

Ground-state scattering lengths for potassium isotopes determined by double-resonance photoassociative spectroscopy of ultracold ^{39}K

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We use double-resonance photoassociative spectroscopy of ultracold ^{39}K atoms to precisely determine the triplet $a^3\Sigma_u^+$ scattering length for the various isotopes of potassium. Photoassociation of free ^{39}K atoms to the pure long-range $0_g^-(v'=0, J'=2)$ level is followed by stimulated emission to high-lying levels of the $a^3\Sigma_u^+$ potential. The binding energies of levels within 5 GHz below the lowest ground-state $4s_{1/2}(f_a=1) + 4s_{1/2}(f_b=1)$ hyperfine asymptote are measured by both trap loss and ionization detection. The locations of these near-threshold hyperfine-coupled molecular levels allow us to constrain the triplet potential and thereby determine the triplet scattering length. The result for ^{39}K , $a_t = -33 \pm 5 a_0$ ($1a_0 = 0.0529177 \text{ nm}$), is a factor of ~ 5 improvement over previous determinations and establishes that a large ^{39}K Bose-Einstein condensate will not be stable.

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Ultracold collisional properties of ground-state atoms are dominated by s -wave scattering lengths. These quantities determine the static and dynamic properties of a Bose-Einstein condensate [1–4], elastic cross sections, and the thermalization rates. The accurate determination of scattering lengths is challenging because of their extreme sensitivity to the weak, near-threshold long-range interatomic potentials [5–7]. Photoassociative spectroscopy (PAS) of ultracold atoms is well suited to this, and, along with other methods, has been utilized to determine scattering lengths for various alkali atoms: Li [8,9], Na [10,11], K [12–14], Rb [15–17], and Cs [18,19]. Double-resonance PAS is particularly useful [8,9,15], since it probes the highest-lying vibrational levels of two bound ground-state atoms. In the present work, we perform double-resonance PAS of ultracold ^{39}K atoms near the $4s_{1/2} + 4s_{1/2}$ atomic asymptote, thereby reducing the uncertainty of the triplet $a^3\Sigma_u^+$ scattering length, a_t , by a factor of ~ 5 from previous measurements [12–14]. This allows us to determine that $a_t < 0$ for ^{39}K . We also significantly improve the values of a_t for the bosonic isotope ^{41}K , the fermionic isotope ^{40}K , and for mixed-isotope interactions.

Three previous measurements of a_t for potassium isotopes have been reported [12–14]. Two are based on high-resolution PAS of the two pure long-range states of $^{39}\text{K}_2$, 0_g^- and 1_u . In the 0_g^- state analysis, the rotational

intensity distribution of seven vibrational levels, $v=0-6$, is used to determine a triplet scattering length of $a_t = -17 - 0.045(C_6 - \bar{C}_6) \pm 25 a_0$ [12], where $\bar{C}_6 = 3800 \text{ a.u.}$, $1 \text{ a.u.} = 0.0957345 \text{ yJ (nm)}^6$, and $1a_0 = 0.0529177 \text{ nm}$. A similar analysis using the 1_u -state spectrum gives $-60a_0 < a_t < +15a_0$ [13]. In the third experiment, the thermalization rate of cold ^{40}K atoms is measured and used to derive the cold collision parameters for all three potassium isotopes, giving a result for ^{39}K of $-80a_0 < a_t < -28a_0$ [14]. These measurements indicate that a_t is probably small and negative for ^{39}K , but the need for further improvement is obvious.

In this paper, we use double resonance PAS of ultracold ^{39}K atoms near the ground $4s_{1/2} + 4s_{1/2}$ atomic asymptote to accurately measure the binding energies of several hyperfine and rotational components of the next to last vibrational level of the $a^3\Sigma_u^+$ potential. We then obtain a greatly improved value for a_t by determining the best fit between theoretical bound states obtained from a coupled-channel calculation and the experimental binding energies.

