of the shifts between two states, we determine an absolute interaction shift of the triplet \( |↑_{Na}↑_{Cs}⟩ \) by measuring the triplet least-bound \( (v = −1) \) binding energy, which can be related to the triplet scattering length directly through a single channel QDT [37–39]. We measure the least-bound \( (v = −1) \) triplet binding energy with two-photon Raman spectroscopy, as schematically shown in Fig. 3(a). When the two-photon detuning is resonant with the binding energy, the atoms are transferred to the molecular state which is observed as simultaneous loss of both the Na and Cs atom [28], as shown in the spectrum in Fig. 3(b). The resonance positions are plotted in Fig. 3(c) as a function of different tweezer powers in order to extrapolate the binding energies without light shift. For these experiments, the optical tweezer light is also used as the Raman beams. A linear extrapolation to zero power gives a \( N = 0 \) binding energy of 297.6(1) MHz.

The single-channel QDT relates the binding energies to the scattering lengths. Due to the large hyperfine constant of Cs and for greater accuracy, we extend the QDT to a two-scale theory that includes the \( −C_S/r^6 \) potential in addition to \( −C_S/r^6 \) (see SM for details [49]) using the theoretical \( C_S \) and \( C_S \) coefficients from Refs. [30, 50, 51]. The \( N = 0 \) binding energy then gives a triplet scattering length \( a_T = α(↑_{Na}↑_{Cs}) = 30.4(6) a_0 \), where \( a_0 \) is the Bohr radius. The scattering lengths are summarized in Table 1.

From the triplet scattering length from the binding energy measurement, the pseudo-potential model then gives an absolute interaction shift of \( ΔE_g(|↑_{Na}↑_{Cs})/ℏ = 1.40 \) kHz, which can be used to obtain an absolute shift for each state in the interaction shift measurements. The measurement \( Δν_1 \) in Fig. 2(b) gives an absolute interac-
We note that, despite the higher temperature of the atoms as compared to those in typical bulk-gas-based [0x0] experiments, we compare the MQDT calculation to the Na Cs interactions completely characterized at temperatures of 10 μK. The B-field is ramped on, the atoms are merged into the same tweezer and held for 100 ms, and then the atoms are separated and imaged individually to check for loss. Because there are no lower energy states with the same total $J_F$, the inelastic loss must occur through anisotropic interactions such as the electron spin-spin interactions.

Our search uses a separate optical tweezer apparatus, shown in Fig. 4(a), with Helmholtz coils that are capable of producing a magnetic field, $B$, up to 1000 G at the atoms’ location. The atoms are prepared in the lower hyperfine manifolds, $|Na(F = 1, m_F = -1)\rangle$ and $|Cs(F = 3, m_F = -3)\rangle$, without Raman sideband cooling at about 100 μK. The B-field is ramped on, the atoms are merged into the same tweezer and held for 100 ms, and then the atoms are separated and imaged individually to check for loss. Because there are no lower energy states with the same total $J_F$, the inelastic loss must occur through anisotropic interactions such as the electron spin-spin interactions.

Using our measured scattering parameters, the two-scale MQDT predicts two Feshbach resonances for this hyperfine state, where the closed channel is either the rotational ground $N = 0$ (s-wave) or excited $N = 1$ (p-wave) of a molecular state which has the approximate quantum number $\nu = -1$ and $Na (F = 2)Cs(F = 3)$ in the low-field limit. Na-Cs loss spectroscopy is shown in Fig. 4(c) and (d) for two different magnetic field ranges. The s-wave resonance is observed at 652.1(4) G, and the narrower p-wave Feshbach resonance is observed at 791.10(5) G, which agree with the MQDT predictions with an accuracy of 1.4% and 0.9% respectively. These are summarized in Table I.

We note that, despite the higher temperature of the atoms as compared to those in typical bulk-gas-based
searches, the tight confinement and exact preparation provided by the optical tweezer increases the sensitivity to losses due to Feshbach resonances.

