Collisions of Doubly Spin-Polarized, Ultracold ^{85}Rb Atoms

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We study the collisions of doubly spin-polarized ^{85}Rb atoms at millikelvin temperatures using photoassociation spectroscopy. Because the atoms are spin polarized, only triplet collisional states are formed. This leads to photoassociation spectra of a particularly simple form, which provide a very direct probe of the ground state collision. These spectra are analyzed to yield the ground state triplet scattering length $-1000a_0 < a_T < -60a_0$ for ^{85}Rb, $+85a_0 < a_T < +140a_0$ for ^{87}Rb, and the product of the D-line dipole matrix elements $d(P_{1/2})d(P_{3/2}) = 8.75 \pm 0.25$ a.u.

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Rapidly developing techniques for trapping and cooling neutral atoms using laser fields are opening up a wide array of new applications. These include the construction of very precise atomic clocks [1–3], sensitive electric dipole moment searches [4], and possible studies of quantum collective phenomena such as Bose-Einstein condensation. Success in each of these applications hinges critically on understanding the long-range interactions between cold atoms. Cold collision cross sections are very sensitive to long-range atomic interactions and play a dominant role in many experiments. For example, collisional frequency shifts may limit the accuracy of cold atomic fountain clocks [2,3]. Moreover, efforts to achieve Bose-Einstein condensation in a dilute laser-cooled gas depend critically on the ground state scattering length, which must be positive and preferably large [5–8].

Despite their importance, long-range interactions between atoms have been determined by conventional molecular spectroscopy in only a limited number of cases. The lack of extensive data is due in part to the difficulty of populating long-range states starting from the molecular ground state. On the other hand, these states are readily populated in collisions between ultracold atoms, in particular, by photoassociation spectroscopy [9]. Photoassociation spectra of Na [10,11], Rb [12,13], and Li [14] have already been obtained directly yielding detailed information on their long-range excited state interactions. In this paper, we present new ^{85}Rb photoassociation data and analysis, and show for the first time that atomic ground state interaction parameters can be determined from photoassociation spectra.

A crucial aspect of our experiment is that we doubly spin polarize the colliding atoms. This is important, since the ground state collision is thereby restricted to the triplet channel, and the analysis becomes relatively straightforward. An experiment using unpolarized atoms would need to determine both singlet and triplet parameters simultaneously. As a result of the spin polarization, we are able to observe clear, quantum-statistical features of the collisions. By doubly polarizing the atoms, and choosing a suitable excited state, we obtain spectra which provide a very simple and direct probe of the ground state collision: Each peak effectively measures the amplitude of a particular partial wave of the collision, thus yielding detailed information about the ground state interatomic potential in a narrow radial range that may be varied by the choice of excited state energy. This is, to our knowledge, the first report of doubly spin-polarized ultracold collisions of alkali atoms.

In our experiment, we load approximately 10^4 ^{85}Rb atoms into a far off-resonance optical dipole force trap (FORT) [15]. The FORT laser beam is a linearly polarized, Gaussian beam containing about 1.5 W of optical power and focused to a waist of about 10 μm. The FORT laser is tuned to 12 289 cm^{-1}, which lies between two well-resolved photoassociation peaks. To define a quantization axis, a magnetic field of 7 G is applied along the FORT laser beam propagation ($\hat{z}$) direction.

Once the atoms are loaded into the FORT, they are exposed to a combination of laser fields for 200 ms. Each 200 ms period is broken into a series of 5 μs cycles in which the atoms are irradiated by four laser fields in sequence. During the first 2.5 μs of each cycle, only the FORT laser is applied. During the next 0.6 μs, only two optical pumping (OP) beams interact with the atoms. One of these is tuned to the ^{85}Rb $5^2S_{1/2}(F = 3) \rightarrow 5^2P_{3/2}(F = 3)$ transition and is circularly polarized. It has an intensity of 100 μW/cm^2 and propagates along the z direction. The other optical pumping beam is tuned to the ^{85}Rb $5^2S_{1/2}(F = 2) \rightarrow 5^2P_{3/2}(F = 3)$ transition. During the last 1.9 μs of each cycle, only the photoassociation (PA) laser field is applied, which is linearly polarized, propagates in the z direction, and has an intensity in the range from 20 to 80 W/cm^2. The combined effect of these fields is to trap the atoms, to keep them optically pumped into the ^{85}Rb $5^2S_{1/2}(F = 3, M_F = 3)$ state, and to induce photoassociation transitions. Alternation of the fields in time prevents the light shift of the FORT beam from disrupting the optical pumping or from shifting and broadening the photoassociation resonances. At least 95% of the atoms are in the doubly spin-polarized state.

