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Singlet s-wave scattering lengths of ^{6}Li and ^{7}Li

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Photoassociation of ultracold lithium atoms into bound vibrational levels of the $A\,^1\Sigma_u^+$ excited state is used to probe the $X\,^1\Sigma_g^+$ ground-state interaction potential of $^6\mathrm{Li}_2$ and $^7\mathrm{Li}_2$. It had been predicted that the s-wave photoassociation signal strength would pass through a minimum as a function of vibrational level for positive s-wave scattering length. We report the observation of this novel effect, and use the location of the minimum to precisely determine the singlet s-wave scattering length for both isotopes. The sensitivity of this technique is demonstrated by distinguishing the minima for collisions involving ^{7}Li atoms in different hyperfine states. [S1050-2947(96)50306-9]

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New developments in cooling atomic gases have enabled investigations of novel effects in the ultracold energy regime, including the recent attainment of quantum degenerate gases and observation of Bose-Einstein condensation in weakly interacting systems [1–3]. Collisional interactions are crucial to both the dynamical evolution and the steady-state behavior of these systems and are relevant to searches for superfluidity in an atomic gas [4], advances in high precision measurements [5], and the creation of more highly accurate atomic clocks based on laser-cooled neutral atoms [6]. An important parameter for understanding these collisions is the s-wave scattering length a. At low temperatures where the thermal de Broglie wavelength is large compared to the interaction length scale, the effect of the two-body interaction potential in elastic scattering can be described using the s-wave scattering length [7]. For atoms colliding with relative momentum $\hbar k$, the scattering length is related to the s-wave phase shift δ_0 by $a=-\lim_{k\to 0}\tan\delta_0/k$.

Two ground-state alkali-metal atoms can interact via the electronic spin-singlet $X\,^1\Sigma_g^+$ or the spin-triplet $a\,^3\Sigma_u^+$ molecular potentials. The scattering length is extremely sensitive to the interaction potential. For potentials supporting bound states, as is the case for all alkali-metal dimers, the sign and magnitude of the scattering length are largely determined by the location of the highest-lying bound state of the potential [8]. A potential giving a state that is only weakly bound leads to a large positive value for a corresponding to a net repulsion, but if the potential were slightly shallower so that this state were barely unbound, a would change discontinuously to a large and negative value [8]. For the $a\,^3\Sigma_u^+$ state, two-photon spectroscopy has been used to directly locate the highest lying vibrational level of $^7\mathrm{Li}_2$ and a_T was found to be $-27.3\pm0.8a_0$ [9]. For the straight state of ^{7}Li, Côté *et al.* and Moerdijk *et al.* have independently predicted a value, contained in the interval $32a_0<a_S<38a_0$, by comparing an empirical potential to spectroscopic data [10].

Photoassociation is the formation of a molecule by photoabsorption during a collision of free atoms [11]. While photoassociative spectroscopy is a powerful technique for probing long-range, high-lying bound states of diatomic molecules, it can also provide details of the low-energy, ground-state unbound wave function [12]. Napolitano *et al.* first suggested that since the scattering length is a property of the zero-energy wave function, scattering lengths could be determined from photoassociative spectra [13]. One-photon photoassociative spectra of Rb have been used to determine that $-1200a_0<a_T<-80a_0$ for ^{85}Rb, and $99a_0<a_T<119a_0$ for ^{87}Rb [14]. Côté *et al.* recently pointed out that the s-wave contribution to photoassociative spectra is extremely sensitive to the sign of a [15]. The long-range asymptotic wave function for the free atoms is $u\propto(R-a)$, so u has a node at $R=a$ when $a>0$. Since the amplitude of the excited-state wave function is greatest near its outer turning point, and the transition strength is a function of the overlap of the ground- and excited-state wave functions, a minimum in the photoassociation signal is expected for vibrational levels with outer turning points near $R\simeq a$ for the positive a case [15]. Based on the qualitative dependence on vibrational level observed in the photoassociation signal strengths of the $X\,^1\Sigma_g^+\rightarrow A\,^1\Sigma_u^+$ and $a\,^3\Sigma_u^+\rightarrow 1\,^3\Sigma_g^+$ transitions of $^7\mathrm{Li}_2$ [16], Côté *et al.* found $a_T<0$ and $a_S>0$ for ^{7}Li [15]. In the previous experimental work, the signal strengths were observed to monotonically decrease as more deeply bound levels were probed [16]. However, since $a_S>0$, a recurrence of the singlet signal strengths was predicted. In this paper, we report the observation of this recurrence. We present quantitative measurements of the one-photon photoassociation signal strengths of transitions to vibrational levels of the $A\,^1\Sigma_u^+$ state of both $^6\mathrm{Li}_2$ and $^7\mathrm{Li}_2$, and use this information to extract precise values of a_S for both isotopes.

