Probing the unitarity limit at low laser intensities

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Abstract. We show how photoassociation (PA) near a magnetically tuned Feshbach resonance can probe the unitarity limit at low laser intensities. This limit occurs when the scattering matrix element in the PA process reaches its quantum limit of one, and is usually difficult to access, requiring large laser intensities. We show how giant enhancements of the PA rate in the vicinity of a Feshbach resonance can reach it, and we predict the appearance of new features with varying magnetic fields and laser intensities. We investigate these effects for a Bose gas of $^7$Li atoms at temperatures near quantum degeneracy and give a simple analytical model. We discuss potential applications of our results, obtained without adjustable parameters, to high-precision spectroscopy.

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1. Introduction

Over the last decade, photoassociation (PA) of ultracold atoms [1] has been widely used to study long-range interactions and to form ultracold molecules [2, 3]. More recently, magnetoassociation (MA)—using magnetically induced Feshbach resonances [4]—has been crucial to realize molecular Bose–Einstein condensates (BEC) [5] and investigate the BEC–BCS cross-over regime [6]. Several phenomena of ultracold physics, e.g. vibrational quenching or reactions involving ultracold molecules [7], occur near the unitarity limit, where scattering matrix elements reach unity. To date, only a few PA experiments [8]–[12] have reached the saturation regime where the unitarity limit becomes accessible. Several effects could appear in PA at this limit, such as Autler–Townes like splittings [13] or blue light shifts [14] due to the proximity of high-lying levels of the electronic ground state.

In the present paper, we show how the unitarity limit can be readily accessed by combining elements of both PA and MA, and how new spectroscopic features appear. In addition, we demonstrate that our molecular physics based coupled-channel calculations—sensitive to the full wave functions of both scattering and target states—lead to remarkable agreements with recent measurements, without adjustable parameters. Such quantitative theoretical treatment of binary processes is crucial to reveal many-body effects present in ultracold gases, and is also required if one hopes to use ultracold atoms to measure the variation of fundamental constants [15].

Observation of saturation has been reported in a mixture of $^6\text{Li}–^7\text{Li}$ [8], and in an optical lattice loaded with Cs [9]. Theoretical studies [16] and experiments in BECs have also been conducted, but only PA in a sample of $^7\text{Li}$ reached the unitarity regime near the critical temperature [10, 11] and in a BEC [11], whereas PA in a BEC of Na showed no trace of saturation [12]. The range of intensities needed together with unfavorable transition matrix elements make the unitarity regime difficult to reach. More ideal candidates to attain this regime are alkaline earth metals with better Franck–Condon factors at large distance such as $^{86}\text{Sr}$ [17], or ultracold Rydberg gases where their strong interactions lead to large saturation effects [18]. However, large intensities might still be required for alkaline earth metals, with possible power broadening issues, multi-photon excitations or multi-photon ionization, and while saturation occurs at low laser intensities in ultracold Rydberg samples, the role of many-body physics can be crucial.

Combining both PA and MA techniques can overcome these difficulties and allow study of PA near the unitarity limit at low laser intensities, without the need for Floquet or multi-photon treatments. This approach, labeled Feshbach optimized photoassociation (FOPA), has been introduced in [19] (and references therein) for the formation of large amounts of ultracold molecules, and recently demonstrated experimentally in [11]. For the sake of clarity and simplicity, we focus on new features arising from the unitarity limit in non-degenerate ultracold gases. In particular, to compare with recent experiments of Hulet and co-workers [10, 11], we consider a bosonic gas of $^7\text{Li}$, where FOPA is performed to the $1^3\Sigma^+_g$ excited molecular state, with atoms initially prepared in the $|f = 1, m = 1\rangle$ or $|f = 2, m = 2\rangle$ hyperfine states (see figure 1). Scattering properties for these entrance channels are well understood and have been experimentally investigated [10, 11, 20].
Figure 1. FOPA: two colliding atoms (1) undergo a Feshbach resonance occurring when a bound level (2) (green wave function) coincides with the continuum state (blue wave function). The coupling between both open and closed channels enhances the probability density at short separation. A photon can then associate the pair of atoms into a deeper bound level $v$ (3) of an excited potential (red) to form a bound molecule.