The experimental setup has been described in previous publications [20,21]. Figure 1 shows the level scheme for the Λ -type pump-dump double resonance transition. A pair of potassium atoms, each in the $f_\alpha=1$ ground state is initially photoassociated at ν_1 to the $v'=0, J'=2$ rovibrational level of the pure long-range 0_g^- state, about 180 GHz below the $4s_{1/2} + 4p_{3/2}$ asymptote. f_α with $\alpha=a(b)$ denotes the total atomic hyperfine quantum number for atom $a(b)$, respectively. A second photon ν_2 then stimulates transitions to bound levels of the lowest triplet state $a^3\Sigma_u^+$, just below the $4s_{1/2} + 4s_{1/2}$ ground-state asymptote. This experiment uses the intermediate 0_g^- state because it is accessible by photoassociation from $f_a=1 + f_b=1$ atomic collisions and because it has a large transition probability to weakly bound vibrational levels of the $a^3\Sigma_u^+$ state. However, the Franck-

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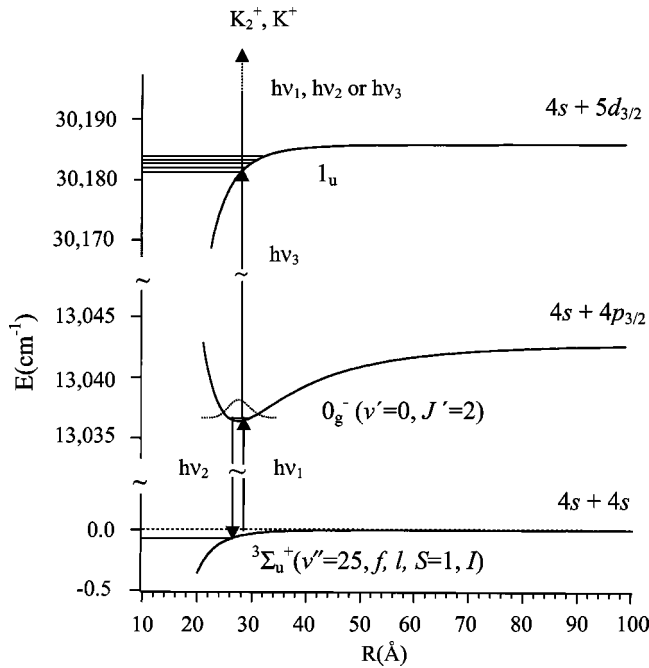


FIG. 1. Level scheme for Λ -type two-photon photoassociation spectroscopy. For the ionization depletion detection scheme, an additional photon ν_3 excites molecules from the 0_g^- intermediate state to a highly excited autoionizing 1_u state below the $4s+5d$ atomic limit. Resonances appear as dips in the ion signal.

Condon factors only favor the two most weakly bound vibrational levels of the $a^3\Sigma_u^+$ state. Overlap of the 0_g^- vibrational wave functions with more deeply bound levels is negligible.

Two detection schemes are used. For a trap loss detection scheme, atomic fluorescence of the trapped ^{39}K atoms is collected, while for a second scheme ions are detected using an additional narrowband laser to excite the intermediate state to a highly excited autoionizing 1_u state below the $4s+5d$ limit [22]. When the dump laser is tuned to resonance with a $a^3\Sigma_u^+$ level, an increased trap loss and a decrease of the ion signal are observed. A heterodyne system measures the frequency difference between the dump and pump lasers.

An unusual feature of double resonance through the 0_g^- state is that photoassociation to the lowest vibrational levels (e.g., $v'=0-4$ for K_2) does not induce significant trap loss [20]. These vibrational levels have so little kinetic energy that when the molecules undergo bound-free radiative decay, the atoms do not gain enough kinetic energy to escape from the trap. Nevertheless, stimulated transitions to levels of the $a^3\Sigma_u^+$ state do cause loss as these molecules are not trapped by the MOT.