The agreement of the Feshbach resonance locations with the MQDT model combined with a series of tweezer-based measurements represents an important validation of the use of effective theory for interactions. The interaction of composite particles, and in particular molecules, is generally too difficult for the standard coupled-channel approach because the number of coupled-channels is simply too large [31–33]. This difficulty requires that we look for effective theories to describe the interactions that require a minimum number of parameters, and efficient ways to measure these parameters. This tweezer scheme can also be used to probe atom-molecule and molecule-molecule interactions, providing both high effective densities and exact preparation of the collisional partners. The measurement of the interaction shift of an atom in the presence of a molecule could also be used for non-destructive state-sensitive detection of the molecule, which is a challenge for molecules without closed optical transitions. For Na-Cs in particular, the observation of a Na-Cs Feshbach resonance for exactly two atoms is an important step towards creating a single Feshbach molecule, and the eventual coherent creation of a ro-vibrational ground state molecule.

We thank Lewis Picard, Eliot Fenton, and Frederic Condlin for experimental assistance. This work is supported by the Arnold and Mabel Beckman Foundation, as well as the NSF (PHYS-1806595 and through Harvard-MIT CUA), AFOSR (FA9550-19-1-0089), and the Camille and Henry Dreyfus Foundation. J. T. Z acknowledges support from an NDSEG fellowship. The work at Toledo was supported by NSF (PHY-1607256).

* jhood@fas.harvard.edu
  ni@chemistry.harvard.edu

[1] C. A. Regal, M. Greiner, and D. S. Jin, Phys. Rev. Lett. 92, 083201 (2004).
[2] M. Greiner, O. Mandel, T. Esslinger, T. W. Hänsch, and I. Bloch, Nature 415, 39 (2002).
[3] F. Böttcher, J.-N. Schmidt, M. Wenzel, J. Hertkorn, M. Guo, T. Langen, and T. Pfau, Phys. Rev. X 9, 011051 (2019).
[4] L. Tanzi, E. Lucioni, F. Famà, J. Catani, A. Fioretti, C. Gabbanini, R. N. Bisset, L. Santos, and G. Modugno, Phys. Rev. Lett. 122, 130405 (2019).
[5] L. Chomaz, D. Petter, P. Ilzhöfer, G. Natale, A. Trautmann, C. Politi, G. Durastante, R. M. van Bijnen, A. Patscheider, M. Suhnen, M. J. Mark, and F. Ferlaino, Phys. Rev. X 9, 021012 (2019).
[6] K.-K. Ni, S. Ospelkaus, M. H. G. de Miranda, A. Pe’er, B. Neyenhuis, J. J. Zirbel, S. Kotochigova, P. S. Julienne, D. S. Jin, and J. Ye, Science 322, 231 (2008).
[7] P. K. Molony, P. D. Gregory, Z. Ji, B. Lu, M. P. Köppinger, C. R. Le Sueur, C. L. Blackley, J. M. Hutson, and S. L. Cornish, Phys. Rev. Lett. 113, 255301 (2014).
[8] T. Takekoshi, L. Reichsöllner, A. Schindewolf, J. M. Hutson, C. R. Le Sueur, O. Dulieu, F. Ferlaino, R. Grimm, and H.-C. Nägerl, Phys. Rev. Lett. 113, 205301 (2014).
[9] J. W. Park, S. A. Will, and M. W. Zwierlein, Phys. Rev. Lett. 114, 205302 (2015).
[10] M. Guo, B. Zhu, B. Lu, X. Ye, F. Wang, R. Vexiau, N. Bouloufa-Maafa, G. Quéméré, O. Dulieu, and D. Wang, Phys. Rev. Lett. 116, 205303 (2016).
[11] B. Gao, E. Tiesinga, C. J. Williams, and P. S. Julienne, Phys. Rev. A 72, 042719 (2005).
[12] J. Weiner, V. S. Bagnato, S. Zilio, and P. S. Julienne, Rev. Mod. Phys. 71, 1 (1999).
[13] J. Amato-Grill, N. Jepsen, I. Dimitrova, W. Lunden, and W. Ketterle, Phys. Rev. A 99, 033612 (2019).
[14] A. Goban, R. Hutson, G. Marti, S. Campbell, M. Perlin, P. Julienne, J. Dincao, A. Rey, and J. Ye, Nature 563, 369 (2018).
[15] N. Schlosser, G. Reymond, I. Protosenko, and P. Grangier, Nature 411, 1024 (2001).
[16] B. Darquié, M. P. A. Jones, J. Dingjan, J. Beugnon, S. Bergamini, Y. Sortais, G. Messin, A. Browaeys, and P. Grangier, Science 309, 454 (2005).
[17] Y. Miroshnychenko, W. Alt, I. Dotsenko, L. Förster, M. Khudaverdyan, D. Meschede, D. Schrader, and A. Rauschenbeutel, Nature 442, 151 (2006).
[18] D. D. Yavuz, P. B. Kulatunga, E. Urban, T. A. Johnson,
N. Proite, T. Henage, T. G. Walker, and M. Saffman, Phys. Rev. Lett. 96, 063001 (2006).

