Suppression of power broadening in strong-coupling photoassociation in the presence of a Feshbach resonance

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Abstract
The photoassociation (PA) spectrum is analysed in the presence of a magnetic Feshbach resonance. A nonperturbative solution of the problem yields analytical expressions for PA linewidth and shift which are applicable for arbitrary PA laser intensity and magnetic field tuning of Feshbach resonance. We show that by tuning the magnetic field close to the Fano minimum, it is possible to suppress power broadening at increased laser intensities. This occurs due to quantum interference of PA transitions from unperturbed and perturbed continuum. Line narrowing at high laser intensities is accompanied by large spectral shifts. We briefly discuss the important consequences of line narrowing in cold collisions.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Over the years, photoassociation and Feshbach resonance have become important tools in the manipulation of ultracold collisions. Recent experimental [1–3] and theoretical [4–7] works on photoassociation (PA) near a magnetic field Feshbach resonance (MFR) [8] give rise to the exciting possibilities of coherent control of atom–molecule conversion. In a recent experiment, Junker et al [1] have demonstrated an asymmetric spectral line shape and saturation in PA in the presence of MFR. Asymmetric line shape is a characteristic feature of the Fano effect [9] which arises in different areas such as atomic [10], particle [11] and condensed matter physics [12]. When a pair of colliding atoms under the influence of the Feshbach resonance are photoassociated into an excited molecular state, PA transitions can occur in two competing pathways. The presence of a Feshbach resonance largely perturbs the continuum states of colliding atoms. As a result, continuum states get hybridized with one or more bound states embedded in the continuum. PA transitions from unperturbed and perturbed continuum states can interfere, resulting in the asymmetric Fano line shape. The unique aspect of the MFR-induced Fano effect in PA is that the continuum-bound coherence can be controlled by tuning the magnetic field. Feshbach resonances and photoassociation have a common feature: both are the effects of continuum-bound interacting systems. In the case of a Feshbach resonance, an initial continuum state is coupled to a bound state embedded in the continuum by the hyperfine interaction of two atoms. In the case of PA, the continuum state gets coupled to an excited diatomic molecular state by a single photon. Both can be treated within the framework of Fano’s theory. In fact, based on Fano’s method, theory of one- and two-photon PA has been developed by Bohn and Julienne [13, 14]. Two-photon PA [15] has been shown to lead to quantum interference. Furthermore, quantum interference has been demonstrated in the coherent formation of molecules [16].

Apart from MFR-induced modification, PA under intense laser fields (strong-coupling regime) can further modify the continuum states due to the strong continuum-bound dipole coupling. The Fano effect in the strong field regime [17–19] leads to ‘confluence of bound-free coherences’ [17] which can result in a number of effects such as line narrowing [20], Autler–Townes splitting [17] and nonlinear Fano effect [21]. Continuum-bound coherences in strongly interacting ultracold atoms can lead to a number of profound physical effects.
which may not be observed in the weak coupling regime. A decade ago, Vuletic et al [22] experimentally observed line narrowing in the spectrum of trap loss of atoms due to a tunable Feshbach resonance. In Fano’s theory [9], a continuum interacting with a bound state is exactly diagonalized leading to a ‘dressed’ continuum state. In linear Fano theory, the optical transition matrix element between this dressed continuum and any other bound state is obtained perturbatively by the use of a Fermi Golden rule. In the strong coupling regime, the optical coupling of the dressed continuum with any other state needs to be treated nonperturbatively either by a ‘double diagonalization’ technique [17] or by other diagonalization techniques such as used in [23].

Here we explore the possibility of suppression of power broadening in strong-coupling PA by manipulation of continuum-bound coherences with a Feshbach resonance. As the simplest possible model, we consider optically coupled two bound states interacting with a common continuum as shown in figure 1. We demonstrate that by tuning the magnetic field close to Fano minimum where excitation probability vanishes [9], it is possible to obtain line narrowing in the PA spectrum with large shifts at high laser intensities. Large light shift with narrow linewidth may be useful in the efficient tuning of elastic scattering length by optical means [24–26].

The paper is organized in the following way. In the following section, we discuss in brief the formulation of the problem. In section 3, we present the analytical results on line narrowing and enhancement of shifts. In section 4, we discuss the numerical results and the paper is concluded in section 5.