In Fig. 2, we show examples of spectra as a function of $\nu_2 - \nu_1$ tuned between 1 GHz and 6 GHz below the $f_a = 1 + f_b = 1$ threshold. A group of hyperfine-rotation coupled energy levels is observed in both the trap-loss and the ion-depletion spectra. The lines belong to the next to last vibrational level of the $a^3\Sigma_u^+$ state, $v''=25$. In principle, we should also observe the hyperfine-rotational structure of the

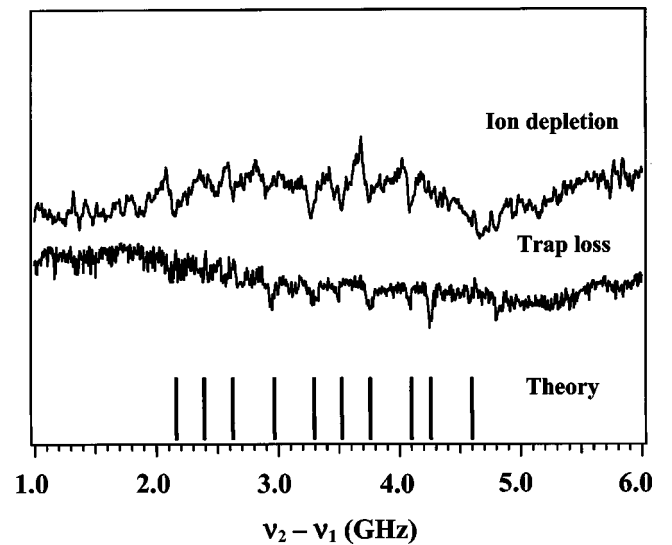


FIG. 2. Sample spectra of the hyperfine levels of the $v''=25$ level of the $a^3\Sigma_u^+$ state, between 1 GHz and 6 GHz below the $f_a = 1 + f_b = 1$ atomic threshold. Predicted line positions are shown for comparison. The broad dip in the ion signal centered at 4.7 GHz corresponds with a 0_u^+ photoassociation line for laser ν_2 .

last bound level ($v''=26$) of the $a^3\Sigma_u^+$ state about 1 GHz below the $f_a = 1 + f_b = 1$ asymptote. However, we have not found unambiguous lines in this region in either the trap loss or the ion-depletion spectrum. This is partially due to overlap of these molecular lines with strong atomic Raman resonances in the trap loss spectrum and with the one-photon photoassociation signals in the ion-depletion spectrum.

We observe twelve spectral lines between 1.5 GHz and 4.7 GHz by analyzing seven trap-loss spectral scans and ten ion-depletion spectral scans. Six (four) of these ten ion spectra use the $v'=0$ (2), $J'=2$, 0_g^- intermediate state, respectively. We accept as real spectral features only those lines that appear in at least four separate scans. The line positions of the $v'=0$ and 2 ion-depletion spectra differ on average by only 2 MHz. Some lines are better observed in trap loss or by ion detection. In fact, four lines are only visible in ion depletion. Table I shows the average of the line positions over all runs in which these lines are readily observable. The averages have a statistical uncertainty that is ≤ 20 MHz. However, we feel that the more significant uncertainty is systematic. The systematic uncertainties are a combination of two factors. The line positions in the trap-loss spectra are on average 29 MHz closer to the atomic threshold than the corresponding ion-depletion spectra while the ion-depletion spectra often show a dispersive type of line shape. For the dispersive spectra we nevertheless choose the minimum ion signal as the line position. Consequently, we believe that the total uncertainty of the line positions is approximately 40 MHz.

The spectra are assigned using a coupled-channels analysis [12,13,15]. The Hamiltonian used in this analysis contains the $X^1\Sigma_g^+[V_0(R)]$ and $a^3\Sigma_u^+[V_1(R)]$ potential, the nuclear rotation $\hbar^2\mathbf{I}^2/(2\mu R^2)$, and the atomic hyperfine interaction $V_{\text{HF}} = a_{\text{hf}}(\mathbf{i}_a \cdot \mathbf{s}_a + \mathbf{i}_b \cdot \mathbf{s}_b)$. Here, \mathbf{I} is the nuclear mechanical angular momentum, μ is the reduced mass, R is the

TABLE I. Comparison of the level energies between experiment and theory. All energies are relative to the ground-state $f_a = 1 + f_b = 1$ limit. The experimental uncertainty in the level positions is 40 MHz. The theoretical values are obtained using $a_t = -33a_0$ and $C_6 = 3897$ a.u. The $f = 1$ and 3 levels are degenerate. For clarity only the $f = 3$ label is presented. Not included in the table are two unassigned features in the $v' = 2$ ion-depletion spectrum at -1508 and -1869 MHz.

