Photoabsorption by Ultracold Atoms and the Scattering Length

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It is shown that photoabsorption in a gas of ultracold atoms depends on the sign and magnitude of the scattering length in the scattering of a pair of atoms. Measurements of fluorescence in ^{7}Li are interpreted to show that the triplet scattering length is negative and the singlet scattering length is positive.

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The s-wave scattering length is a critical parameter in theories of Bose-Einstein condensation. They predict that the Bose-Einstein condensate in an assembly of atoms is stable only if a is positive [1,2]. The scattering length a characterizes the scattering of a pair of atoms at ultralow collision energies. Its magnitude and sign are extremely sensitive to the details of the interatomic potential [3,4]. Photoabsorption in a gas of ultracold atoms provides information on the low energy ground state wave function [5]. We point out here that because a is property of the zero-energy wave function its sign can often be determined from measurements of photoassociation spectroscopy. By analyzing the intensities of the photoabsorption features obtained for lithium [6,7] we conclude that a is negative for scattering in the $a\,^3\Sigma_u^+$ state of the ^{7}Li atom pair and positive for scattering in the $X\,^1\Sigma_g^+$ state. For the triplet state, this conclusion is consistent with recent calculations [4,8,9] and an experimental determination of the scattering length [10].

In Fig. 1 we reproduce the measured photoassociation spectrum of a gas of ^{7}Li atoms at a temperature of several mK [7] as a function of the detuning to the red of the $2\,^2S_{1/2}$-$2\,^2P_{1/2}$ transition of ^{7}Li. There occurs a series of strong peak that persists to large detunings and a series of weak peaks that disappears beyond a detuning of 400 GHz. The locations of the peaks demonstrate that the strong series arises from the photoabsorption by $^7Li_2$ molecules in transitions from the free continuum $a\,^3\Sigma_u^+$ state to bound vibrational levels of the excited $1\,^3\Sigma_g^+$ state and the weak series from photoabsorption in transitions from the free continuum $X\,^1\Sigma_g^+$ state to bound vibrational levels of the excited $1\,^1\Sigma_u^+(A)$ state [7]. The relative intensities depend in part on the details of the experiment, but the qualitatively different behavior of the singlet and triplet series is definite; the triplet features persist deeper into the potential well. We confirm the interpretation by calculating the relative intensities of the strong and weak series.

The free-bound absorption rate coefficient at a probe laser frequency ν of a pair of atoms in the singlet or triplet state of the molecules with reduced mass μ at a temperature T may be written [11]

$$\kappa_{f\to b}(\nu,T) = \omega_f \frac{16\pi^3\nu}{3hc}\frac{h^3}{(2\pi\mu k_B T)^{3/2}}\pi\hbar \times \sum_v \exp(-E/k_BT) \times \sum_J \omega_{fJ}\Big[(J+1)\Big|\Big\langle u_b^{v,J+1}|D_{bf}|u_f^{E,J}\Big\rangle\Big|^2 + J\Big|\Big\langle u_b^{v,J-1}|D_{bf}|u_f^{E,J}\Big\rangle\Big|^2\Big], \quad (1)$$

where D_{bf} is the molecular dipole transition moment, $u_b^{v,J-1}$ and $u_b^{v,J+1}$ are the final vibrational v state wave functions corresponding to rotational quantum numbers $J-1$ and $J+1$, respectively, $u_f^{E,J}$ is the initial continuum energy-normalized wave function with rotational angular momentum quantum number J and asymptotic kinetic energy E, and ω_f and ω_{fJ} are statistical weights. If E_b is the energy of the bound state v relative to the dissociation limit of the initial state of $^7Li_2$, the energy E is related to the detuning Δ and the frequency ν through $h\nu = E_b - E = \hbar\omega_p + \hbar\Delta$, where $\hbar\omega_p$ is the energy difference between the dissociation limits of the excited and ground states. The weights ω_f and ω_{fJ} depend on

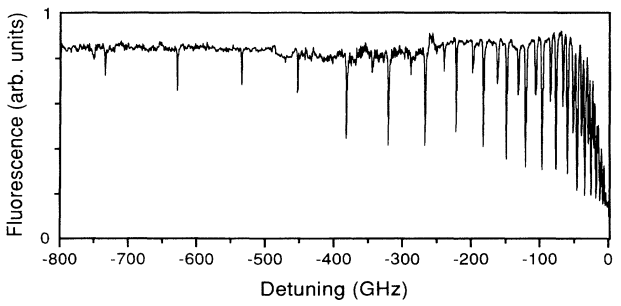

FIG. 1. Experimental trap-laser-induced fluorescence spectrum for ^{7}Li as a function of the detuning Δ in GHz. The fluorescence is directly proportional to the photoassociation spectrum.

