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Interisotope Determination of Ultracold Rubidium Interactions from Three High-Precision Experiments

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Combining the measured binding energies of four of the most weakly bound rovibrational levels of the $^{87}\text{Rb}_2$ molecule with results of two other recent high-precision experiments, we obtain exceptionally strong constraints on the atomic interaction parameters in a highly model independent analysis. The comparison of ^{85}Rb and ^{87}Rb data, where the two isotopes are related by a mass scaling procedure, plays a crucial role. We predict scattering lengths, clock shifts, and Feshbach resonances with an unprecedented level of accuracy. Two of the Feshbach resonances occur at easily accessible magnetic fields in mixed-spin channels. One is related to a d -wave shape resonance.

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After the first realization of Bose-Einstein condensation (BEC) in a dilute ultracold gas of rubidium atoms [1], experiments with the two isotopes ^{87}Rb and ^{85}Rb further led to an amazingly rich variety of BEC phenomena, ranging from the controlled collapse of a condensate with tunable attractive interactions [2] to the realization of an atomic matter wave on a microchip [3]. Because of the large number of groups that have started doing experiments with these atomic species and the growing complexity and subtlety of the planned experiments, there is a clear need for a more precise knowledge of the interactions between ultracold rubidium atoms in the electronic ground state, since these determine most of the properties of the condensate. For instance, despite a widespread interest, to our knowledge, not until now has any experimental group been able to locate the predicted [4] magnetic-field induced Feshbach resonances that can be used to tune the interactions between ultracold ^{87}Rb atoms. Being able to switch on or off these interactions at will by a mere change of magnetic field may well be one of the main assets of matter waves compared to light waves in the new matter wave devices. In an atomic interferometry device, in particular, a nonlinear interaction between interfering waves may be introduced or eliminated by changing a field applied at the intersection point.

In this Letter, combining the results of three very recent high-precision observations, we come close to a complete and model-independent specification of the interaction properties of ultracold rubidium atoms. The fact that two isotopes ^{85}Rb and ^{87}Rb are involved in the measurements makes the constraints exceptionally strong and also increases the predictive power: The interaction properties of any other fermionic or bosonic isotope with mass number 82, 83, 84, or 86 are now known with about the same precision. Using mass scaling to relate the different isotopes, we are able for the first time to deduce for each of the isotopes the exact numbers of bound Rb_2 states with total spin $S = 0$ (singlet) and 1 (triplet). As an illustration of the predictive power, we predict two Feshbach

resonances in mixed-spin scattering channels for ^{87}Rb at easily accessible fields that could lead to new time dependent phenomena in coherent spin oscillations and spin waves. There are numerous effects, such as spinor condensate energy differences, which are proportional to differences of scattering lengths. Because these differences are unusually small in Rb, the potentials must be very accurate to calculate them to reasonable accuracy.

The first of the three high-precision experiments is the recent measurement of four of the highest bound rovibrational levels of the $^{87}\text{Rb}_2$ molecule with 10 kHz precision [5]. The second experiment is the improved characterization [6] of the elastic scattering near a Feshbach resonance in ^{85}Rb , leading to a more precise determination of the resonance field $B_0 = 154.9(4)$ G and the nearby field strength $B'_0 \equiv B_0 + \Delta = 165.85(5)$ G, where the scattering length goes through zero [Δ is the (elastic) resonance width]. The third experimental ingredient going into our analysis is the measurement [7] of 12 148 transition frequencies between $X^1\Sigma_g^+$ vibrational levels of the $(^{85}\text{Rb})_2$, $(^{87}\text{Rb})_2$, and $^{85}\text{Rb}^{87}\text{Rb}$ molecules, leading to a highly accurate singlet Rb + Rb potential [8]. Moreover, within the accuracy of this experiment, a comparison of levels for the three studied isotopomers shows no sign of Born-Oppenheimer breakdown effects, i.e., the observed levels agree with a simple radial Schrödinger equation containing a common singlet potential $V_S(r)$ and the reduced atomic mass. Calculation shows [9] that this justifies neglecting such effects also in our analysis.

