

Photoassociative frequency shift in a quantum degenerate gas

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A light-induced frequency shift is observed in single-photon photoassociative spectra of magnetically trapped, quantum degenerate ^7Li . The shift is a manifestation of the coupling between the threshold continuum scattering states and discrete bound levels in the excited-state molecular potential induced by the photoassociation laser. The frequency shift is observed to be linear in the laser intensity with a measured proportionality constant that is in good agreement with theoretical predictions. This phenomenon has important implications for a scheme to alter the interactions between atoms in a Bose-Einstein condensate using photoassociation resonances.

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Photoassociative spectroscopy of trapped atomic gases has been used to measure interatomic interaction potentials with ultrahigh sensitivity [1]. Although most photoassociation experiments with trapped atoms have been performed at temperatures near 1 mK, sub- μK temperatures can be achieved using evaporative cooling. This is the temperature regime that recent experiments on Bose-Einstein condensation (BEC) [2–4] have been conducted. There are several important improvements to photoassociative spectroscopy that are realized with a quantum degenerate gas. Spectroscopic precision can be increased to unprecedented levels since the energy spread of atoms at sub- μK temperatures is less than 20 kHz. Furthermore, the photoassociation rate is enhanced since it increases proportionally with phase space density $n\lambda_D$ [5], where n is the atomic density and λ_D is the thermal de Broglie wavelength. Finally, higher light intensities can be achieved since at sub- μK temperatures, the spatial extent of the trapped gas can be very small, enabling tighter focusing of the photoassociation laser beam. We have exploited these enhancements to investigate the effect of light intensity on single-photon photoassociation spectra of a magnetically trapped, evaporatively cooled gas of ^7Li . In particular, we have measured a spectral shift proportional to the light intensity [5–8]. This shift is relevant to proposed schemes for utilizing photoassociation to alter the interactions between atoms in a Bose-Einstein condensate [6,7] and for producing ultracold, trapped molecules [5,9–19].

The apparatus used in this experiment, which has been used to produce BEC of ^7Li , has been described previously [20]. Permanent magnets establish an Ioffe-Pritchard-type trap with a depth of 10 mK and a bias field of 1004 G at the trap center. Approximately 5×10^8 atoms in the $F=2$, $m_F=2$ hyperfine sublevel of ^7Li are directly loaded into the trap from a laser-slowed atomic beam using three-dimensional optical molasses. Following loading, the laser beams are extinguished and the atoms are evaporatively cooled using a microwave field to selectively spin flip, and thereby remove the hottest atoms. The final temperatures are between 400 and 650 nK, corresponding to between 3×10^5 and 1×10^6 atoms. Under these conditions, the gas is quantum degenerate, although the fraction of atoms in the condensate is small due to attractive interactions in lithium [21].

Following evaporation, a photoassociation laser beam of frequency ω_1 , and intensity I , is passed through the trapped atoms. A schematic representation of the relevant molecular potentials and energy levels is shown in Fig. 1. A vibrationally and electronically excited molecule may form when ω_1 is tuned to resonance between the continuum level of two

