Photoassociation of a Bose-Einstein Condensate near a Feshbach Resonance

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We measure the effect of a magnetic Feshbach resonance (FR) on the rate and light-induced frequency shift of a photoassociation resonance in ultracold $^7$Li. The photoassociation-induced loss rate coefficient, $K_p$, depends strongly on magnetic field, varying by more than a factor of $10^4$ for fields near the FR. At sufficiently high laser intensities, $K_p$ for a thermal gas decreases with increasing intensity, while saturation is observed for the first time in a Bose-Einstein condensate. The frequency shift is also strongly field-dependent and exhibits an anomalous blue-shift for fields just below the FR.

Increasing interest in producing ultracold molecules by associating ultracold atoms has motivated an improved fundamental understanding of association processes. Photoassociation (PA) and magnetically-tuned Feshbach resonances (FR) are two ways in which gases of ultracold atoms may be connected to the molecular bound states of their underlying two-body interaction potentials. A Feshbach resonance is realized by tuning a molecular bound state near the scattering threshold $E_{th}$. The bound state perturbs the scattering state which can, in turn, strongly affect the rate of PA [2]. The rate of association of a Bose-Einstein condensate (BEC) is of particular interest, as a universal regime is predicted at extremely high rates of association [8, 9, 10, 11, 12] where the rate is limited by a universal transient response of the atom pairs [8], independent of the underlying microscopic association process.

Several groups have measured absolute PA rate coefficients [8, 9, 11, 11, 12]. Of these, however, only two were performed with a quantum gas. In ref. [8], the rate of PA of a sodium BEC was found to increase linearly with laser intensity, without any indication of saturation. We previously investigated PA of $^7$Li with laser intensity, without any indication of saturation. [18]. A uniform magnetic field can be set between 480–900 G to access both sides of the FR located near 736 G [19]. Evaporative cooling is achieved by reducing the optical trap beam intensity. Either Bose condensates with no discernable thermal fraction, or thermal gases with $T > T_c$, can be produced depending on the final optical trap depth.

The PA laser propagates collinearly with the optical trap beam, and is tuned to resonance (peak loss) with a vibrational level $v$ of the $1^3S_g^+$ excited molecular state $^3$Li. There are no unresolved rotational or hyperfine levels. For most of the measurements we tune to the $v = 83$ level, located 60 GHz below the $2^2S_{1/2} + 2^2P_{1/2}$ dissociation limit [21]. The $v = 83$ level is a good compromise between strong free-bound overlap and relatively weak off-resonance scattering from the atomic transition. Excited molecules created by the PA laser decay into pairs of energetic atoms that predominately escape the trap. The duration of the PA laser pulse $\tau$ is adjusted at each intensity $I$ and field to maintain the fractional loss of atoms to approximately 30%. The rise and fall times of the PA pulse are less than 0.3 $\mu$s. The number of remaining atoms following the laser pulse is measured using on-resonant absorption imaging.

The on-resonance loss-rate coefficient, $K_p$, is defined by the time evolution of the density distribution: $\dot{n}(t, r) = -K_p n^2(t, r)$, where any possible time-dependence of $K_p$ during the PA pulse is averaged over. For the BEC data (Fig. 4), $\tau$ is much less than either trap period, and collisional redistribution during the PA pulse can be neglected. In this case, we solve for the time-dependent density distribution analytically and extract $K_p$ by spatially integrating the density and matching the observed and calculated loss after a time $\tau$ [8]. For much of the thermal data (Figs. 1 and 3), however, $\tau$ is comparable to the radial trap period and redistribution is not negligible. Without accounting for the details of the time evolution, we take $K_p$ to be the average of two estimates: the first...
FIG. 1: $K_p$ for PA to $v=83$ for a thermal gas. $I$ is fixed at 1.65 W/cm$^2$, while $\tau$ is adjusted between 0.07 and 270 ms to keep the fractional loss at $30 \pm 6\%$ ($\tau \propto 1/K_p$). The fitted temperatures range between 9–18 K. The spread in initial peak density, 0.5 – $10 \times 10^{12}$ cm$^{-3}$, reflects a significant decrease in density near the FR, which we ascribe to enhanced density near the FR, which we ascribe to enhanced intensity. The statistical uncertainty is 20%. The measurement assumes no redistribution, as for the BEC case, while the second assumes that the distribution remains Boltzmann throughout. Due to the small fractional loss, we find the difference between the two estimates to also be small (<25%), and account for it in the stated uncertainties.

