Negative Scattering Length of Ultracold ^{7}Li Gas

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We have improved the theoretical triplet ground-state potential of $^7Li_2$ by means of an inverse perturbation analysis, and find that the diatomic scattering length for ^{7}Li atoms in the $F = 2$, $m_F = 2$ ground-state hyperfine level is negative. The consequences of this result for achieving Bose-Einstein condensation in a trapped ultracold ^{7}Li gas sample are addressed.

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The ideal degenerate quantum gas, consisting of indistinguishable particles with de Broglie wavelength $\Lambda > n^{-1/3}$, where n is the density of the gas, is a fundamental paradigm of quantum statistical mechanics. Yet there are no observations of even a weakly interacting gas in the degenerate quantum regime. In a weakly interacting gas, the interactions are due to binary collisions; multiple-body collisions occur so infrequently that their effect can be neglected. Unlike quantum liquids, the properties of a weakly interacting quantum gas may be calculated using perturbation theory [1]. The realization of a weakly interacting degenerate quantum gas may provide the opportunity to observe and study the most spectacular predictions of quantum statistical mechanics, Bose-Einstein condensation (BEC), and gaseous superfluidity, in a well-characterized, simple system.

The most attractive candidate to observe these phenomena is a spin-polarized gas of atomic hydrogen atoms, which as composite particles obey Bose-Einstein statistics [2]. Spin-polarized ground-state H interacts via the largely repulsive triplet potential. The attractive van der Waals interaction between atoms produces a 6 K deep well at intermediate range, but this well is too shallow to support bound states. Spin-polarized atomic hydrogen gas is predicted to remain gaseous even for temperatures approaching zero. The most promising strategy for realizing BEC in atomic hydrogen now appears to be (light-induced [3]) evaporative cooling of a spin-polarized H gas sample in a magnetic trap with a three-dimensional magnetic field minimum [4]. However, despite intense effort the necessary low temperature and high density required to observe BEC have not yet been attained. The most recently published results are $n \approx 10^{14}$ cm^{-3} and $T \approx 100$ μK [5], which is about a factor of 3 above the condensation temperature at this density.

Magnetically trapped spin-polarized alkali atoms might also be used to investigate weakly interacting quantum gases. There has been much progress in the past few years in trapping alkali atoms and manipulating them using laser radiation [6]. In particular, alkali atoms have been cooled to near the "recoil limit" $T_R = (\hbar k)^2/mk_B$, where k is the wave number of the near-resonant laser radiation and m is the mass of the atom. T_R ranges from 0.2 μK for Cs to 6 μK for Li atoms laser cooled on their principal optical transitions. Laser radiation at the corresponding wavelengths for these transitions is easily generated since they are in the visible or near-infrared portion of the spectrum. Li is an attractive candidate for these experiments. There are two stable isotopes of Li : ^{7}Li obeys Bose-Einstein statistics, while ^{6}Li obeys Fermi-Dirac statistics. Therefore, it is possible to compare both quantum statistics with a single experimental setup. Li has clear advantages from the theoretical point of view relative to the heavier alkalis. Because of the small number of electrons the diatomic interaction potentials of the Li_2 system can be calculated more accurately. In particular, for the work described below, there is close agreement between different calculations of long range dispersion and exchange potentials which play a decisive role in ultracold collisions. In this respect, Li + Li is an interesting intermediate system between H + H, for which it has been possible to calculate the ground-state potentials rigorously [7] and the heavier alkali diatoms, for which the uncertainties are far larger. Furthermore, the $S = 0$ and $S = 1$ potentials are much less attractive than for the heavier alkalis, which combined with the low atomic mass leads to a lower number of widely spaced bound levels close to the continuum. As a consequence, it is far easier to derive a value for the scattering length from spectroscopic data alone than, for instance, for Cs where very small changes in the potential induce sizable differences in the scattering length and even in the number of bound levels, although much progress has been made to derive the scattering length from measured frequency shifts in a cesium atomic fountain [8]. In addition, the much weaker atomic $\mathbf{S} \cdot \mathbf{I}$ hyperfine interaction makes it possible to devise simple models for the competition of exchange forces and hyperfine coupling in ultracold atom-atom scattering [9]. Finally, the largely unknown role of the spin-spin interaction [10,11] (which makes the dipolar decay rate of the density of a heavier alkali gas in a magnetic trap so uncertain) is expected to be virtually absent in Li + Li, again because of the low Z value.