2. Rate coefficient

The PA rate coefficient $K_{PA}^v$ into a molecular rovibrational level $v$ can be expressed in terms of the relative velocity $v_{rel}$ of the colliding pair and the PA cross section $\sigma_{PA}^v$, according to $K_{PA}^v = \langle v_{rel} \sigma_{PA}^v \rangle$. Here, the brackets stand for an average over the velocity distribution. For a temperature $T$ around and above the critical temperature $T_C$ for Bose–Einstein condensation, a Maxwell–Boltzmann velocity distribution is appropriate [21, 22]. At resonance and assuming a negligible linewidth for the PA laser of intensity $I$, $K_{PA}^v(T, I)$ becomes

$$K_{PA}^v(T, I) = \frac{1}{h Q_T} \int_0^\infty \frac{e^{-\varepsilon/k_B T}}{\varepsilon^2 + \left(\gamma_v + \gamma_s\right)^2} d\varepsilon,$$

where $\varepsilon = \mu v_{rel}^2/2$, $\mu$ is the reduced mass, $Q_T = (2\pi \mu k_B T/h^2)^{3/2}$, and $k_B$ and $\gamma_v$ are the Boltzmann constant and the natural linewidth of the photoassociated level [21, 22], respectively. The stimulated width $\gamma_s$ can be expressed using the Fermi golden rule as

$$\gamma_s = \frac{\pi I}{\varepsilon_0 c} |\langle \phi_{v',J=1}|D(R)|\Psi_{v',I=0}\rangle|^2,$$

where $\varepsilon_0$ and $c$ are the vacuum permittivity and speed of light, respectively. The wave functions $\phi_{v,J=1}(R)$ and $\Psi_{v',I=0}(R)$ of the excited level $v$ and the colliding pair of atoms vary with the internuclear separation $R$, as does the transition dipole moment $D(R)$.

The unitarity limit for the PA process corresponds to the maximum PA rate coefficient reachable for a given temperature. It is obtained when the scattering matrix element in equation (1) is equal to its maximum value of one: $\gamma_v \gamma_s / [\varepsilon^2 + ((\gamma_v + \gamma_s)/2)^2] = 1$. The integration over the Maxwell–Boltzmann velocity distribution can then be performed so that equation (1) gives

$$K_{limit}^v(T) = \frac{k_B T}{h Q_T} = \frac{1}{(2\pi \mu)^{3/2} \sqrt{k_B T}}.$$
We determine $|\Psi_{e,l=0}\rangle$ of two colliding atoms of relative momentum $\hbar k = \sqrt{2\mu \varepsilon}$ by solving the Hamiltonian [18]

$$H = \frac{\hbar^2 k^2}{2\mu} + V_C + \sum_{j=1}^{2} H_j^{\text{int}},$$

where $V_C = V_S(R) P^S + V_T(R) P^T$ is the potential interaction, separated into singlet (triplet) molecular potentials $V_{S(T)}$, with projection operators $P^S(T)$. Atom $j$’s internal energy $H_j^{\text{int}} = (a hf/\hbar^2) \vec{s}_j \cdot \vec{i}_j + (\gamma_e \vec{s}_j - \gamma_n \vec{i}_j) \cdot \vec{B}$ consists of the hyperfine and Zeeman contributions. Here, $\vec{s}_j$ and $\vec{i}_j$ are atom $j$’s electronic and nuclear spins with hyperfine constant $a_{hf}$, and $\vec{B}$ is the magnetic field. Since the nuclear gyromagnetic factor $\gamma_n$ is three orders of magnitude smaller than $\gamma_e$, we neglect it in our calculations.

Finally, we note that in order to compute the stimulated width $\gamma_s$ defined in equation (2), the scattering wave function $|\Psi_{e,l=0}\rangle$ is projected onto the triplet manifold.