The details of the apparatus used to trap lithium and perform photoassociative spectroscopy are described in previous publications [16,17]. A photoassociation laser passed through the cloud of trapped atoms causes the formation of excited-state molecules, which may radiatively decay into bound ground-state molecules or unbound atom pairs. Both decay channels lead to measurable trap loss when the photoassociation laser is tuned to a photoassociation resonance. The increase in loss rate reduces the steady-state number of

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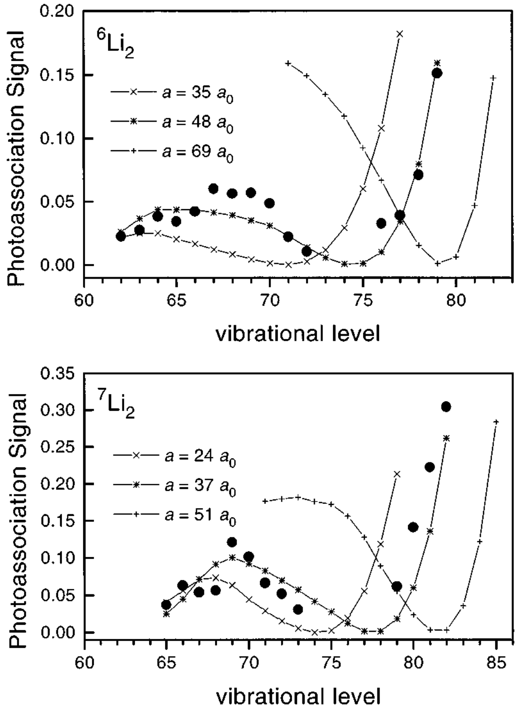

FIG. 1. The circles represent relative photoassociation signal strengths for transitions to vibrational levels of the $A\,^1\Sigma_u^+$ state of ^{6}Li$_2$ and ^{7}Li$_2$. For ^{7}Li$_2$, transitions involving collisions where both atoms are in the $f=2$ hyperfine state are shown, and the binding energies of the data vary from 410 GHz for $v=82$ to 4.4 THz for $v=65$ [16]. For ^{6}Li$_2$, the binding energies vary from 220 GHz for $v=79$ to 3.5 THz for $v=62$ [16], and the photoassociation strength is averaged over the three hyperfine features. The photoassociation signal is the fractional decrease in the observed fluorescence due to the presence of the photoassociation laser on a photoassociation resonance. Results of calculations of the signal strengths for three different values of the scattering length are shown for each isotope.