Figure 1 shows the field dependence of $K_p$ on both sides of the FR for a thermal gas. The data show a pronounced dip in $K_p$ at 710 G, and a maximum, which is more than $10^4$ times larger, located between 725–750 G. Measurements could not be made closer to the FR due to high inelastic collisional loss rates, presumably from a three-body process. A condensate could not be probed on the high-field side of the FR due to its instability for $a < 0$, but at a given field on the low-field side $K_p$ for a condensate was found to be approximately half that of a thermal gas, consistent with quantum statistics.

The variation in $K_p$ can be understood in terms of the Franck-Condon principle and the dependence of the scattering state wavefunction $f(R)$ with magnetic field ($R$ is the internuclear separation). PA predominately occurs at the Condon radius, $R_C$, defined as the outer classical turning point of the excited vibrational energy state. Since $K_p \propto |f(R_C)|^2$, a node in $f(R)$ at $R_C$ results in a minimum in $K_p$. Such minima have been previously observed [21], although with much lower contrast. For sufficiently low $T$ that the collisional wave vector $k$ satisfies $k(R - a) \ll 1$, the asymptotic scattering state $f(R) \sim k^{1/2}(R - a)$, and a node in $f(R)$ appears at $R \simeq a$ [22]. The inset to Fig. 1 shows that the location of the $K_p$ minima shift to higher fields (larger $a$) with increasing $v$, consistent with the fact that $R_C$ increases with $v$. The peak in $K_p$ near 736 G, on the other hand, is a manifestation of the enhancement of $f(R_C)$ when $|a|$ is large due to the FR. The ratio of the asymptotic values of $K_p$ on the high and low field sides of the FR is $\sim 3.5$, which can be understood from the ratio of $|f(R)|^2$ evaluated using the values of $a$ at the two asymptotic fields. Using a coupled-channel calculation [23] with previously determined potentials for lithium [24], we find the scattering lengths at 600 G and 900 G to be $a_1 \simeq 8 a_o$ and $a_0 \simeq -43 a_o$, respectively, and $R_C \simeq 103 a_o$ for $v = 83$, where $a_o$ is the Bohr radius. With these values, the expected ratio is $(R_C - a_0)^2/(R_C - a_1)^2 \simeq 2.4$, in reasonable agreement with measurement.

The effect of the FR on $f(R)$ is also evident in the light-induced spectral shifts. We measured spectral shifts for both a thermal gas and a condensate for fields around the FR. The location of a PA resonance at a particular field and intensity $I$ is determined by fitting the resonance lineshape to a Lorentzian. By taking resonance curves for several values of $I$, we observe that the spectral shift is linear in $I$, and extract the slope, $\Sigma$, plotted in Fig. 2. Far from the FR, the shift is red ($\Sigma < 0$), in agreement with previous measurements of single-photon PA [8]. As the FR is approached from low field, however, $\Sigma$ changes sign, becoming large and positive just below the FR and large and negative just above it in a dispersive-like manner. The field where $\Sigma$ vanishes (710 G), coincides with the location of the zero in $K_p$ shown in Fig. 1.

The spectral shift is a consequence of light-induced mixing of nearby molecular states into $f(R)$, and has been shown to scale linearly with $f(R_C)$ [10]. In previous single-photon experiments the shift was dominated by coupling to the continuum, resulting in a red shift. In the case of PA near a FR, however, the shift is strongly affected by the underlying closed-channel molecular state responsible for the FR. For fields below the FR, the contribution to the shift from the bound state is positive, while above resonance it is negative.