3. Interaction potentials

In earlier work, we have explored PA of $^7\text{Li}$ atoms [23, 24]. However, for our precise calculations, an accurate knowledge of the ground singlet $V_S(R)$ and triplet $V_T(R)$ molecular interaction potentials is needed. We adopted the potentials described in [25]. These potentials were further adjusted to reproduced the scattering properties of two $^7\text{Li}$ atoms [10, 11, 20]. For each curve, we varied the inner wall by shifting the positions of the data points for separations less than the equilibrium separation $R_e$ according to

$$R_{\text{shifted}} = R + s \frac{R - R_e}{R_c - R_e},$$

where $s$ corresponds to the shift of the zero-energy classical turning point $R_c$. The best agreement with [10, 11, 20] was obtained with $s = -0.004$ au, $R_e = 5.04$ au, and $R_c = 3.44$ au for $V_S$, and $s = -0.0215$ au, $R_e = 7.88$ au and $R_c = 6.33$ au for $V_T$. Using these adjusted potentials, we reproduce the Feshbach resonance at 736 G [11] for the $|f = 1, m = 1\rangle$ entrance channel (see figure 2).

For the excited $^1\Sigma^+_g$ potential, we used the potential curve described in [22]. We note that target rovibrational levels close to the dissociation limit of the electronic excited state are not considered here. Thus, the Born–Oppenheimer potential remains a good approximation with the Hund’s coupling case (a) still valid. We also note that the spin–orbit and hyperfine interactions are negligible in our case, and that the Zeeman shift of the triplet excited curve in the presence of a magnetic field would not affect significantly the position of the calculated levels: all these effects are neglected as they do not affect the wave function of the excited rovibrational target state we consider.

Finally, we used the transition dipole moment $D(R)$ described in [22] for transitions from the triplet component of $|\Psi_{e,l=0}\rangle$ to rovibrational levels of the excited $^1\Sigma^+_g$ electronic state.

4. Results

We calculated the PA rate coefficient $K_{\text{PA}}^\nu$ to $\nu = 83$ (using $\gamma_\nu = 12$ MHz [22]) of the $^1\Sigma^+_g$ state as a function of the $B$ field at $T = 10 \mu$K. We choose these values of $\nu$ and $T$ to compare with
We extended a two coupled-channel treatment introduced in [19] to the stimulated rate of PA. A recent measurement at higher $I$ [11] (see below). Figure 2 shows $K_{PA}^{\nu=83}$ for $I = 1 \text{ mW cm}^{-2}$, which displays a maximum at resonance ($B \sim 736 \text{ G}$) and a minimum near $710 \text{ G}$, spanning over eight orders of magnitude. This variation of $K_{PA}^{\nu=83}$ can be understood with a simple model. We extended a two coupled-channel treatment introduced in [19] to the stimulated rate $\gamma_s$ of equation (2), and obtained

$$\gamma_s(I, k, B) = \gamma_s^{\text{off}} |1 + C_1 \tan \delta + C_2 \sin \delta|^2,$$

where $\gamma_s^{\text{off}}(I, k)$ is the off-resonance stimulated rate, $C_1(k)$ and $C_2(k)$ are dimensionless ratios of dipole matrix elements between closed and open channels (see [19]), and $\delta(k)$ is the resonant phase shift. For s-wave as $k \rightarrow 0$, $\gamma_s^{\text{off}}(I, k) \propto I k$, in agreement with Wigner’s threshold law, while $\tan \delta(k) = -k a_{bg} \Delta / (B - B_0)$ depends on the background scattering length $a_{bg}$, and the width $\Delta$ and the position $B_0$ of the resonance (see values in figure 2). The exact $k$ dependence of $C_1$ and $C_2$ varies with the target level $\nu$: they tend to a constant for a short-range/deeply bound state, and go as $k^{-1}$ for long-range/very extended state. Introducing equation (6) into equation (1), we obtain the solid curves in figure 2, with $\gamma_s^{\text{off}} = 0.8 \text{ kHz}$, and fitted values $C_1 = 6$, and $C_2 = 1$. The agreement with the full multi-channel treatment is excellent.