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Photoassociation transitions induced by the PA laser promote colliding pairs of Rb atoms into specific excited bound Rb_2^* states. In order to obtain a spectrum of these states, we repeat the loading and 200 ms irradiation period for a succession of PA laser frequencies [12,13]. At the end of each cycle we detect the number of atoms remaining in the trap with laser-induced fluorescence. Because the excited Rb_2^* states decay predominantly to free pairs of atoms with kinetic energy sufficient to leave the trap, the photoassociation resonances are detectable as a reduction in the fluorescence.

A photoassociation spectrum of a single 0_g^- vibrational level at 12 573.05 cm^{-1} is shown in Fig. 1. In Fig. 1(a), we show the spectrum observed when the atoms are maintained in the $5^2S_{1/2}$, $F = 3$ level but are otherwise unpolarized. A pure rotational spectrum spanning the range from $J = 0$ to 4 is observed. In Fig. 1(b), we show a spectrum recorded with doubly spin-polarized atoms. The odd rotational lines disappear as a consequence of spin statistics.

In order to realize a determination of the $^{85}Rb_2$ ground state parameters, we recorded data similar to that shown in Fig. 1 for a series of vibrational levels of the 0_g^- state that asymptotically connects to the $5^2S_{1/2} + 5^2P_{1/2}$ separated atom limit [16,17]. Five vibrational levels were used in the analysis, with $J = 0$ level energies of -3.365, -4.088, -4.901, -5.812, and -6.827 cm^{-1} with respect to the barycenter of the $5^2S_{1/2} + 5^2P_{1/2}$ dissociation limit, which corresponds to an energy of 12 578.864 cm^{-1} in our spectrum. The outer turning points of these states range from $41.6a_0$ to $46.7a_0$.

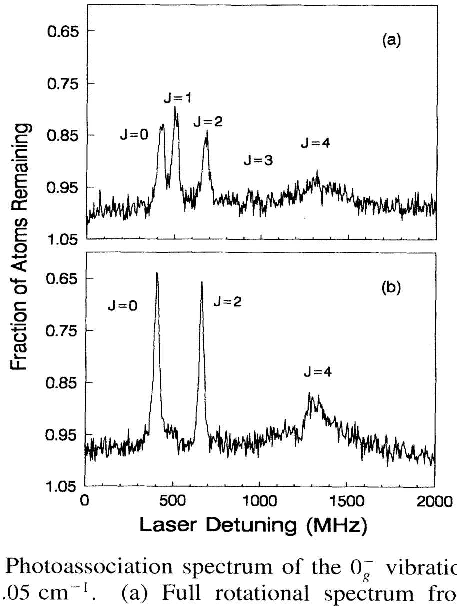

FIG. 1. Photoassociation spectrum of the 0_g^- vibrational level at 12 573.05 cm^{-1}. (a) Full rotational spectrum from $J = 0$ to 4 observed for atoms in $5^2S_{1/2}$, $F = 3$ level but otherwise unpolarized. (b) Spectrum recorded with doubly spin-polarized atoms. Odd rotational lines disappear as a consequence of spin statistics.

The photoassociation spectrum is conveniently described in the dressed-state picture. In that framework each of the Rb + Rb* rovibrational states $|\Omega JM\rangle$ is a discrete state embedded in the ground state continuum [18]. As a result of the interaction with the PA laser field, it acquires a finite partial width γ_L for decay into each of the ground state channels in addition to its spontaneous linewidth γ_0. γ_L is proportional to the PA laser intensity I_L and is given by Fermi's golden rule,

$$\gamma_L = 2\pi |\langle \Omega JM | [\mathbf{d}(1) + \mathbf{d}(2)] \cdot \mathbf{E}_L | SM_S l m_l, \epsilon \rangle|^2, \tag{1}$$

where $\mathbf{E}_L = E_L \boldsymbol{\sigma}_L$ is the PA laser field and $|SM_S l m_l, \epsilon\rangle$ is the energy-normalized continuum ground state. Accordingly, the squared S-matrix element for photoassociation followed by spontaneous emission is given by a Breit-Wigner expression. To first order in I_L

$$|S_{\Omega JM, SM_S l m_l}|^2 = \frac{\gamma_0 \gamma_L(\Omega JM \rightarrow SM_S l m_l)}{(\epsilon + E_g + \hbar\omega_L - E_e)^2 + \frac{1}{4}\gamma_0^2}. \tag{2}$$

Here E_g is the asymptotic Rb + Rb internal energy, E_e is the energy of the $|\Omega JM\rangle$ state, and ϵ is the collision energy. Equation (2) is thermally averaged and summed over J, M, l, m_l to obtain the rate coefficient. The above expressions, without the directional dependences, were first used to analyze photoassociation spectra by Napolitano *et al.* [19]. Expanding $|\Omega JM\rangle$ in atomic fine-structure states [16,17] coupled to total electronic angular momentum jm_j,

$$\gamma_L = \frac{2\pi I_L}{\epsilon_0 c} \left| \sum_j (J\Omega\, j - \Omega | l0)(jm_j l m_l | JM) \times \int_0^\infty dr\, c_j(r) u_{\Omega J}(r) \times \langle jm_j | [\mathbf{d}(1) + \mathbf{d}(2)] \cdot \boldsymbol{\sigma}_L | SM_S \rangle u_l(r) \right|^2. \tag{3}$$