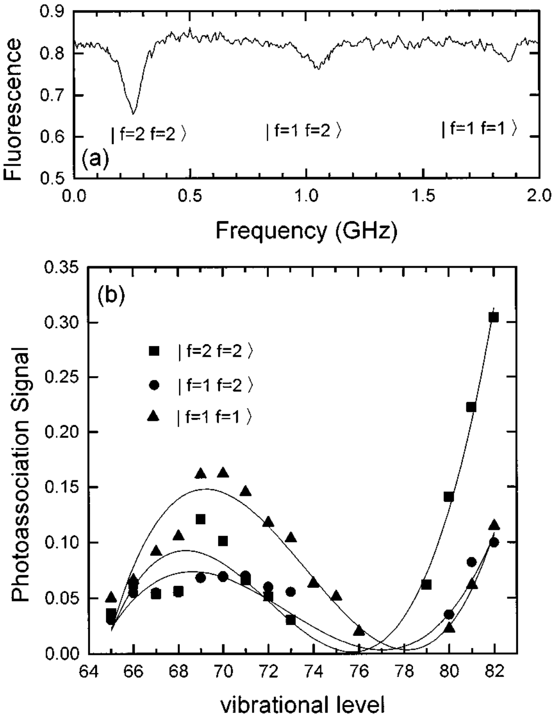

FIG. 2. (a) Spectrum of the $v=81$, $N=1$ level of the $A\,^1\Sigma_u^+$ state of ^{7}Li$_2$. Three features, separated by the atomic ground-state hyperfine splitting, are observed corresponding to collisions with both atoms having $f=2$, one atom with $f=1$ and the other with $f=2$, and both atoms having $f=1$. The fluorescence is given in arbitrary units. (b) Photoassociation signals for each hyperfine feature of each vibrational level of the $A\,^1\Sigma_u^+$ state of ^{7}Li$_2$. The lines are a polynomial fit to the data to help identify the node for each hyperfine series. The resulting minimum in the photoassociation signal occurs at a different location for the different collision channels.

atoms in the trap, which is detected by a photodiode monitoring the trap-laser-induced fluorescence of the trapped atoms. Since the loss-rate increase will be larger for a resonance with greater transition strength, the number of atoms in the trap is a relative measure of the strength of the photoassociative transition. While the signals saturate for large photoassociation signals, the analysis concerns the location of the photoassociation minimum where the signals are small and proportional to photoassociation strength.

The photoassociation signal strengths for transitions into vibrational levels of the $A\,^1\Sigma_u^+$ state of ^{6}Li$_2$ and ^{7}Li$_2$ are shown in Fig. 1. Only transitions to $N=1$ rotational levels are considered, since they correspond to s-wave collisions that dominate at a trap temperature of 1–2 mK. For both isotopes, the transition strength decreases as the vibrational quantum v is decreased, goes through a minimum, and then recurs at lower v, as predicted for $a>0$. Also shown in Fig. 1 are calculations of signal strengths for both isotopes assuming several values of a_S. The sensitivity of the signal strength to a, evident in Fig. 1, enables an extremely precise determination of their values.

Hyperfine structure is easily resolvable in the spectra of both isotopes [18] and plays a role in extracting the singlet scattering lengths. For large internuclear separation R in the initial free state, the value of the total angular momentum of each atom f is a good quantum number. For ^{7}Li, the nuclear spin is 3/2, so $f=1$ or 2. In collisions of two atoms there are three possible combinations of f values: both atoms can have $f=1$; one atom can have $f=1$ and the other $f=2$; or both can have $f=2$. Hyperfine structure arises in the singlet spectra because each of the three combinations has both singlet and triplet character, so a transition to an excited singlet state picks out the singlet component in the free state, giving three transition features separated by the ground-state hyperfine splitting of the atom, as in Fig. 2(a) [18]. At large R, the three combinations result in three separated interaction potentials. But at small R, the total electronic spin is a good quantum number because of the dominance of the exchange interaction relative to the hyperfine interaction [19].

In Fig. 2(b) the photoassociative signal for each hyperfine feature for ^{7}Li$_2$ is shown. The minimum of the photoassociative signal is different for the different collision possibilities, graphically demonstrating the sensitivity of signal strengths to very small differences in the potential. By comparing with the theoretical calculations shown in Fig. 1(b), we find the best match for $a=(39\pm3)a_0$ for collisions between two $f=1$ atoms, $(33\pm5)a_0$ for an $f=1$ and an $f=2$

atom, and $(31\pm3)a_0$ for collisions of two $f=2$ atoms. Although these do not represent the scattering lengths for any particular collision, the true singlet scattering length will be between these values and would be given by the location of the signal strength minimum in the limit of no hyperfine interaction. Therefore, we give $a_S=(34\pm5)a_0$, in good agreement with the results of Ref. [10]. The ground-state hyperfine splitting for ^{6}Li is significantly smaller than for ^{7}Li (228.2 MHz vs 803.5 MHz) and we cannot distinguish the different minima. We find $a_S=(47\pm3)a_0$ for ^{6}Li.

Single-photon photoassociative signal strengths are sensitive to both the sign and magnitude of the *s*-wave scattering lengths in the ultracold regime. This is particularly true for $a>0$ where the location of the minimum in signal strength precisely constrains a. The method is well suited to lithium where the long-range asymptotic region corresponds to the region of significant photoassociation signal strength.

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