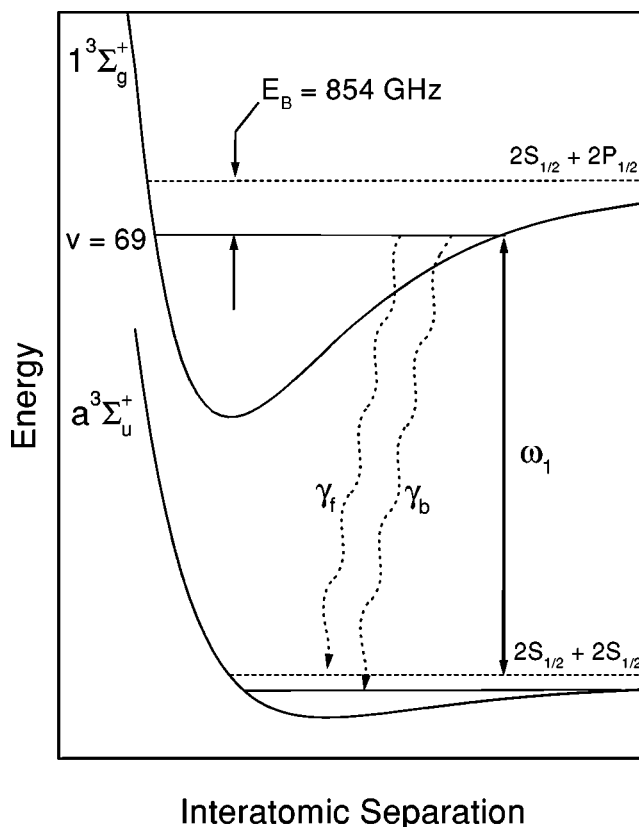


FIG. 1. Relevant molecular potentials and energy levels. The upper potential dissociates to a $2S_{1/2}$ and a $2P_{1/2}$ atom while the lower potential dissociates to two $2S_{1/2}$ atoms. The spontaneous emission rates γ_b and γ_f are to bound and free states, respectively. The binding energy of the $v=69$ level is given relative to dissociation. A laser of frequency ω_1 drives the transition between a state of two free atoms and an electronically excited molecule. Since the atoms are spin polarized, they only couple to the spin-triplet potentials.

colliding atoms and a bound level in the excited-state molecular potential. The excited molecule may spontaneously decay, most probably into a pair of hot atoms or possibly into a ground-state molecule, resulting in a detectable reduction of trapped atoms. The trapped cloud is probed *in situ* using the phase-contrast imaging technique described in Ref. [20] in order to determine the number of remaining atoms N . Since photoassociation removes a significant number of atoms, only one image may be obtained per evaporative cooling cycle. Therefore, to build up a resonance curve, the entire cycle is repeated many times for different values of ω_1 .

The photoassociation light is derived from a low-power (~ 10 mW), grating-stabilized, external cavity diode laser. The laser is side locked to a 3 GHz free-spectral-range (FSR) scannable Fabry-Perot cavity in order to reduce acoustical jitter to an rms amplitude of ~ 1 MHz as measured with an optical spectrum analyzer. A 750 MHz FSR scanning etalon is used to measure the relative frequency separation between the diode laser and another laser that is locked to the $2S_{1/2}-2P_{3/2}$ atomic resonance. Slow feedback to the 3 GHz cavity maintains this frequency separation to within ~ 3 MHz. An ~ 8 mW beam from the diode laser is injected into a tapered optical amplifier, providing up to 300 mW of output power at the injected frequency. An acousto-optic modulator and a mechanical shutter are used to chop the amplified beam on and off. The beam is subsequently coupled into a single-mode optical fiber, reducing pointing jitter and intensity variation across the beam profile. The output of the fiber, limited to 70 mW, is focussed at the position of the atoms to a $1/e^2$ intensity radius that ranges between 60 and 120 μm . The laser beam waist is always larger than the $1/e$ density radius of the atoms of ~ 40 μm . The photoassociation laser beam is directed nearly parallel to the laser beam used for imaging the atom cloud, which allows for the imaging optics to be used to ensure spatial overlap of the photoassociation laser beam with the atom cloud.

The laser frequency ω_1 is tuned to near resonance with the $v=69$ vibrational level of the excited molecular potential. Since the binding energy of this level, 854 GHz, is much larger than either the 10 GHz fine-structure splitting of the $2P$ atomic state or any hyperfine interaction, the total electronic spin \mathbf{S} decouples from both the electronic orbital angular momentum \mathbf{L} and the total nuclear spin \mathbf{I} . In this case, the light field only couples to \mathbf{L} , and \mathbf{S} and \mathbf{I} do not change during the photoassociation transition. Therefore, the selection rules for the total spin $\mathbf{G}=\mathbf{S}+\mathbf{I}$, and its projection M_G , are $\Delta G=0$ and $\Delta M_G=0$ [22], and there is no first-order Zeeman shift in the transition energy. Since both the ground state and excited molecular potentials are Σ states, corresponding to zero projection of \mathbf{L} onto the internuclear axis, there is no change in the projection of \mathbf{L} , and the transition dipole is oriented along the trap magnetic field bias direction (0,0,1). Geometric constraints, however, require the photoassociation laser beam to propagate along the (1,1,1) direction, so that only 2/3 of the light intensity can be polarized along the transition dipole. In this paper, the reported intensities correspond to those actually measured.