Before examining the effect of FOPA on saturation, we performed a calculation, using equation (1), at $B = 0$ and $T = 500 \text{ nK}$ to compare with experimental results for $^7\text{Li}$ atoms in $|f = 2, m = 2\rangle$ (a pure triplet interaction) [10]. Figure 3 shows the results: our calculated (solid curve) and measured (circles) rates are in agreement well past the range of validity of the perturbative prediction (dotted line) [10]. Saturation is observed for the highest reported intensities as the rate nears the theoretically predicted unitarity limit of $2.5 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1}$. We repeated the calculation for $|f = 1, m = 1\rangle$ (mostly triplet interaction) at $B = 0$ and near the resonance with $740 \text{ G}$: the $B = 0$ result (solid curve) is only slightly different from the pure
Figure 3. $K_{PA}^{v=83}$ into $^{13}\Sigma^+_g$ of $^7\text{Li}_2$ at $T = 500 \text{nK}$ versus laser intensity, with $B = 0$ (solid curve) and near resonance at $B = 740 \text{G}$ (dashed curve) for the $|1, 1\rangle$ state. We also compare our calculation for $|2, 2\rangle$ at $B = 0$ with the experimental data of [10] (circles). The dotted line is the perturbative rate of $5.8 \times 10^{-10}$ $(\text{cm}^3\text{s}^{-1}/\text{W cm}^{-2})$, while the horizontal dot-dashed line indicates the saturation limit of $2.5 \times 10^{-8}$ cm$^3$ s$^{-1}$.

triplet $|2, 2\rangle$ case, while the PA rate at $740 \text{G}$ (dashed line) increases rapidly until it reaches the saturation limit at intensities ten times smaller than for $B = 0$, to then slowly decrease. This particular shape can be explained by the integrand in equation (1) which takes the form $\gamma_s\gamma_v/[(\gamma_s + \gamma_v)/2]^2$ as $\varepsilon \to 0$; as $\gamma_s \propto I$ increases, so does the integrand, until it reaches its maximum of one at $\gamma_s = \gamma_v$, after which it starts to decrease as $\gamma_s^{-1} \propto I^{-1}$ at large intensities. We note that this behavior has recently been observed experimentally [11].

One can use this low intensity saturation to probe how reaching the unitarity limit may affect the PA lineshape. In figure 4, we display $K_{PA}^{v=83}$ as a function of the intensity $I$. We observe the appearance of a double-minimum structure that becomes more pronounced as $I$ grows. This feature is a consequence of the unitarity limit: as $I$ increases, the PA rate reaches its maximum value at $\gamma_s = \gamma_v$, and then decreases at higher $I$. However, $\gamma_s$ also varies with $B$ according to equation (6), and as $B$ goes through the Feshbach resonance at large enough $I$, $\gamma_s$ will reach the value $\gamma_v$ twice, once on the way ‘up’ and once on the way ‘down’ from the resonance, leading to two maximum values of $K_{PA}^{v=83}$. The insets compare the results from the multi-channel treatment (white circles) and the simple two-channel model results (red solid curves) using equation (6) for 1 mW cm$^{-2}$ and 10 mW cm$^{-2}$: in both cases, the agreement is good. We note that the double-minimum feature (figure 4) appears quickly, even at rather low intensity. While the first minimum arises from a small $\gamma_s$ due to a poor wave function overlap between entrance and target states (destructive interference) near a particular $B$ field, the nature of the second minimum is quite different. In fact, it occurs at the resonance $B_0$, where $\gamma_s$ is maximum and extremely large, so much that any pair of atoms photoassociated is photodissociated right away into the continuum (Rabi flopping), leading to a very poor formation rate.