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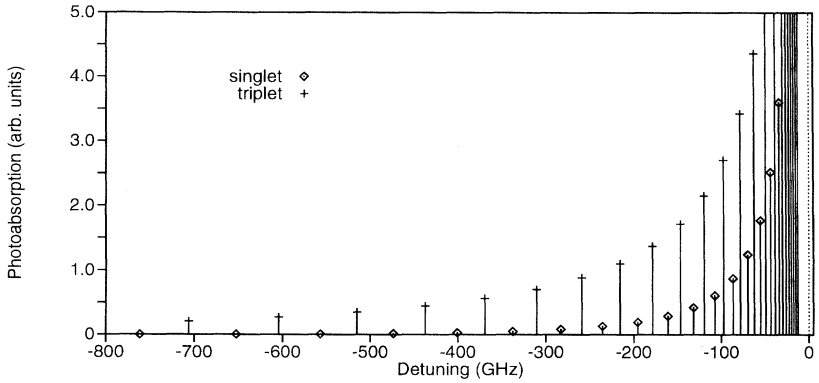

FIG. 2. Relative theoretical photoabsorption strengths for ^{7}Li at 1 mK as a function of the detuning Δ in GHz. The values for $|\Delta| < 50$ GHz extend outside the frame of the figure.

the initial hyperfine state of the interacting atoms and on the nuclear spin I. For a statistical distribution of hyperfine states in $^7Li_2$, $\omega_f = 1/32$, $\omega_{fJ} = 5$ for J odd and 3 for J even for the singlet transitions, and $\omega_f = 3/32$, $\omega_{fJ} = 3$ for J odd and 5 for J even for the triplet transitions [12].

To calculate the radial wave functions we solved the partial wave equations with the singlet and triplet ground state potentials of Côté, Dalgarno, and Jamieson [4] and singlet and triplet excited state potentials constructed from the available empirical Rydberg-Klein-Rees data [13,14], and *ab initio* data [15] and theoretical values of the long range coefficients [16]. For the transition dipole moments we used the calculated values of Ratcliff, Fish, and Konowalow [17], scaled to the accurate limiting form of Marinescu and Dalgarno [18].

Figure 2 presents our predicted spectrum at a temperature of 1 mK at which only s-wave scattering contributes in the initial free state. In the figure, we show the peak intensities and we do not include the highest levels near zero detuning, because for them spin-orbit and other couplings, which we ignore, are important. The theoretical photoabsorption spectrum is the mirror image of the experimental spectrum of Fig. 1. The theory predicts, as observed, a strong triplet series and a weak, much less extended, singlet series.

The relative intensities of the absorption peaks are related directly to the singlet and triplet scattering lengths. We show in Fig. 3 the bound state wave functions of the $v = 89$ level of the $1\,^1\Sigma_u^+(A)$ state, corresponding to a detuning of 107 GHz, and the $v = 80$ level of the $1\,^3\Sigma_g^+$ state, corresponding to a detuning of 120 GHz. The wave functions are rapidly oscillating with small amplitude with the maximum amplitude occurring for the extended outer lobes centered at separations R between $80a_0$ and $85a_0$. The free wave functions for an energy $k_BT = 3.2$ mK are also illustrated in Fig. 3. It is evident that because of cancellation at smaller R the major contribution to the dipole matrix element comes from the outer lobe region.

A more quantitative understanding can be reached by considering the zero temperature limit. In the outer region, the free wave function takes its asymptotic form [19]

$$u_f^{E,0}(R) \simeq \left(\frac{2\mu}{\pi\hbar^2 K}\right)^{1/2}\left(1 - \frac{R}{a}\right)\sin\delta_0(K), \qquad (2)$$

where K is the wave number such that $\hbar^2K^2 = 2\mu E$ and $\delta_0(K)$ is the s-wave scattering phase shift which varies for low K as

$$\sin\delta_0(K) \sim -Ka + \mathcal{O}(K^2). \qquad (3)$$

Replacing $D_{bf}(R)$ by $D_{bf}(\overline{R}_v)$ where $\overline{R}_v$ is the center of the outer lobe, we obtain the approximate formula