The above-mentioned $0_g^-(S + P_{1/2})$ electronic state has a number of simplifying features that facilitate the analysis considerably. First, it is a pure triplet state [16], so that no singlet amplitude is coupled in by the excitation. Second, as with any of the $\Omega = 0$ states, it has negligible second-order hyperfine energy shifts. Third, at the relevant interatomic distances near the outer turning point, this 0_g^- state is, to very good approximation, a product of independent atomic states $S_{1/2}$ and $P_{1/2}$, coupled to form $j = 0$. As a consequence, only states for which $J = l =$ even are excited. Thus our data display directly the quantum statistics of the atoms: Because they are bosons, they may only collide in even ground state partial waves. Note that the magnetic field has negligible influence on the spectrum.

We find γ_L for the 0_g^- state to be a product of a geometrical coefficient and a squared radial integral $\int dr\, u_{\Omega J}(r)\, d_{eg}(r)\, u_l(r)$, in abbreviated notation. Notice that a $j = 0$ component of the upper state cannot be excited by σ^+ laser light starting with doubly polarized ground state atoms with $S = M_S = +1$. Indeed, a full calculation of the excitation rate, taking into account all components of the 0_g^- state, shows this photoassociation rate to be smaller than that for σ^- light by 2 orders of magnitude. This prediction is confirmed by experiment.

Because of the small ranges of ϵ and l involved, cold collisions have the unique property of being insensitive to the detailed behavior of the badly known inner parts of the interatomic potential. The variation of the radial wave function with E and l is a very small first-order perturbation up to a rather large radius r_0. The only relevant information is the accumulated information contained in the phase $\phi(E, l)$ of the rapidly oscillating wave function at r_0 and its first derivatives for $E = l = 0$ [20]. Model calculations show that for $l \leq 2$ and $r_0 = 30a_0$, the calculated photoassociation rates are sufficiently insensitive to the precise values of the first derivatives that they can be taken reliably from an *ab initio* calculation [21]. This insensitivity was used and explained previously in Refs. [22,23]. A similar accumulated-phase method was adopted for the excited state. At large distances where excitation of the above-mentioned five vibrational levels occur, our calculated results are almost independent of the dispersion parameters C_{ne} ($n \geq 6$) for the excited state and C_{ng} ($n \geq 8$) for the ground state, provided that these are taken within the bounds of the present uncertainty [17,24–26].

From the energies of the five measured $J = 0$ levels we derive a value 8.75 ± 0.25 a.u. for the product $d(P_{1/2})d(P_{3/2})$ of D-line dipole matrix elements [27], which determines the strength of the resonant-dipole $1/r^3$ potential in the present r range. This value is in agreement with the most accurate previous measurements $d^2(P_{1/2}) = 8.43 \pm 0.20$ a.u. [28], $d^2(P_{3/2}) = 9.19 \pm 0.18$ a.u. [28], $d^2(P_{3/2}) = 9.08 \pm 0.28$ a.u. [29], and $d^2(P_{3/2}) = 8.68 \pm 0.16$ a.u. [30].

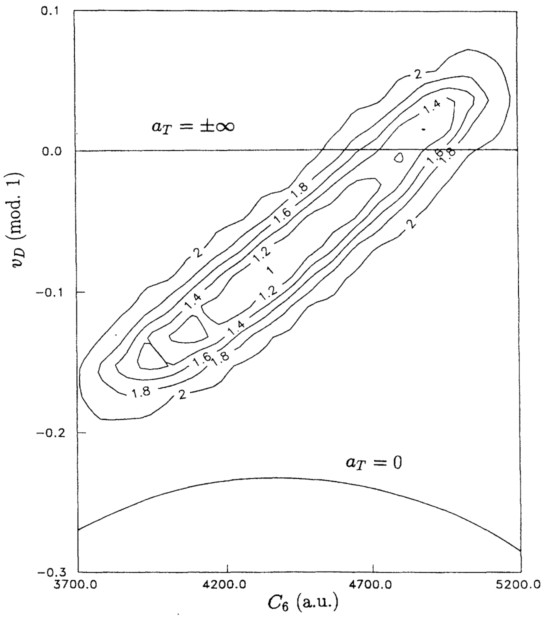

FIG. 2. Contour plot of total χ^2 surface for ratios of $J = 0$ and 2 photoassociation peak areas, as a function of C_{6g} and s-wave vibrational quantum number v_D at dissociation. Lines where a_T changes sign are indicated.