[19] A. M. Kaufman, B. J. Lester, M. Foss-Feig, M. L. Wall, A. M. Rey, and C. A. Regal, Nature 527, 208 (2015).

[20] J. D. Thompson, T. G. Tiecke, A. S. Zibrov, V. Vuletić, and M. D. Lukin, Phys. Rev. Lett. 110, 133001 (2013).

[21] M. A. Norcia, A. W. Young, and A. M. Kaufman, Phys. Rev. X 8, 041054 (2018).

[22] S. Sasaki, J. T. Wilson, B. Grinkemeyer, and J. D. Thompson, Phys. Rev. Lett. 122, 143002 (2019).

[23] J. P. Covey, I. S. Madjarov, A. Cooper, and M. Endres, Phys. Rev. Lett. 122, 173201 (2019).

[24] P. Xu, J. Yang, M. Liu, X. He, Y. Zeng, K. Wang, J. Wang, D. J. Papoular, G. V. Shlyapnikov, and M. Zhan, Nat. Commun. 6, 7803 (2015).

[25] L. R. Liu, J. D. Hood, Y. Yu, J. T. Zhang, N. R. Hutzler, T. Rosenband, and K.-K. Ni, Science 360, 900 (2018).

[26] Q. Guan, V. Klinkhamer, R. Klemt, J. H. Becher, A. Bergschneider, P. M. Preiss, S. Jochim, and D. Blume, Phys. Rev. Lett. 122, 083401 (2019).

[27] P. Sompet, S. S. Szigeti, E. Schwartz, A. S. Bradley, and M. F. Andersen, Nat. Commun. 10, 1889 (2019).

[28] L. R. Liu, J. D. Hood, Y. Yu, J. T. Zhang, N. R. Hutzler, T. Rosenband, and K.-K. Ni, Phys. Rev. X 9, 021039 (2019).

[29] L. Anderegg, L. W. Cheuk, Y. Bao, S. Burchesky, W. Ketterle, K.-K. Ni, and J. M. Doyle, (2019), arXiv:1902.00497.

[30] O. Docenko, M. Tamanis, J. Zaharova, R. Ferber, A. Pashov, H. Knöckel, and E. Tiemann, J. Phys. B 39, S929 (2006).

[31] M. Mayle, B. P. Ruzic, and J. L. Bohn, Phys. Rev. A 85, 062712 (2012).

[32] M. Mayle, G. Quéméner, B. P. Ruzic, and J. L. Bohn, Phys. Rev. A 87, 012709 (2013).

[33] P. D. Gregory, M. D. Frye, J. A. Blackmore, E. M. Bridge, R. Sawant, J. M. Hutson, and S. L. Cornish, Nat. Commun. 10, 3104 (2019).

[34] C. Monroe, D. M. Meekhof, B. E. King, S. R. Jefferts, W. M. Itano, D. J. Wineland, and P. Gould, Phys. Rev. Lett. 75, 4011 (1995).

[35] A. M. Kaufman, B. J. Lester, and C. A. Regal, Phys. Rev. X 2, 041014 (2012).

[36] Y. Yu, N. R. Hutzler, J. T. Zhang, L. R. Liu, J. D. Hood, T. Rosenband, and K.-K. Ni, Phys. Rev. A 97, 063423 (2018).

[37] B. Gao, Phys. Rev. A 58, 4222 (1998).

[38] B. Gao, Phys. Rev. A 64, 010701 (2001).

[39] B. Gao, Phys. Rev. A 78, 012702 (2008).