This set of extremely precise measurements calls for a very careful construction of the interatomic total spin $S = 0$ and 1 potentials, depending on the interatomic separation r . We combine the singlet potential of Ref. [7] with a long-range part equal to the difference $V_{\text{disp}} - V_{\text{exch}}$ of a dispersion term and an exchange term, starting at a variable radius r_S between $21a_0$ and $23.5a_0$ ($1a_0 = 0.529$ Å). The part $V_{\text{disp}}(r)$ includes C_6 , C_8 , and C_{10} terms and retardation, while $V_{\text{exch}}(r)$ is assumed to be given by the asymptotic form $\frac{1}{2}Jr^{7/2\alpha-1}\exp(-2\alpha r)$, derived by Smirnov and

Chibisov [10] for r values where the overlap of the electron clouds is sufficiently small [$\alpha = 0.554a_0^{-1}$, following from the ionization potential $\frac{1}{2}\alpha^2$ of the Rb atom in atomic units (a.u.)].

The triplet potential is subject to a larger uncertainty. For its short-range part an *ab initio* potential is usually taken. To get rid of this model dependence, we use the accumulated phase method [11]: The “history” of the atom-atom motion is summarized by a boundary condition at an interatomic distance r_0 , in the form of the phase $\phi_T(E, l)$ of the oscillating triplet radial wave function ψ depending on energy E and angular momentum l . Specifying $\phi_T(E, l)$ is equivalent to giving the logarithmic derivative ψ'/ψ at $r = r_0$. In all of our previous work, we neglected the singlet-triplet mixing by the hyperfine interaction V_{hf} of the nuclear and electronic spins in the range $r < r_0$, in order to deal with pure singlet and triplet radial waves until the boundary. Here, however, we introduce a new variant that allows us to choose a larger r_0 than would otherwise be possible: We include the adiabatic mixing by V_{hf} in the two-atom spin states but still neglect its influence on the radial wave functions to avoid dependence on the history other than via the pure triplet phase. Model calculations show that in this form the scattering calculations have the required accuracy for r_0 values up to $16a_0$. The experimental data for either ultracold or weakly bound atoms that we analyze comprise a small E and l range near $E = l = 0$. In this range, a first-order Taylor expansion $\phi_T(E, l) = \phi_T^0 + E\phi_T^E + l(l+1)\phi_T^l$ is adequate, which reduces the information contained in $V_T(r)$ for $r < r_0$ to three phase parameters only. In principle, these would be needed for both the ^{85}Rb and ^{87}Rb systems. However, since we expect Born-Oppenheimer breakdown effects to be negligible also for the triplet channel in the distance range $r < r_0$, we use mass scaling to express ϕ_T^0 , ϕ_T^E , and ϕ_T^l for ^{85}Rb in terms of the three phase parameters for ^{87}Rb . Beyond r_0 we construct $V_T(r)$ from $V_S(r)$ by adding $2V_{\text{exch}}(r)$.

Applying this method, we carry out a full quantum scattering calculation for a set of eight experimentally measured quantities. This set consists of five quantities for ^{87}Rb and three for ^{85}Rb . The ^{87}Rb data are the four bound state energies and the ratio of scattering lengths $a_{1-1}/a_{21} = 1.062(12)$ for atomic scattering in condensates of ^{87}Rb atoms in the hyperfine states $(f, m_f) = (1, -1)$ and $(2, 1)$ [12]. For ^{85}Rb we include the Feshbach resonance fields B_0 and B'_0 , as well as the energy 0.7(1) mK of the g -wave shape resonance observed in the scattering of a pair of cold atoms in the total spin $S = 1$ state [13].