Measured (MHz)	Assignment $l(S,I)f$	Calculated (MHz)	Meas. - Calc. (MHz)
-2171	4 (1,3)4	-2167	-4
-2390	4 (1,1)2	-2394	+4
-2664	4 (1,3)3	-2628	-36
-2923	4 (1,3)2	-2969	+46
-3288	2 (1,3)4	-3292	+4
-3501	2 (1,1)2	-3520	+19
-3757	2 (1,3)3	-3754	-3
-4094	2 (1,3)2	-4095	+1
-4257	0 (1,3)3	-4250	-7
-4611	0 (1,3)2	-4591	-20

internuclear separation, a_{hf} is the atomic hyperfine constant, and \mathbf{s}_α and \mathbf{i}_α are the atomic electron spin and nuclear spin, respectively. The structure of the Hamiltonian ensures that \mathbf{I} and $\mathbf{f} = \mathbf{f}_a + \mathbf{f}_b = \mathbf{S} + \mathbf{I}$ are conserved. $\mathbf{S} = \mathbf{s}_a + \mathbf{s}_b$ and $\mathbf{I} = \mathbf{i}_a + \mathbf{i}_b$ denote the total electronic and nuclear spins of the molecule, respectively. Moreover, the identical ^{39}K nuclei require that only states with even $l + S + I$ exist.

The interaction V_{HF} lifts the hyperfine degeneracy of the $S = 1$ $a^3\Sigma_u^+$ state and causes singlet-triplet mixing. From perturbation theory the effect of V_{HF} on the $a^3\Sigma_u^+$ state is, after some algebra, given by $(1/4)a_{\text{hf}}[f(f+1) - S(S+1) - I(I+1)]$ with the restriction that for even (odd) l only the odd (even) I states exist. The $S = 0$ $X^1\Sigma_g^+$ state has a zero first-order hyperfine splitting. Strong singlet-triplet mixing occurs when the vibrational levels of the two potentials are closely spaced compared to $a_{\text{hf}}/h = 231$ MHz. For K_2 the vibrational spacings of both the $X^1\Sigma_g^+$ and $a^3\Sigma_u^+$ states are sufficiently large that only bound levels within the last GHz below the hyperfine asymptotes are strongly mixed. For a quantitative description of such states a full close-coupled calculation is necessary.

For our observed levels with binding energies that are larger than 1 GHz, singlet-triplet mixing can be ignored and a model in which the hyperfine interaction is added perturbatively to the rovibrational levels of the $V_S(R) + \hbar^2 l(l+1)/(2\mu R^2)$ potentials and S and I are good quantum numbers can be used. A comparison of a close-coupled calculation and the model shows agreement to within 5 MHz, which is much smaller than the experimental uncertainties.

Table I shows the assignments of the ten observed lines that can be unambiguously matched with the coupled-channels analysis. All states belong to the $v'' = 25$ vibrational level of the $S = 1$ $a^3\Sigma_u^+$ state. The observed lines are labeled by $l(S,I)f$. In the assignment, we have also used the fact that

TABLE II. Determinations of scattering lengths in a_0 of various isotopic combinations of K. The first column labels the isotopes of the colliding K partners. The next three columns give the triplet, $(f_a, f_b)fl = (1,1)2s$ and $(9/2,9/2)8s$ scattering lengths. The latter two, given where appropriate, are obtained from a close-coupled calculation that includes a $X^1\Sigma_g^+$ potential [13]. The $(9/2,9/2)8s$ scattering length for 40-40 has been independently determined to be $157 \pm 20a_0$ [14]. The error budget combines uncertainties in the experimental binding energies, the uncertainty in C_6 based on the values reported in Ref. [24], and the uncertainty in the ^{39}K singlet scattering length [13] where appropriate. The current experimental binding energies can not be used to determine C_6 .

	a_t	$a_{(1,1)2s}$	$a_{(9/2,9/2)8s}$
39-39	-33 ± 5	-45 ± 15	
40-40			169 ± 9
41-41	60 ± 2	65 ± 5	
39-40	-4000 ± 3000		
39-41	177 ± 8	160 ± 20	
40-41	97 ± 2		

odd l $a^3\Sigma_u^+$ and all $X^1\Sigma_g^+$ levels cannot be accessed for the downward transition from a $J' = 2$ 0_g^- rotational level [10]. In fact, for the $J' = 2$ level only $l = 0, 2,$ and 4 states can be observed.