Figure 2 shows several resonance curves corresponding to different values of I . The data points are normalized to back-

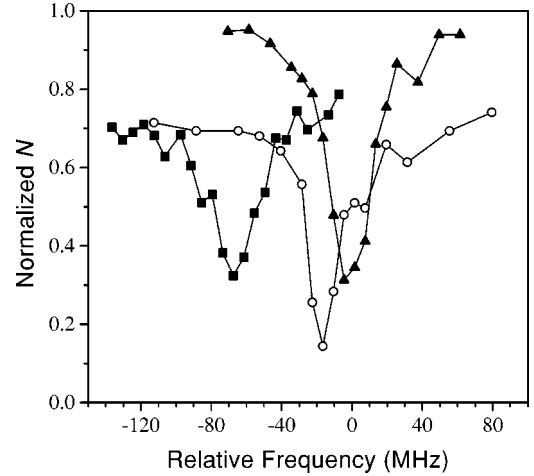


FIG. 2. Single-photon photoassociation resonance curves for the $v=69$ vibrational level of the $1^3\Sigma_g^+$ potential of $^7\text{Li}_2$. The signal is the normalized number of atoms remaining following the photoassociation pulse. The intensities, Gaussian $1/e^2$ intensity radii, and pulse durations are: 290 W/cm^2 , 120 μm , and 140 μs (solid squares); 53 W/cm^2 , 60 μm , and 1 ms (open circles); and 12 W/cm^2 , 95 μm , and 10 ms (solid triangles). At higher intensity, the signal baselines do not normalize to unity as the large intensity variation across the spatial extent of the atoms leads to a large dipole force perturbation.

ground images obtained without the photoassociation pulse to account for drift in trap loading conditions. The data clearly demonstrate that the resonance is red-shifted with increasing I . As the intensity is varied, the photoassociation pulse time is adjusted to maintain a relatively large and constant signal size. The resonance spectral widths are observed to be between 20 and 30 MHz. The natural linewidth for a long-range vibrational level, such as $v=69$, is $\sim 2\Gamma_a$ [23], where $\Gamma_a=2\pi\times 5.9$ MHz is the natural linewidth of the atomic $2P$ excited state of ^7Li . There are several other mechanisms that contribute to the broadening of the observed lineshapes. The relatively large depth of signal leads to a saturation broadening, which is the case even for low I since the pulse duration is extended to maintain a relatively constant signal size. Additionally, inhomogeneous broadening caused by the variation of laser intensity across the spatial extent of the atom cloud is expected to contribute as much as 10 MHz to the width for the largest values of I . The temperature of the gas is sufficiently low that thermal broadening is negligible compared with Γ_a . The observed resonance curves are sufficiently symmetrical to be fit reasonably well to a Lorentzian lineshape in order to locate the resonance center. Figure 3 shows the frequency shift of the measured resonance peaks as a function of I . These data fit a line of slope -245 ± 10 ($\text{kHz cm}^2/\text{W}$).

The origin of the frequency redshift is explained in Ref. [8]. It arises from coupling the various threshold scattering states to the excited-state bound level v and will be a generic feature of near-threshold resonant scattering. In particular, in the Fano theory for a bound state coupled to the continuum, the energy shift is proportional to the integral [24]

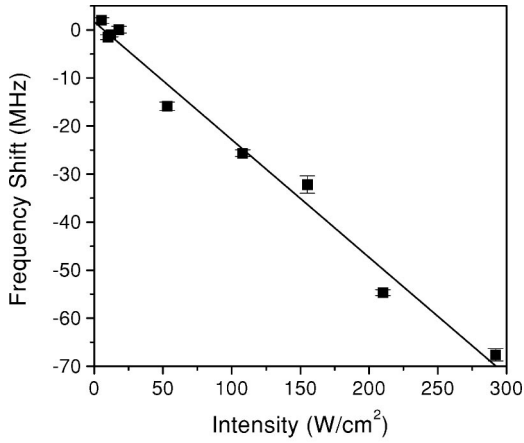


FIG. 3. Photoassociation frequency shift as a function of light intensity. The points correspond to the peaks of measured spectra obtained as described in the text. The line is a fit to these points giving a slope of -245 ± 10 (kHz cm²)/W. The error bars indicate the uncertainty in the fitted line center. The uncertainty in the light intensity is $\pm 10\%$. The quoted uncertainty in the fitted slope is the change required to increase the value of the goodness-of-fit parameter (unreduced χ^2) by unity.

$$\delta\epsilon \propto \int dE' \frac{V(E')^2}{E - E'}, \quad (1)$$

where $V(E')$ represents the density of continuum states at energy E' . The integrand in Eq. (1) is positive when $E' < E$ and negative when $E' > E$, where E is the unperturbed resonance energy. The density of continuum states $V(E')$ always increases versus energy E' no matter if the gas is confined to a trap, or in free space. Therefore, the negative part of the integral will contribute more strongly, and Eq. (1) gives rise to a red shift.