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Although alkali atoms are attractive for these investigations, spin-polarized alkali atoms are not as weakly interacting as spin-polarized hydrogen. For example, the depth of the van der Waals well in the triplet potential of Li is 480 K and is predicted to support 10 bound vibrational energy levels for ^{6}Li and 11 for ^{7}Li [12]. Many more bound levels will be supported by the triplet potential of the heavier alkali atoms. On a time scale during which three-body collisions can be neglected, however, these bound levels are not formed and a description of the properties of the gas in terms of a single quantity, the two-body s-wave scattering length $a = -\lim_{k\to 0} \tan\delta_0(k)/k$, is appropriate at ultracold temperatures ($k \mid a \mid \ll 1$). The sign of a then turns out to be of crucial significance. $a > 0$ represents an effectively repulsive potential, and collisions can be modeled as those between hard spheres of radius a. The thermodynamic properties of such a gas can be obtained as a power series expansion in the parameter $(na^3)^{1/2}$ [1]. For negative values of the scattering length the interaction is effectively attractive and the Bose-condensed state is predicted to be unstable in a spatially homogeneous system [13]. Also other features are lost which are characteristic of BEC: (1) The usual discussion of the Bose condensate in terms of a Bogoliubov transformation [13] crucially depends on a positive value of the scattering length [14]; (2) closely connected with this, the quasiparticle concept breaks down; (3) the quartic term for the free energy as a function of the order parameter $\langle\psi\rangle$ is negative, precluding the application of the Landau theory of second-order phase transitions. It is not clear at present how this situation changes in the inhomogeneous situation of a trap experiment. It is possible in principle that a fractionally significant occupation of a single-particle ground state occurs in such an experiment as a transient phenomenon. The zero point energy may help stabilize the system against collapse for some time. A treatment of the inhomogeneous time-dependent Bose system with $a < 0$ does not exist in the literature. It is clear, however, on the basis of the above-mentioned aspects of the homogeneous system that a negative sign of a will have profound influence on a possible transient BEC type state occurring in such an experiment. For instance, the Hartree-Fock equation for the condensate wave function, known as the Gross-Pitaevskii equation [15], does not have a solution. At the present time, the many fascinating questions associated with the dynamics of an inhomogeneous Bose gas with a negative scattering length remain unexplored theoretically as well as experimentally. This underlines the importance of determining the sign of a for the various alkali gases for which BEC is attempted experimentally.

In this paper we present results of calculations which show that the scattering length for the scattering of two Li ground-state atoms with $F = 2$, $m_F = 2$ is negative. Apart from the sign of the scattering length, knowledge of the Li + Li interaction potentials is crucial for predicting the rate of decay of a trapped Li gas sample due to dipolar relaxation and three-body recombination. Work along this line is planned as a continuation of this project.

The starting point of the present work was the ^{6}Li triplet ground-state potential obtained by Zemke and Stwalley [12] via a "dense" Rydberg-Klein-Rees analysis of spectroscopic data from Ref. [16] and the addition of short- and long-range analytical parts. Using this potential for ^{7}Li we calculate a scattering length $-16a_0 < a < -10a_0$ depending on the values of the dispersion and exchange parameters. When solving the Schrödinger equation for the $^7Li_2$ triplet levels, however, we find discrepancies ΔE_{vJ} with the spectroscopic term values which exceed the experimental uncertainties. In view of this, we have improved the potential by means of an inverse perturbation analysis (IPA) [17]: a correction ΔV is obtained in the form of a linear combination $\sum_i c_i f_i(r)$ of suitably chosen basis functions. Equating the first-order energy perturbations to the desired energy shifts produces a set of linear equations for the coefficients c_i, which allows ΔV to be calculated. This procedure is repeated until convergence, using the new "unperturbed" eigenfunctions in every iteration. The experimental data set includes the $v \leq 7$ levels from Ref. [16]. It is now being extended and improved by new measurements. Preliminary results [18] support the quality of the IPA potential derived here. In Fig. 1 we present the discrepancies $\Delta E_{vJ} = (E_{vJ} - E_{00})^{\text{calc}} - (E_{vJ} - E_{00})^{\text{exp}}$ for all bound $v \leq 7$ levels before and after IPA.

We consider the potential thus obtained to be sufficiently accurate to determine the scattering length in the following way. We define a phase difference $\phi(E, J)$ between radial solutions of the Schrödinger equation at energy $E < 0$ and angular momentum J found by outward and inward integration starting with regular boundary conditions. We consider only high levels for which this

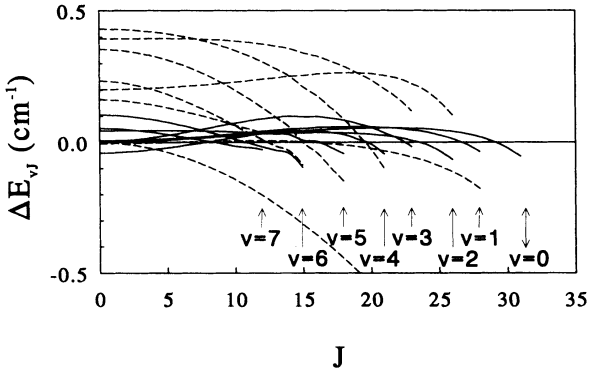

FIG. 1. Differences of experimental and theoretical energy eigenvalues $[\Delta E_{vJ} = (E_{vJ} - E_{00})^{\text{calc}} - (E_{vJ} - E_{00})^{\text{exp}}]$ before (dashed lines) and after IPA (solid lines) for rovibrational levels with $v \leq 7$. The values of v corresponding to the lines can be recognized by their maximum J values, which are indicated by the arrows.