With a least-squares search routine, we determine optimal values for the parameters C_6 , C_8 , J , $\phi_T^0(^{87}\text{Rb})$, $\phi_T^E(^{87}\text{Rb})$, and $\phi_T^l(^{87}\text{Rb})$. C_{10} is kept fixed at the value calculated by Marinescu *et al.* [14], but the effect of $\pm 10\%$ variations around this value and an estimated upper bound for the influence of higher dispersion terms are included in the final error bars. Column A of Table I summarizes

TABLE I. Interaction parameters (a.u.) derived from experiments without (column A) and including (column B) the requirement $V_S = V_{\text{disp}} - V_{\text{exch}}$ for $r_0 < r < r_S$.

Quantity	A	B
$C_6/10^3$	4.703(9)	4.698(4)
$C_8/10^5$	5.79(49)	6.09(7)
$C_{10}/10^7$	7.665 (Ref. [14])	7.80(6)
$C_{11}/10^9$...	-0.86(17)
$C_{12}/10^9$...	11.9 (Ref. [22])
$J \cdot 10^2$	0.45(6)	0.42(2)
$a_T(^{87}\text{Rb})$	+98.98(4)	+98.99(2)
$a_S(^{87}\text{Rb})$	+90.4(2)	+90.0(2)
$a_T(^{85}\text{Rb})$	-388(3)	-387(1)
$a_S(^{85}\text{Rb})$	+2795 $^{+420}_{-290}$	+2400 $^{+370}_{-150}$
$\nu_{DT}(\text{mod } 1)$, $n_{bT}(^{87}\text{Rb})$	0.4215(3), 41	0.4214(2), 41
$\nu_{DS}(\text{mod } 1)$, $n_{bS}(^{87}\text{Rb})$	0.455(1), 125	0.456(1), 125
$\nu_{DT}(\text{mod } 1)$, $n_{bT}(^{85}\text{Rb})$	0.9471(2), 40	0.9470(1), 40
$\nu_{DS}(\text{mod } 1)$, $n_{bS}(^{85}\text{Rb})$	0.009(1), 124	0.011(1), 124

the main results of the calculations. We find a value for C_6 in agreement with the theoretical value 4691(23) obtained by Derevianko *et al.* [15]. The C_8 value agrees with that calculated by Marinescu *et al.* [14]. To our knowledge, this is the first experimental determination of C_8 from a combined set of cold-atom + bound state data. Our analysis also yields the first experimental value of the strength of V_{exch} from such data. The coefficient J agrees with the most recent theoretical value in Ref. [16]. Table I also gives the values of the pure singlet and triplet scattering lengths for both ^{85}Rb and ^{87}Rb , following from C_6 , C_8 , J , and $\phi_T^0(^{87}\text{Rb})$, as well as the fractional vibrational quantum numbers at dissociation $\nu_D(\text{mod } 1)$ [11] and the numbers of bound states n_b . The reduced minimum χ^2 value is 0.5.

The foregoing makes clear that a major step forward has been made possible by the new experiments, two of which make use of a Bose-Einstein condensate. This is a firm basis for making a variety of interesting predictions. As a first example, we predict the ^{87}Rb $f = 1$ spinor condensate to be ferromagnetic, i.e., it is favorable for two $f = 1$ atoms to have their spins parallel, because the mean field interaction is more repulsive for total $F = 0$ than for $F = 2$: The calculated scattering lengths are $a_{F=2} = +100.4(1)a_0$ and $a_{F=0} = +101.8(2)a_0$. In a recent preprint, Klausen *et al.* [17] independently came to this conclusion of a ferromagnetic spinor condensate by calculating the scattering lengths for several assumed numbers of triplet bound states.

We are also able to predict collisional frequency shifts in an ^{87}Rb fountain clock for arbitrary choices of partial densities of atomic hyperfine states. Table II compares our calculated fractional frequency shifts normalized to total atom density n for two recent experiments [18,19]. We find good agreement with the three measured shifts.

For various applications, there is widespread interest for predictions of magnetic-field values at which Feshbach

TABLE II. Predictions of collisional frequency shifts for the ^{87}Rb fountain clock, compared to two recent experiments.