References [6] and [8] give simple, approximate expressions for the frequency shift. In Ref. [6], the magnitude of the frequency shift is given as $\beta\Omega^2/\delta\epsilon_v$, where, in their notation, $\Omega^2 = (\Gamma_a^2/2)I/I_{sat}$, $I_{sat} = 5.1$ mW/cm² is the saturation intensity of the atomic resonance, $\delta\epsilon_v$ is the energy difference between vibrational levels v and $v+1$, and β is a numerical factor that characterizes the overlap integral between the ground and excited-state wave functions. As per the discussion following Eq. (7) of Ref. [6], $\beta \approx 0.8\pi^2(1 - a_T/r_t)$ in the low-temperature limit, where $a_T = -27.6a_0$ is the ground-state triplet scattering length [25], a_0 is the Bohr radius, and r_t is the classical outer turning point of vibrational level v . For the $v=69$ level, $r_t = 44a_0$ and $\delta\epsilon = 120.6$ GHz [26], giving 242 (kHz cm²)/W for the magnitude of the theoretical light-induced shift, when the factor of $2/3$ accounting for the polarization of the photoassociation beam is taken into account. A prediction of 250 (kHz cm²)/W was obtained from the authors of Ref. [8]. Both predictions agree quite well with the data.

The primary subject of Ref. [6] is not the light-induced frequency shift, but rather an optical method for altering the interactions between ground-state atoms using photoassocia-

tion resonances. If the laser frequency is tuned near a molecular resonance, there is some probability that the atom pair will be excited to the upper potential shown in Fig. 1. If the laser detuning is made sufficiently large, the probability of excitation is small, but since the interaction potential of the excited state is much deeper than that of the ground state in the long-range region, a small excitation amplitude can have a substantial effect on the overall interaction [6,7]. The degree of change in interaction induced by the photoassociation laser, as well as the amount of induced loss, depends sensitively on the frequency detuning of the laser from the molecular resonance, necessitating a detailed understanding of the light-induced frequency shift described above.

This scheme for altering the interactions between atoms is an optical analog to magnetically-tuned Feshbach resonances [27], which have been used to alter the interactions between atoms in Bose-Einstein condensates [28,29]. Evidence for these optically induced Feshbach resonances has recently been obtained in a gas of ultracold sodium atoms [30]. There are several possible advantages of an optical method, such as the speed in which optical fields may be switched, the generality of the method for different atomic species, and the simplicity of applying optical fields, in comparison to changing magnetic field strengths. Such a technique may be particularly useful for probing the dynamics of condensate collapse in the case of attractive interactions [31–33], and we have performed a detailed analysis for a condensate of ⁷Li atoms.

Spontaneous light scattering induced by the photoassociation laser, even when detuned far from the molecular resonance, severely limits the utility of this technique for altering the interaction between condensate atoms. Our analysis suggests that this technique cannot be used to produce large condensates of ⁷Li atoms by making the self-interactions repulsive, because the induced loss rate exceeds the condensate growth rate. On the other hand, the interaction can be made more attractive for long enough (~ 10 ms) to induce a collapse for relatively small condensates without significant spontaneous scattering losses. Using $v=69$, we calculate that the magnitude of the scattering length a can be made three times larger for $I=600$ W/cm² and $\Delta = -165$ MHz. To achieve such an intensity with the ~ 70 mW of available laser power, the Gaussian $1/e^2$ intensity radius of the laser beam must be set to ~ 90 μ m. Under these conditions, however, we observe that the laser beam produces a substantial loss of trapped atoms accompanied by a significant change in shape of the atom cloud. We attribute these observations to heating of the atoms caused by the strong dipole force that arises from the large intensity gradient across the atom cloud. We conclude that substantially more laser power is required to make even modest changes to the scattering length using this technique.

In conclusion, we find that the light-induced shift is substantial, and must be taken into account, for example, in schemes for producing trapped ultracold molecules using photoassociation [5,9–19]. It is important to understand the laser-induced frequency shift since the success of these mol-

ecule production schemes depends on detailed knowledge of the intermediate-state spectral location. The origin of the shift is well understood in terms of continuum coupling, and its magnitude, predicted by semi-analytic theories, is in excellent agreement with experiment.

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