[40] T. M. Hanna, E. Tiesinga, and P. S. Julienne, Phys. Rev. A 79, 040701 (2009).

[41] B. Gao, Phys. Rev. A 84, 022706 (2011).

[42] C. Makrides and B. Gao, Phys. Rev. A 89, 062718 (2014).

[43] Y. Cui, M. Deng, L. You, B. Gao, and M. K. Tey, Phys. Rev. A 98, 042708 (2018).

[44] T. Busch, B.-G. Englert, K. Rzążewski, and M. Wilkens, Found. Phys. 28, 549 (1998).

[45] E. L. Bolda, E. Tiesinga, and P. S. Julienne, Phys. Rev. A 66, 013403 (2002).

[46] D. Blume and C. H. Greene, Phys. Rev. A 65, 043613 (2002).

[47] Z. Idziaszek and T. Calarco, Phys. Rev. A 74, 022712 (2006).

[48] J. F. Bertelsen and K. Mølmer, Phys. Rev. A 76, 043615 (2007).

[49] See supplementary material.

[50] A. Derevianko, J. F. Babb, and Dalgarno, Phys. Rev. A 63, 052704 (2001).

[51] S. G. Porsev and A. Derevianko, J. Chem. Phys. 119, 844 (2003).
The simplest description of the interaction between two alkali-metal atoms is a three-parameter model based on a single-scale multichannel quantum defect theory (MQDT) [S1–S5]. Here “single-scale” corresponds to the fact the theory is built on the solutions for the long-range potential $-C_6/r^6$ which has a single length scale $\beta_6 = (2\mu C_6/\hbar^2)^{1/4}$. And the three parameters can be taken as the singlet s-wave scattering length $a_S$, the triplet s-wave scattering length $a_T$, and the $C_6$ coefficient. The theory provides the simplest description of low-energy alkali interactions with or without magnetic field, including magnetic Feshbach resonances in all partial waves.

Depending on the specific system, the description of the magnetic Feshbach spectrum by this simple model has a typical accuracy of a few percent to about 8% for heavier systems with large hyperfine splittings [S5]. To understand these deviations and their improvements, one can keep in mind that an accurate description of a magnetic Feshbach resonance requires a simultaneous accurate descriptions of both the open and the closed channels, in particular the bound spectrum in the closed channels over an energy range corresponding to the channel energy spacing. For alkali-metal atoms, the channels are separated by the hyperfine splittings, and the requirement for accurate Feshbach spectrum translates into the requirement of accurate bound spectrum over a binding energy range of $|\Delta E^{\text{hf}}|$. Thus even when we are at ultracold temperatures where a single scattering length would suffice to describe the interaction in the open channel, the understanding of the scattering length itself, its relation to scattering lengths in other channels, and its tuning through the Feshbach spectrum, would require an underlying understanding over a much broader range of energies of at least $|\Delta E^{\text{hf}}|$. The validity and the accuracy of single-scale MQDT depends on the validity that the energy variation over this energy range is due solely to the $-C_6/r^6$ potential, which is less valid for systems with large hyperfine splittings such as our system with a Cs atom that has a hyperfine splitting of around $9$ GHz or $0.4$ K.

There are different options for improving upon this single-scale baseline result. Focusing on effective theories that do not rely on the details of the short-range potential, the easiest and the simplest improvement to implement is to introduce parameters that characterize the energy dependence and the partial-wave dependence of the single-scale short-range parameters [S6, S7]. A more fundamental approach that requires the fewest number of extra parameters, an important criteria for a good effective theory of interaction, is to go to a shorter length scale through the inclusion of higher order terms in the asymptotic expansion for the potential, such as

$$-\frac{C_6}{r^6} - \frac{C_8}{r^8},$$

which is used as the reference potential for our 2-scale MQDT and is a more accurate representation of the real atomic interaction at shorter distances. The additional $-C_8/r^8$ terms has a corresponding length scale $\beta_8 = (2\mu C_8/\hbar^2)^{1/(n-2)} = (2\mu C_8/\hbar^2)^{1/6}$ that is smaller than $\beta_6$.