The assigned levels are used in a least-squares fit to find a triplet potential that reproduces the bound states and consequently allows us to calculate the triplet scattering length. For the fitting procedure, an approximate potential for the $a^3\Sigma_u^+$ state is constructed from short-range Rydberg-Klein-Rees potentials [23] smoothly connected to an analytic long-range dispersion and exchange potential. This potential, however, is not sufficiently accurate to predict the level positions of weakly bound vibrational levels and ultracold scattering properties. Therefore we change the shape of this approximate potential by varying the C_6 dispersion coefficient and adding/subtracting a parametrized short-range correction potential $\Delta V_S(R)$ [12–14].

The best shape of the $a^3\Sigma_u^+$ potential is determined according to the reduced χ^2 criterion. Bounds for the uncertainty are determined by $\chi_{\text{min}}^2 + 1$, where $\chi_{\text{min}}^2 \approx 0.3$ is the smallest value of χ^2 . To determine the sensitivity of the fitting to the leading dispersion coefficient C_6 , the fitting process is repeated for several fixed values of C_6 ranging from 3600 a.u. to 4000 a.u. Unfortunately, the minimum of χ^2 is nearly independent of C_6 . We find a best fit for $a_t = -33 \pm 4 - 0.05\delta C_6 a_0$, where δC_6 is the deviation from $C_6 = 3897$ a.u., a recent high-precision calculation by Derevianko *et al.* [24], and the uncertainty of $\pm 4 a_0$ derives from the measurement uncertainty of 40 MHz. If the calculation of Ref. [24] is assumed to be correct, its uncertainty of 15 a.u. contributes only $1 a_0$ to the error budget, and our final result is $a_t = -33 \pm 5 a_0$.

Table II shows scattering lengths of various isotopic combinations of K using the appropriate reduced mass in the Hamiltonian and observing that within the Born-Oppenheimer approximation the $X^1\Sigma_g^+$ and $a^3\Sigma_u^+$ potentials are the same for all isotope combinations. For the bosonic

isotopes the a_t scattering length characterizes the collision between two doubly polarized atoms. The $a_{(f_a, f_b)fl} = a_{(1,1)2s}$ scattering length characterizes the collisions between two $f_a=1$, $m_a=-1$ atoms where m_α is the magnetic projection quantum number of f_α . For the fermionic ^{40}K collision the $a_{(9/2,9/2)8s}$ scattering length describes the collision between an $f_a=9/2, m_a=9/2$ and an $f_b=9/2, m_b=7/2$ atom. Both $a_{(1,1)2s}$ and $a_{(9/2,9/2)8s}$ scattering lengths depend on the $X^1\Sigma_g^+$ potential [13]. Note that if the range of C_6 of Ref. [24] is found to be incorrect then the values quoted in Table II will have to be adjusted. In principle, the unambiguous observation of the next vibrational level downward would allow theory to extract C_6 and its uncertainty.

In summary, we have observed hyperfine-coupled molecular bound levels in a region between 1 GHz and 6 GHz below the $4s(f_a=1) + 4s(f_b=1)$ atomic asymptote of $^{39}\text{K}_2$ by double-resonance photoassociative spectroscopy. The observed levels belong to the next to last vibrational level $v''=25$ of the lowest triplet state $a^3\Sigma_u^+$. Using the purely long-range 0_g^- state as the intermediate state greatly simplifies the observed lower-state spectrum since only levels with triplet

character are dipole-allowed and observed. Binding energies of the observed molecular lines are analyzed by the coupled-channels calculation and used to determine the triplet scattering length for the various isotopes. The result for ^{39}K , $a_t = -33 \pm 5a_0$ reduces the uncertainty of the triplet scattering length by a factor of five compared to previous results derived from the excited-state spectroscopy. The unambiguously negative triplet scattering length shows that ^{39}K cannot be cooled into a stable Bose-Einstein condensate in a large sample unless the Feshbach resonances predicted in Ref. [7] can be utilized. However, the predicted positive scattering lengths for ^{41}K show that it is a good candidate for Bose-Einstein condensation.

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