A recent experiment reached the saturation regime at low intensity [11]. In figure 5, we compare our multi-channel results at $T = 10 \mu\text{K}$ and $I = 1.6 \text{W cm}^{-2}$ with those of [11], where the atom loss rate $K_p$ is twice our $K_{PA}^{v}$, since two atoms are lost during the PA process. Our results are within or close to the uncertainty of [11]; when taking into account the variation in temperatures between data points ($\sim 9–18 \mu\text{K}$ [11]), the discrepancy vanishes. We note that there are no adjustable parameters in our theory, and that such agreement for the absolute
rate coefficient is remarkable. Figure 5 also shows zooms of the minimum and the maximum. A different approach was recently used to explore the minimum [26]. We also note that the experimental points at saturation near the resonance are the least accurate, and that the point at 745 G was particularly challenging to obtain [11], consistent with the large and negative scattering length \( a \) (i.e. attractive interaction: see figure 2).
We note here that since FOPA reaches that maximum PA rate coefficient possible, one could select the excited level $v$ that spontaneously decays mostly into a deep level $v_g$ of the electronic ground state. For example, 57.4\% of $(v = 9, J = 1)$ in $1^3\Sigma^+_u$ decays into $v_g = 0$ of $a^3\Sigma^+_u$ [3], while 23.1\% of $(v = 3, J = 1)$ in $1^1\Sigma^+_u$ decays into $v_g = 0$ of $X^1\Sigma^+$. Assuming that the unitarity limit of $5.5 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ is reached in a sample of density $\rho = 10^{11} \text{ cm}^{-3}$ with an illuminated volume $V = 10^{-6} \text{ cm}^3$, a rate [27] of $R = r\rho^2 V K \sim 10^7 \text{ mol s}^{-1}$ is expected ($r$: branching ratio). This compares well with recent experiments using stimulated Raman adiabatic passage (STIRAP) to produce ground state molecules starting from a Feshbach bound state [28]; about $3 \times 10^4$ ground state molecules are produced during the entire cycle, including creation of Feshbach molecules, taking $\sim 10–30 \text{ s}$, i.e. about $10^3 \text{ mol s}^{-1}$.

Another characteristic of FOPA is the sensitivity of $K_{\text{PA}}^v$ to the exact level $v$ being probed. In fact, whereas the maximum of $K_{\text{PA}}^v$ near the resonance at 736 G does not vary much, the exact position of the minimum is very sensitive to $v$. In figure 6, we show $K_{\text{PA}}^v$ for $I = 1 \text{ mW cm}^{-2}$ as a function of $B$ for levels $v = 60–98$, assuming $\gamma_v = 12 \text{ MHz}$ for all those levels ($\gamma_v$ is roughly constant for this range of levels [22]). While the maximum remains at 736 G, the minimum follows a ‘croissant trajectory’ as $v$ decreases, first moving to lower $B$ at large $v$ and then to higher $B$ at smaller $v$, even passing to the right of the resonance at lower $v$. The reason for this sensitivity resides in the nature of the minimum, i.e. a poor overlap of the target level $v$ and scattering wave functions. Any small difference in the target wave functions will be amplified and will result in a shift of the minimum. It is worth noting that such a shift was observed in recent measurements for $v = 82–84$ [11], consistent with our results. This sensitivity to minute

\[ \text{Figure 5. Comparison of } 2K_{\text{PA}}^{v=83} \text{ (white circles) with data of } [11] \text{ (green squares) at } 10 \mu\text{K} \text{ and } 1.6 \text{ W cm}^{-2} \text{ for } ^7\text{Li}_2. \text{ The insets show the minimum and maximum (linear scale); only the point at } 745 \text{ G is outside the uncertainty of } 45\% \text{ [11]. The dashed horizontal line corresponds to twice the saturation limit of } 5.5 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}. \]
changes in the target level wave function could actually be used as a spectroscopic tool for high-precision measurements of both the scattering and bound states. Similar shifts take place at higher laser intensities, where the double-minimum feature allows for a more complex behavior.

We note that the very sharp variation of the PA rate near either minimum could become a possible tool to measure the variation of fundamental constants, such as that proposed for the fine structure constant $\alpha$ in [15].

5. Conclusions

In conclusion, we have shown that by marrying the PA with a Feshbach resonance into the FOPA technique, it is possible to access the unitarity limit at low laser intensity (e.g. ten times smaller than at $B = 0$, see figure 3). This approach removes problems related to multi-photon excitations/ionization. It also allows the probing of saturation effects such as double-minimum in lineshapes and the variation of the minimum rate with the level $v$ being probed. By adjusting the potential curves to match those positions, high-precision spectroscopy of scattering and bound states could be achieved. The remarkable agreement with recent measurements, obtained without adjustable parameters, illustrates the predictive power of our approach for binary processes. Finally, we note that since PA probes the density of states in the continuum [29], FOPA might yield interesting results in degenerate gases. In brief, FOPA is a promising new technique to probe the unitarity limit at low laser intensities, and could be used for tests of fundamental constants and to investigate many-body physics.

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