A 2-scale MQDT description of alkali-metal interactions thus uses only one more parameter, the $C_8$, than the single scale theory for a total of 4 parameters that can be taken as $a_S$, $a_T$, $C_6$, and the $C_8$. It is more accurate in its description of scattering lengths and Feshbach spectrum, and can cover a greater range of energies when needed, similar to what has been demonstrated in QDT for $-C_1/r - C_4/r^4$ potential [S8]. The theory is formally the same as the single-scale theory except for the details of the QDT functions. These details will be presented elsewhere.

We point out that similar theoretical analysis can also be carried out using numerical formulations of MQDT [S9–S12] and other numerical methods of similar spirit which are already multiscale (see, e.g. [S13, S14]). The main difference of our approach is that it tries to make the best use of the existing single-scale analytic solutions [S15], and is formulated to show explicitly how physics at different length scales are related, and the progression from the single-scale theory that is sufficient at small energies and requires fewer parameters, to multiscale theories at shorter length scales that requires more parameters. In such an approach, an extra parameter or the complexity of the theory is added.
only when necessary. This characteristic is important in the context of an effective theory of interactions, especially when the parameters of the theory need to be determined from a limited number of experimental measurements.

For our particular system of \(^{23}\text{Na}^{133}\text{Cs}\), we have taken the long-range potential parameters \(C_6 = 3227 \text{ a.u.}\) and \(C_8 = 3.681 \times 10^5 \text{ a.u.}\) from Docenko et al. \cite{S16}. They are much more reliably determined by theory \cite{S17, S18} than the parameters \(a_S\) and \(a_T\), which are sensitive to short-range complex molecular interactions. With still a limited number of experimental measurements, only the latter parameters are determined experimentally as discussed in the main text. With more future measurements, especially those related to more deeply bound molecular states, we anticipate more precise determinations of all parameters including the \(C_6\) and \(C_8\) coefficients in a future study.

### INTERACTION SHIFT CALCULATION

In the limit of small collision energy, the molecular potential \(V(r)\) can be replaced with a Fermi pseudopotential consisting of the scattering length \(a\) and a regularized \(\delta\)-potential,

\[
V(|r|) = \frac{2\pi\hbar^2a}{\mu} \delta^{(3)}(r) \frac{\partial}{\partial r} r.
\]

For low energies, the Fermi pseudopotential reproduces the correct wavefunction outside the range of interactions. The Fermi pseudopotential approximation accuracy is characterized by the ratio of the characteristic van der Waal’s length \(\beta_0 = (2\mu C_6/R^2)^{1/4}\) to the relative oscillator length \(\beta_R = \sqrt{\hbar/m\omega_R}\) \cite{S19, S20}. The Na mass is 23 amu and Cs mass is 133 amu. The measured trapping frequencies are \((\omega_{\text{Na},x}, \omega_{\text{Na},y}, \omega_{\text{Na},z}) = 2\pi \times (109, 118, 20) \text{ kHz},\) and \((\omega_{\text{Cs,x}}, \omega_{\text{Cs,y}}, \omega_{\text{Cs,z}}) = 2\pi \times (130, 140, 24) \text{ kHz}.\) In our experiment, \(\beta_0 = 6 \text{ nm}\) is much smaller than the relative oscillator lengths \(\beta_{R,\text{Axial}} = 158 \text{ nm}\) and \(\beta_{R,\text{Radial}} = (65, 67) \text{ nm}\).

The Hamiltonian for two different atoms interacting in an anisotropic 3D harmonic trap is given by

\[
H = \sum_{i=x,y,z} \left( \frac{1}{2} m_1 \dot{x}_{1,i}^2 + \frac{1}{2} m_1 \omega_{1,i}^2 x_{1,i}^2 \right) + \sum_{i=x,y,z} \left( \frac{1}{2} m_2 \dot{x}_{2,i}^2 + \frac{1}{2} m_2 \omega_{2,i}^2 x_{2,i}^2 \right) + V(|r_1 - r_2|),
\]