$(\frac{1}{n} \frac{\Delta\nu}{\nu})_{\text{exp}}$ (10^{-24} cm 3)	Ref.	Present theory (10^{-24} cm 3)
-56_{-21}^{+84}	[18]	-72.5 ± 3.3
$-50(10)_{-34}^{+22}$	[19]	-32.8 ± 0.7
$-60(16)_{-46}^{+29}$	[19]	-41.5 ± 2.9

resonances are to be expected in the scattering of two ^{87}Rb atoms in the $(f, m_f) = (1, +1)$ state. With our interaction parameters, we expect them at the four resonance field values B_0 given in Table III together with the widths Δ . The B_0 values are to be compared with the values 383, 643, 850, and 1018 G predicted in 1997 [4]. It is interesting that the broadest resonance at 1004 G shows a doublet structure [9].

Figure 1 shows Feshbach resonances that we predict to occur in the mixed-spin channels $(2, +1) + (1, -1)$ and $(2, -1) + (1, +1)$ at easily accessible field values of 1.9 and 9.1 G, respectively. The graphs show the predicted field-dependent scattering lengths $a(B)$, which are complex functions due to the presence of exothermal inelastic decay channels. The generalized analytic expression for the field dependence in this case is [9]

$$a(B) = a_\infty \left(1 - e^{2i\phi_R} \frac{\Delta_{\text{el}}}{B - B_0 + \frac{1}{2}i\Delta_{\text{inel}}} \right), \quad (1)$$

with Δ_{el} and Δ_{inel} the (in)elastic resonance widths and ϕ_R a resonance phase constant, arising due to inelasticity. Note that the real part of the scattering length does not go through infinity. It turns out that the 1.9 G resonance is an $l = 2$ resonance, which couples via the spin-spin interaction V_{ss} to the s -wave incident channel. Actually, this resonance is the ‘‘hyperfine analog state’’ [9] in the $(2, +1) + (1, -1)$ scattering channel of the d -wave shape resonance of ^{87}Rb occurring in the spin-stretched $(2, +2) + (2, +2)$ spin channel [13], i.e., a state with essentially the same spatial dependence and differing only in its hyperfine spin structure [20]. It is located at a comparable low energy above threshold. In a similar way, the $l = 0$ resonance at 9.1 G is the hyperfine analog state of two of the $l = 0$ bound states observed [5] at roughly 25 MHz below threshold in the $(2, +2) + (2, +2)$ and $(1, -1) + (1, -1)$ channels, belonging to the same rotational band as the d -wave shape resonance. They might play a role in the damping of coherent spin oscillations of the type which are being observed in experiments at JILA [21].

Until now we assumed the expression for V_{disp} to be valid for interatomic distances larger than r_S . We now ex-

TABLE III. Resonance fields B_0 and widths Δ for ^{87}Rb .

B_0 (G)	403(2)	680(2)	899(4)	1004(3)
Δ (mG)	<1	15	<5	216

tend V_{disp} with C_{11} and C_{12} terms and assume it to be valid also between $18a_0$ and r_S . This leads us to a more ambitious approach that allows us to determine C_{10} and C_{11} as two more free parameters in the least-squares search: We take into account the additional constraint arising from the equality $V_S(r) = V_{\text{disp}}(r) - V_{\text{exch}}(r)$ by imposing this equality at five r points as additional ‘‘experimental data’’ with a standard deviation of 0.5%. We thus effectively include the bound states of Ref. [7] with outer turning points in the range considered. In the search we take C_{12} equal to the theoretical value 11.9×10^9 of Ref. [22]. In column B of Table I, the resulting optimal parameter values are given together with error bars based on a 25% uncertainty in C_{12} . We find a value for C_{10} differing from the theoretical value 7.665×10^7 a.u. [14] by only 1.8%. While the above attractive C_n terms with even n arise from the interatomic multipole-multipole interaction in second order, a C_{11} term is expected [23] as a repulsive third-order dispersion term arising from the mutual dipole excitation and deexcitation of the atoms with an intermediate quadrupole transition between excited states in each of the atoms. Note that the ratio $C_{11}/C_{12} = -0.072$ is comparable to the rigorous value -0.028 for H atoms [24] and the *ab initio* ratio -0.041 for Cs atoms [25]. The remaining residue of the fit, concentrated at the smallest radii in the radial interval, may well be due to the summed contributions of further (attractive and repulsive) dispersion terms beyond the C_{12} contribution plus correction terms to the Smirnov-Chibisov exchange expression. Note that the values of the lower dispersion coefficients are dominated by