phase has an unambiguous value in the classically allowed region. We define ϕ as a continuous function of energy, equal to $v\pi$ for E equal to an eigenvalue E_{vJ}. Calculations for various choices of $V(r)$ indicate that the differences of ϕ with E and J over the relevant E and J ranges are negligibly changed by reasonable changes in V. This is clear intuitively, since differentiation of the phase $\phi \simeq \int k(r)dr$ with respect to E or $J(J+1)$ tends to emphasize the dependence on the more reliable long-range potential. We therefore assume that our IPA potential describes sufficiently accurately the variations of $\phi(E, J)$ with E and J and determine the change $\Delta\phi$ in $\phi(0,0)$ needed to fit the differences of the higher measured levels. Figure 2 shows the discrepancies between measured and calculated energy differences for the $v = 6, 7, 8$ and 9 levels as a function of $\Delta\phi$ or equivalently v_D. The diagram also shows the value of the scattering length, which can be calculated for each value of $\phi(0,0)$. The $\Delta\phi$ values for the various energy splittings do indeed differ by small parts (3% at most) of the period π. Furthermore, very small changes of $\phi(0,0)$ are apparently sufficient to fit the experimental values. We find $-0.04 < \Delta\phi < +0.035$ or $10.79 < v_D < 10.81$. For the scattering length we find a negative value: $-19.9a_0 < a < -12.2a_0$. Varying the dispersion and exchange coefficients over their error bars [12] enlarges this interval to $-27.8a_0 < a < -7.1a_0$.

As in the case of Cs [11,19] the upper level of the lower hyperfine manifold is another possible candidate for BEC (^{7}Li: $F = 1$, $m_F = -1$). This state is also a low-field seeking state for magnetic fields less than 140 G, but at higher fields the sign of its Zeeman shift reverses due to hyperfine mixing. This imposes a rather low maximum trap depth of only 1 mK. The existence of this alternative in the case of the alkalis is of great importance, since it may be essential for avoiding the "relaxation explosion" upon crossing the BEC phase transition line which has been predicted [20] for the case of magnetically trapped H: The magnetic dipole decay rate for the $F = 1$, $m_F = -1$ state goes to zero for $B \to 0$. It will be necessary to derive the ^{7}Li singlet potential in order to carry out the coupled-channel calculations necessary to derive the scattering length for the scattering of $F = 1$, $m_F = -1$ atoms. Like in the case of Cs [11] it is to be expected that resonances will show up for particular values of the external magnetic field [21]. The ability to change the sign of the scattering length by a variation of B in the environment of a suitable resonance is an intriguing possibility.

In conclusion, we have found convincing evidence for a negative sign of the scattering length for ^{7}Li + ^{7}Li triplet scattering and discussed its implications for the collective quantum behavior of an ultracold ^{7}Li gas sample in a magnetic trap.

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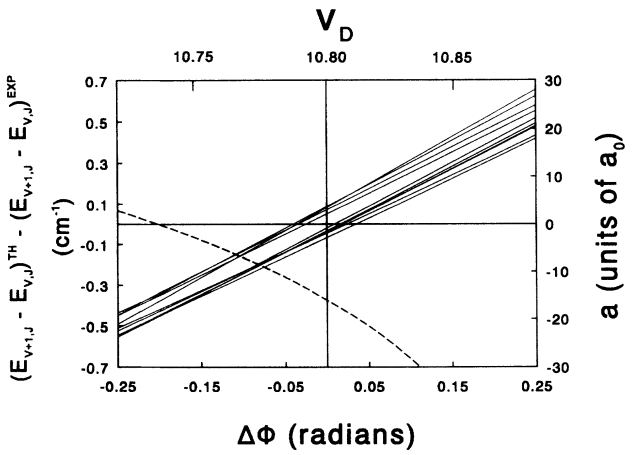

FIG. 2. Discrepancies between measured and calculated energy differences (left axis, solid lines) and scattering length (right axis, dashed line) as a function of $\Delta\phi$ or v_D. The $(v+1, J)$-(v, J) combinations for the lines from above to below at $\Delta\phi = 0$ are (8,9)-(7,9), (8,11)-(7,11), (8,6)-(7,6), (8,4)-(7,4), (7,11)-(6,11), (7,9)-(6,9), (7,6)-(6,6), (7,4)-(6,4), (9,4)-(8,4), and (9,6)-(8,6).

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