where \(V(|r_1 - r_2|)\) is the spherically symmetric molecular potential for the two atoms. The trapping frequencies are in general different for the two atoms due to different polarizabilities and masses. The trapping frequencies can also be different for all three axes in an anisotropic trap. In terms of the relative and center-of-mass coordinates, \(r_R = r_1 - r_2\) and \(r_C = \frac{m_1 r_1 + m_2 r_2}{m_1 + m_2}\), the Hamiltonian can be re-expressed in terms of the reduced mass \(\mu = \frac{m_1 m_2}{m_1 + m_2}\), total mass \(M = m_1 + m_2\), relative frequencies \(\omega_{R,i} = \sqrt{\frac{m_2 \omega_{1,i}^2 - m_1 \omega_{2,i}^2}{m_1 + m_2}}\), and center-of-mass frequencies \(\omega_{C,i} = \sqrt{\frac{m_1 \omega_{1,i}^2 + m_2 \omega_{2,i}^2}{m_1 + m_2}}\) as

\[
H = \sum_{i=x,y,z} \left( \frac{1}{2} M \dot{x}_{C,i}^2 + \frac{1}{2} M \omega_{C,i}^2 x_{C,i}^2 \right) + \left( \sum_{i=x,y,z} \left( \frac{1}{2} \mu \dot{x}_{R,i}^2 + \frac{1}{2} \mu \omega_{R,i}^2 x_{R,i}^2 \right) + V_{int}(|r_R|) \right) + \sum_{i=x,y,z} \mu(\omega_{C,i}^2 - \omega_{R,i}^2)x_{R,i}x_{C,i}.
\]

The center-of-mass part of the Hamiltonian is just a single particle 3D harmonic oscillator. The second term, the relative Hamiltonian, is a 3D harmonic oscillator with a regularized \(\delta\)-function at the origin, for which analytic solutions exist for cases that the trap has spherical \cite{S21} and cylindrical \cite{S22} symmetries.

The final term in the Hamiltonian in Eq. S.3 couples the center-of-mass and relative coordinates. If the trapping frequencies of the two atoms are the same for each axis (for example in the case when the two atoms are the same species), then the last term vanishes, and then the relative and center-of-mass coordinates are separable. But when the two atoms are different species, as in our case, then this term can be important, which is discussed for the spherical case in Refs. \cite{S23, S24}. The trapping frequencies depend on the polarizability and mass as \(\sqrt{\alpha(\lambda)/m}\). At the tweezer wavelength of \(\lambda = 976 \text{ nm}\), the measured Cs trapping frequencies are \(\approx 19\%\) larger than those of Na.

To find the eigenenergies of the full Hamiltonian of Eq. S.3, we first ignore the last term so that the Hamiltonian is separable into center-of-mass and relative coordinates. In our experiment, the axial trapping frequency (along the axis of the tweezer and labeled as \(z\)) is approximately 5.6 times smaller than the two radial trapping frequencies (\(x\) and \(y\)). There is also a \(7\%\) difference between the two radial axes, but we initially assume they are the same and add the difference later as a correction. Therefore, we use the cylindrical harmonic oscillator wavefunctions as the solutions of the center-of-mass Hamiltonian, and they are labeled as \(|n,l,m_z\rangle\), where \(n\) and \(l\) are the principal and
angular momentum quantum numbers for the radial part, and \( m_z \) is quantum number for 1D harmonic oscillator for the axial part, and have eigenenergies

\[
E_{n,l,m_z}/(\hbar \omega_z) = (2n + |l| + 1)\eta + (m_z + 1/2),
\]

where we define \( \eta \) as the ratio of the radial to axial trapping frequency.

For the relative Hamiltonian, we use the analytic cylindrical solutions from Ref. [S22]. These solutions require that the axial trapping frequency is an integer multiple \( \eta \) of the radial trapping frequency. We define \( \eta = 6 \), which is close to the actual values of 5.6, and we will include the remaining terms later as a correction. The analytic solutions are given for the interacting states, but there also many relative states which have zero wavefunction at the \( \delta \)-function, and therefore are unaffected. For example any state with \( l \neq 0 \) or odd \( m_z \) has a zero at \( \delta \)-function. The complete basis includes both the interacting states from Ref. [S22] as well as all of the non-interacting states. The non-interacting states are solutions to the cylindrical harmonic oscillator, and so are just cylindrical harmonic oscillator wavefunctions.