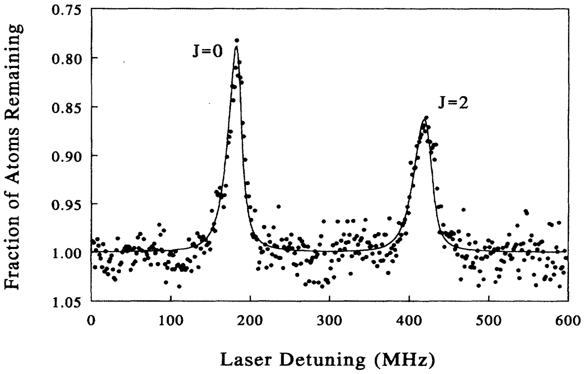

FIG. 3. Comparison of theoretical and experimental line shapes for rotational levels at 12 573.96 cm^{-1}. Each J peak arises only from the partial wave $l = J$.

The analysis of the data is carried out as follows. As a first step, we use the strongest $J = 0$ and 2 rotational line shapes to determine an optimum temperature for each point of a grid of C_{6g}-ϕ_g values by a least-squares fit to the measured data points. For this purpose, C_{6g} is taken to lie between 3500 and 6000 a.u. and ϕ_g is allowed to span a full π range. Using these temperatures, we apply a least-squares fit to the ratios of the $J = 0$ peak areas over the C_{6g} and ϕ_g plane (least-squares function χ_1^2), which constrains these parameters to lie in a narrow strip and determines the temperature to be 500 ± 100 μK. We then calculate the χ_2^2 and χ_3^2 functions associated with the four $J = 2$ and one $J = 2$ to $J = 0$ ratios of peak areas, respectively, over the limited ranges of C_{6g} and ϕ_g found previously. We find that χ_2^2 sets about the same limits on C_{6g} and ϕ_g as χ_1^2. However, χ_3^2 sets a limit that corresponds to a different strip in the C_{6g}-ϕ_g plane. The intersection of these strips gives the estimated values and uncertainties of C_{6g} and ϕ_g. Figure 2 shows a contour plot of the total χ^2 surface combining the above three sets of ratios of peak areas. Instead of ϕ_g, we use the more transparent (fractional) s-wave vibrational quantum number v_D at dissociation as a parameter. The lines where the triplet scattering length changes a_T sign are indicated. While C_{6g} is experimentally constrained to about the full range of theoretically predicted values [17,24,31], v_D(mod1) is found to be in the interval between $+0.07$ and -0.19. Including the uncertainty in d^2, C_{6e}, and C_{8g}, we find a_T to be negative with at least 80% probability. For the recently predicted value $C_{6g} = 4426$ a.u. from Ref. [24], which is believed to be correct within a few percent [26], we find $v_D(^{85}\text{Rb, mod1}) = -0.09 \pm 0.07$ and $-1000a_0 < a_T(^{85}\text{Rb}) < -60a_0$. Figure 3 shows a comparison of theoretical and experimental line shapes of the $J = 0$ and 2 rotational levels at 12 573.96 cm^{-1} for this value of C_{6g} and $v_D(\mathrm{modl}) = -0.09$.

A simple $\sqrt{m}$ scaling procedure enables us to find the corresponding results for the ^{87}Rb isotope. On the basis of the triplet potential of Krauss and Stevens [21] we take the number of s-wave radial nodes within r_0 to be 34 ± 3. We then derive ϕ_g for ^{87}Rb and by radial integration to larger distances, introducing five additional nodes, find $v_D(^{87}\mathrm{Rb}, \mathrm{modl}) = +0.37 \pm 0.10$ and $+85a_0 < a_T(^{87}\mathrm{Rb}) < +140a_0$. Also for the total theoretical C_{6g} range a_T is found to be positive.

To summarize, we have used high-resolution photoassociation spectroscopy to study the collisions of ultracold ^{85}Rb atoms. By doubly spin polarizing the atoms, and choosing the particular excited state 0_g^- ($5^2S_{1/2} + 5^2P_{1/2}$), we obtain spectra which provide a very simple and direct probe of the ground state collision: Each peak effectively measures the amplitude of a particular partial wave of the ground state collision, in a narrow radial range that may be varied by the choice of excited state energy. Because of this, we obtain detailed information about the ground state interatomic potential. Analysis of these spectra reveals that the ground state triplet scattering length of ^{85}Rb is large and negative, so that no stable Bose condensate is possible for this state of this isotope. Using mass scaling arguments, we are led to the opposite conclusion for the ^{87}Rb isotope. Apparently, its triplet scattering length is positive so that it remains a viable candidate for observation of a stable Bose-Einstein condensate.

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