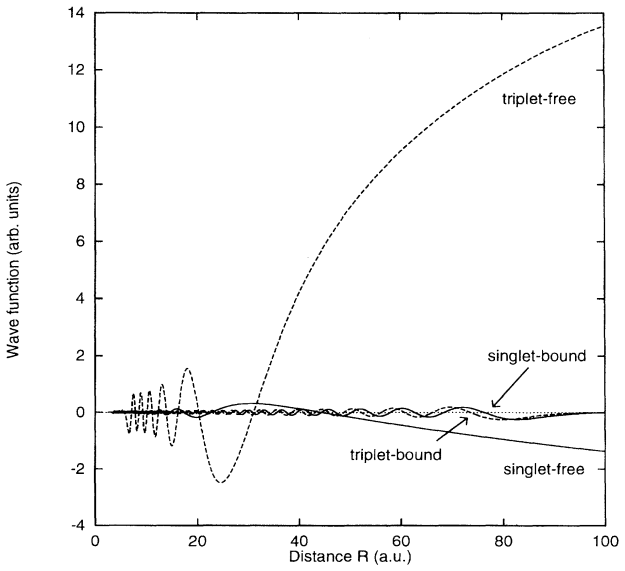

FIG. 3. Bound singlet ($v = 89$) and triplet ($v = 80$) wave functions and free singlet and triplet wave functions at an energy of $k_BT = 10^{-8}$ a.u. (3.2 mK).

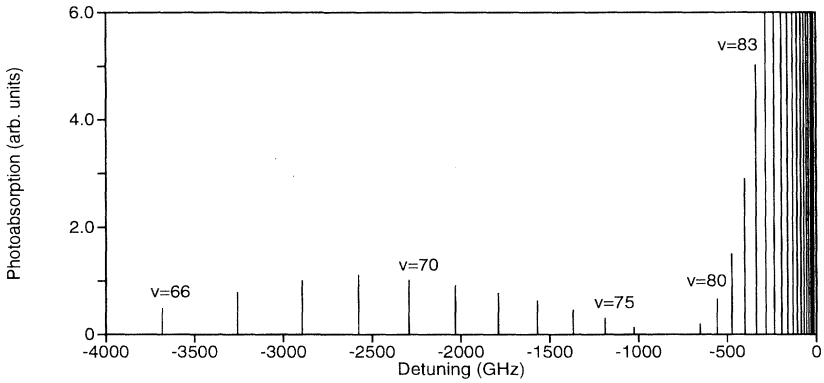

FIG. 4. Photoabsorption for the singlet transitions of ^{7}Li. The labels v are the vibrational quantum numbers of the excited $1\,^1\Sigma_u^+(A)$ state. The intensities for $v > 83$ extend beyond the scale of the figure.

$$\left|\left\langle u_b^{v,1}|D_{bf}|u_f^{E,0}\right\rangle\right|^2 \simeq [D_{bf}(\overline{R}_v)]^2\left(\frac{2\mu}{\pi\hbar^2 K}\right)\left(1-\frac{\overline{R}_v}{a}\right)^2 L^2\left|u_b^{v,1}(\overline{R}_v)\right|^2 \sin^2\delta_0(K)$$

$$\simeq \left(\frac{2\mu K}{\pi\hbar^2}\right)[D_{bf}(\overline{R}_v)]^2 (a-\overline{R}_v)^2 L^2\left|u_b^{v,1}(\overline{R}_v)\right|^2, \tag{4}$$

where L is the width of the outer lobe. The formula suggests immediately that the intensities will be generally larger for negative a than for positive a.

As the detuning increases and lower vibrational levels are accessed, $\overline{R}_v$ diminishes. If $\overline{R}_v$ is above $|a|$ for high v, the matrix element will decrease slowly with v in the case of a negative a and rapidly with v in the case of a positive a, passing through zero when $\overline{R}_v \sim a$ and then increasing for lower v. This behavior is demonstrated in Fig. 4 which shows the calculated photoabsorption spectrum for the singlet transitions of ^{7}Li. The scattering length for the initial state is positive with magnitude $37a_0$.

The theoretical values do indeed show this behavior for $^7Li_2$. Although the relative intensities of the experimental data in Fig. 1 are affected by details of the experiment, they are sufficiently consistent with the theory to demonstrate conclusively that the $^7Li_2$ triplet scattering length is negative and the singlet scattering length is positive, in agreement with calculations using the most reliable interatomic potentials [4,8,9] and the triplet scattering length inferred from a spectroscopic measurement of the least bound state of the $a\,^3\Sigma_u^+$ ground state potential [10]. Reliable values of the magnitude a can be obtained by more quantitative data on the relative strengths of the fluorescence peaks.

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