One complication is that when \( \eta \), the ratio of the radial to the axial trapping frequency, is an integer, there is a subspace of cylindrical harmonic oscillator states with \( l = 0 \) and even \( m_z \) that are degenerate and have the same energy from Eq. S.4. In each degenerate subspace with \( N_{\text{deg}} \) states, the non-interacting states are a linear superposition of the degenerate eigenstates \( \psi_i \). We find these amplitudes \( c_i \) using a Gram-Schmidt procedure, which requires that \( \sum_{i=1}^{N_{\text{deg}}} c_i \psi_i(0) = 0 \). In each subspace, there is only one interacting state, for which the analytic solution is used, and \( N_{\text{deg}} - 1 \) non-interacting states.

For the interacting states, the energies are given by the transcendental equations [S22]

\[
\mathcal{F}(-(E - E_0)/2, \eta) = -\sqrt{2\pi}/a,
\]

where \( \mathcal{F}(x, \eta) \) is given by

\[
\mathcal{F}(x, \eta) = \frac{\sqrt{\pi} \Gamma(x)}{\Gamma(x + \frac{1}{2})} \sum_{m=1}^{n-1} F(1, x; x + \frac{1}{2}; e^{i(2\pi m/\eta)}) - \frac{2\sqrt{\pi} \Gamma(x)}{\Gamma(x - \frac{1}{2})}.
\]

Here \( F(a, b; c, x) \) denotes the hypergeometric function and \( \Gamma(x) \) is the Euler gamma function. The energy \( E \) and \( E_0 \) are in units of the axial trap energy \( \hbar \omega_z \), and so the ground state energy \( E_0 = \eta + \frac{1}{2} \).

Now that we have solution in the separable and cylindrical case, the next step is to include the non-separable and asymmetric correction terms by diagonalizing the total matrix in the combined center-of-mass and relative cylindrical bases. For the matrix, we include all states with energies up to \( 20 \omega_{R,z} \). The matrix elements are calculated numerically using the cylindrical wavefunctions, which for completeness are given here:

\[
\Psi_{n,l,m_z}(\rho, \theta, z) = \Psi_{n,l}^{\text{radial}}(\rho, \theta) \Psi_{m_z}^{\text{axial}}(z),
\]

with the normalized radial harmonic oscillator wavefunction

\[
\Psi_{n,l}^{\text{radial}}(\rho, \theta) = \sqrt{\frac{2^n}{a_z^2(n + |l|)!}} e^{-\rho^2/(2a_z^2)} (r/a_\perp)^{|l|} L_n^{(|l|)}(r^2/a_\perp^2) e^{i\theta} \sqrt{2\pi},
\]

and the normalized 1D harmonic wavefunction

\[
\Psi_{m_z}^{\text{axial}}(z) = \frac{1}{\sqrt{2m_z!a_z^2}} \frac{1}{\sqrt{\pi}} e^{-z^2/(2a_z^2)} H_{m_z}(z/a_z).
\]

Here the radial and relative oscillator lengths are defined as \( a_\perp = \sqrt{\hbar/(\mu \omega_\perp)} \) and \( a_z = \sqrt{\hbar/(\mu \omega_z)} \). \( H_{m_z} \) are the Hermite-Gaussian functions, and \( L_n^{(|l|)} \) are the generalized Laguerre polynomials.

The eigenenergies of the matrix are calculated as a function of the scattering length, which is shown in Fig. 1 of the main text.

**PERTURBATION THEORY**

We use first-order perturbation theory to estimate the shift of the Na-Cs 3D motional ground state due to the interaction, which is approximated by the Fermi pseudopotential interaction \( V(|\mathbf{r}_1 - \mathbf{r}_2|) = \frac{2\pi \hbar^2 a}{\mu} \delta^{(3)}(|\mathbf{r}_1 - \mathbf{r}_2|) \partial \overline{r} \).
Using the Cartesian harmonic oscillator bases \(|n_{Na,x}^{Na}, n_{Na,y}^{Na}, n_{Na,z}^{Na}; n_{Cs,x}^{Cs}, n_{Cs,y}^{Cs}, n_{Cs,z}^{Cs}\rangle\), first-order perturbation theory gives a ground state shift of \(\Delta E_g \approx \frac{2\hbar^2}{\mu} \langle 0, 0, 0; 0, 0, 0 \rvert \partial^3 \rangle r_1 - r_2) \frac{\partial}{\partial r_1^{\beta(3)}} \langle 0, 0, 0; 0, 0, 0 \rangle\), which simplifies to

\[
\Delta E_g \approx a \left( \frac{2\hbar^2}{\mu \sqrt{\pi}} \frac{1}{\beta_{\text{eff}}^3} \right),
\]  