The data of Fig. 1 demonstrate that $K_p$ can be extraordinarily large near the FR, making it an ideal system for exploring saturation. Figure 3 shows measured values of $K_p$ vs. $I$ for a thermal gas at a field just above the FR. This data can be analyzed in terms of a Fano model of a bound state coupled to a continuum, where the on-resonance intensity-dependent loss rate coefficient can be
where $K$ is the energy dependence of the low temperatures in our experiment we neglect any counts for broadening of the PA resonance. Because of the high-intensity regime is still evolving, but a frame-

ciferies saturation. By fitting the measured lineshape to a fit to Eq. 1, which yields 4.1 W/cm$^{2}$. The data clearly show the predicted rollover in $K_p$ we obtain

$\Sigma = 4.1 \times 10^{-9}$ cm$^{3}$/s and $I_{sat} = 4.1$ W/cm$^{2}$. A linear fit to the slope for small $I$ gives $8.2 \pm 1.1 \times 10^{-9}$ cm$^{3}$ s$^{-1}$/W cm$^{-2}$, where the uncertainty refers only to statistical uncertainty.

work for comparison of the data may be established by defining $K_p$ in terms of a characteristic length, $L$, as $K_p = (h/m)L$. The smallest relevant length scale is the average interatomic separation, evaluated at the peak density $n_o$. Taking $L = n_o^{-1/3}$, we obtain the “rogue photodissociation” limit, $K_{pd}$, a universal regime where saturation in the rate of condensate loss is predicted due to the dissociation of excited molecules into the hot pair continuum. The NIST BEC experiment achieved $K_p \approx K_{pd}$, but with no indication of saturation. For the data of Fig. 4, $n_o = 1.6 \times 10^{12}$ cm$^{-3}$, giving $K_{pd} \sim 8 \times 10^{-9}$ cm$^{3}$/s. Our measured $K_{max}$ is nearly 20 times greater than $K_{pd}$. More recent calculations, however, have shown that while dissociation does impose a rate limit on condensate loss, it is not as stringent as $K_{pd}$. While the highest- $I$ data of Fig. 4 are well into the “dissociation regime”, our measured $K_{max}$ is, nonetheless, nearly 7 times greater then predicted from the equations given in ref. [3]. We note that in the experiment atoms removed from the condensate but left with energies below the trap depth (500 nK) may be indistinguishable from condensate atoms by our detection method. This effect cannot explain the discrepancy with theory, however, as it would result in an apparent suppression of $K_p$ at the onset of photodissociation.

The observed saturation could be explained by a higher than expected limit imposed by dissociation, perhaps due
to cross-coupling between the PA and Feshbach resonances. An alternative explanation is provided by quantum mechanical unitarity. If we take $L \propto 2R_{TF}$, where $R_{TF} \approx 10 \mu m$ is the radial Thomas-Fermi radius, then $K_a \sim 1.8 \times 10^{-7} \text{cm}^3/\text{s}$, in good agreement with the measured value of $1.4 \times 10^{-7} \text{cm}^3/\text{s}$. A preprint has recently appeared that directly models our experiment with good quantitative results, but it does not determine whether the rate limit is imposed by dissociation or unitarity.

We have presented well-characterized measurements of PA near a FR, which we hope will serve to constrain and guide theory, especially in the previously unexplored strong coupling regime. Condensate loss rates as high as the unitarity limit have been demonstrated by combining PA and Feshbach resonances. Future studies should explore the dynamical behavior of loss since $K_p$ is predicted to be time-dependent in the dissociation regime. Finally, Feshbach enhanced PA may prove useful in realizing the long-sought goal of coherent oscillation between atomic and molecular condensates or for efficient production of ground-state molecules using a two-photon process.

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