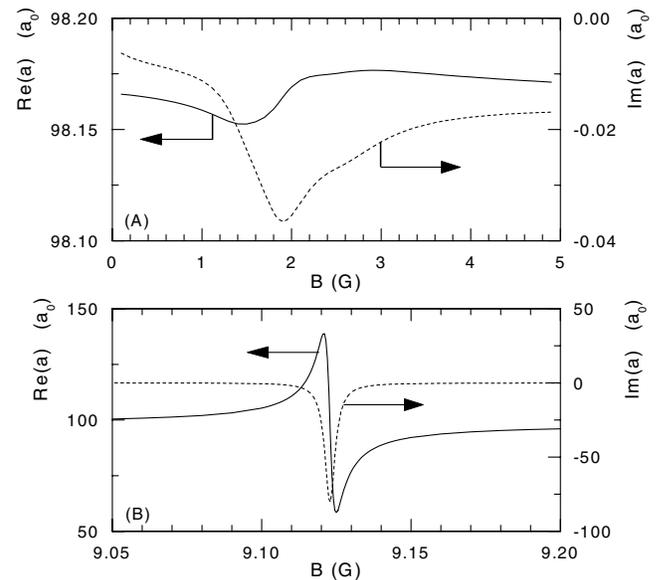


FIG. 1. (A) Real (solid line) and imaginary (dashed line) parts of the scattering length $a(B)$ in the s -wave ^{87}Rb $(2, +1) + (1, -1)$ mixed spin scattering channel, showing the presence of a Feshbach resonance at 1.9 G. The imaginary part is proportional to the summed rate coefficient G for decay into all open channels. (B) Same for Feshbach resonance in the $(2, -1) + (1, +1)$ channel at 9.1 G.

the close-to-threshold measurements, whereas the higher ones are determined primarily by the Seto potential in the middle range $r_0 < r < r_S$. We expect that experiment will prove the value of this more ambitious approach.

For completeness, we point out that a weak contribution to the total atom-atom force is still missing in the above picture: the interatomic spin-spin interaction V_{ss} . One component of V_{ss} is the well-known magnetic dipole interaction between the valence electron spins of the interacting atoms. An additional contribution, which arises from the electronic spin-orbit coupling as a second-order effect, has been experimentally determined for the first time for rubidium atoms by Freeland *et al.* [5]. Calculation shows that V_{ss} has a negligible influence on the previous analysis.

In summary, combining the results of three recent high-precision experiments, we have come close to a complete and model independent specification of the interaction properties of cold rubidium atoms. We have determined the van der Waals coefficients C_6 , C_8 , C_{10} , and C_{11} , and the strength J of the exchange interaction. We have thus reached a consistent picture of the interactions, with which it is possible to predict essentially all parameters needed for a complete description of a rubidium Bose-Einstein condensate or thermal gas of any isotope in an arbitrary spin state. New experimental data, in particular on the Feshbach resonances, will undoubtedly be helpful to confirm the above consistent picture and to further narrow down the error limits. We believe that our approach sets an example for similar experimental and theoretical work for other (combinations of) atomic species. From a theoretical point of view, it is fascinating that it is possible to achieve a level of precision for the interaction properties approaching that for collisions of cold hydrogen atoms, based on a combination of experimental results and a sound framework of collision physics. Additional details and their relevance for future experiments will be the subject of a future publication [9].

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