(S.10)

where the effective 3D oscillator length is defined in terms of the 1D oscillator lengths,

\[
\beta_{\text{eff}} = \left( (\beta_{Na,x}^{Na})^2 + \beta_{Cs,x}^{Cs} ) (\beta_{Na,y}^{Na})^2 + \beta_{Cs,y}^{Cs} ) (\beta_{Na,z}^{Na})^2 + \beta_{Cs,z}^{Cs} \right)^{1/6}.
\]  

(S.11)

The oscillator lengths are \(\beta = \sqrt{\frac{\hbar}{m\omega}}\), where \(m\) is the mass of the atom and \(\omega\) is the trapping frequency.

---

[S1] B. Gao, E. Tiesinga, C. J. Williams, and P. S. Julienne, Phys. Rev. A 72, 042719 (2005).
[S2] T. M. Hanna, E. Tiesinga, and P. S. Julienne, Phys. Rev. A 79, 040701 (2009).
[S3] B. Gao, Phys. Rev. A 84, 022706 (2011).
[S4] C. Makrides and B. Gao, Phys. Rev. A 89, 062718 (2014).
[S5] Y. Cui, M. Deng, L. You, B. Gao, and M. K. Tey, Phys. Rev. A 98, 042708 (2018).
[S6] M. Li, L. You, and B. Gao, Phys. Rev. A 89, 052704 (2014).
[S7] M. Li and B. Gao, Phys. Rev. A 91, 032702 (2015).
[S8] H. Fu, M. Li, M. K. Tey, L. You, and B. Gao, New J. Phys. 18, 103016 (2016).
[S9] F. H. Mies, J. Chem. Phys. 80, 2514 (1984).
[S10] F. H. Mies and P. S. Julienne, J. Chem. Phys. 80, 2526 (1984).
[S11] J. P. Burke, C. H. Greene, and J. L. Bohn, Phys. Rev. Lett. 81, 3355 (1998).
[S12] B. P. Ruzic, C. H. Greene, and J. L. Bohn, Phys. Rev. A 87, 032706 (2013).
[S13] E. G. M. van Kempen, S. J. J. M. F. Kokkelmans, D. J. Heinzen, and B. J. Verhaar, Phys. Rev. Lett. 88, 093201 (2002).
[S14] R. Pires, M. Repp, J. Ulmanis, E. D. Kuhne, M. Weidemüller, T. G. Tiecke, C. H. Greene, B. P. Ruzic, J. L. Bohn, and E. Tiemann, Phys. Rev. A 90, 012710 (2014).
[S15] B. Gao, Phys. Rev. A 58, 1728 (1998).
[S16] O. Docenko, M. Tamanis, J. Zaharova, R. Ferber, A. Pashov, H. Knöckel, and E. Tiemann, J. Phys. B 39, S929 (2006).
[S17] A. Derevianko, J. F. Babb, and A. Dalgarno, Phys. Rev. A 63, 052704 (2001).
[S18] S. G. Porsev and A. Derevianko, J. Chem. Phys. 119, 844 (2003).
[S19] E. L. Bolda, E. Tiesinga, and P. S. Julienne, Phys. Rev. A 66, 013403 (2002).
[S20] D. Blume and C. H. Greene, Phys. Rev. A 65, 043613 (2002).
[S21] T. Busch, B.-G. Englert, K. Rzążewski, and M. Wilkens, Found. Phys. 28, 549 (1998).
[S22] Z. Idziaszek and T. Czarnecki, Phys. Scr. 82, 065502 (2005).
[S23] J. F. Bertelsen and K. Mølmer, Phys. Rev. A 76, 043615 (2007).
[S24] F. Deuretzbacher, K. Plassmeier, D. Pfannkuche, F. Werner, C. Ospelkaus, C. Ospelkaus, K. Sengstock, and K. Bongs, Phys. Rev. A 77, 032726 (2008).