2. Formulation of the problem

The dressed state of a system of two colliding atoms interacting with a PA laser in the presence of MFR can be written as

\[
|\Psi_E\rangle = \frac{1}{r} \left[ \int dE' b_E r|\Psi_E^F (r)|1 + \sum_{i=2,3} \Phi_i (r)|i\right],
\]

where \(r\) is the relative coordinate of the two atoms, \(E\) is an energy eigenvalue, \(|1\rangle\) (|2\rangle) represents the internal electronic states of a open(closed) channel |1\rangle |2\rangle and |3\rangle denotes the electronic state of the excited molecule. \(\Phi_i (i = 2, 3)\) and \(\Psi_E^F\) are the wavefunctions of the perturbed bound and continuum states, respectively. Here \(b_E\) is the density of states of the unperturbed continuum. In writing the above equation, we have assumed that the PA laser can couple only a particular ro-vibrational state of the excited molecule, and the bound state \(\Phi_2\) has zero angular momentum. Let the hyperfine spin coupling between channels 1 and 2 be denoted by \(V(r)\). Let \(\Omega_1 (r)\) and \(\Omega_2 (r)\) represent the molecular Rabi couplings of the excited state |3\rangle with the ground states |1\rangle and |2\rangle, respectively. In the absence of these three couplings, let the unperturbed bound states be denoted by \(\phi_1 (r)\) and \(\phi_2 (r)\) with bound state energies \(E_1\) and \(E_2\) respectively, and the unperturbed continuum states by \(\psi_E^F\) with asymptotic collision energy \(E'\).

With the use of these unperturbed solutions, we construct three Green’s functions \(G_E (r,r')\), \(G_2 (r,r')\) and \(G_3 (r,r')\) which correspond to channels 1, 2 and 3, respectively. The continuum Green’s function \(G_E (r,r')\) can be written as

\[
G_E (r,r') = -\pi \psi_E^{reg} (r_s) \psi_E^{reg} (r_s), \quad \text{where } r_{(s)} \text{ implies either } r \text{ or } r' \text{ whichever is smaller (greater) than the other. Here } \psi_E^{reg} (r) = \psi_E^{reg} + i \psi_E^{irr} \text{ where } \psi_E^{reg} \text{ and } \psi_E^{irr} \text{ represent regular and irregular scattering functions, respectively. Asymptotically, } \psi_E^{reg} (r) \sim j_0 \cos q_0 r - j_0 \sin q_0 r \text{ and } \psi_E^{irr} (r) \sim (-j_0 \cos q_0 + j_0 \sin q_0) r. \]

Thus at low energy, \(q_0 r \sim k\) is related to Feshbach resonance linewidth \(\Gamma_r \sim k C\), where \(C\) is a constant and \(\mu\) is the reduced mass of the two atoms. \(a_{res}\) is related to the applied magnetic field \(B\) by \(a_{res} = -\frac{\mu a_B}{E_3}\) [27], where \(a_B\) is the background scattering length, \(E_3\) is the resonance magnetic field, \(\Delta\) is a parameter which depends on \(\Gamma_r\) and magnetic moments of the atoms at \(E_3\). Having done so, we can obtain the solutions in the form \(\Phi_3 (r) = A_{PA} \phi_3 (r)\) and \(\Phi_2 (r) = A_{CC} \phi_2 (r)\). The explicit form of \(A_{PA}\) [23] is given by

\[
A_{PA} = \frac{\exp (i \mu q)}{\beta + 1} \frac{\pi \Omega_E}{(\beta + 1)(\Delta_p + i \Gamma_{PA}/2)} - \frac{\pi \Omega_E}{(\beta + 1)(\Delta_p - i \Gamma_{PA}/2)},
\]

where \(\Delta_p = \hbar \delta + E(E_3 - E_{shunt})\). \(E_{shunt}^0\) is the energy shift in the absence of a Feshbach resonance, \(\Omega_E = \int d r_{(s)} \Omega_3 (r) \psi_E^{reg}\) is the continuum-bound molecular dipole coupling and \(\Gamma_{PA}\) is the stimulated linewidth of PA given by \(\Gamma_{PA} = 2 \pi |\Omega_E|^2\). Here \(q\) is Fano’s asymmetry parameter defined by

\[
q = \frac{\Omega_3 + V_{23}}{\pi \Omega_E V_{23}},
\]

where \(\Omega_3 = \int d r \phi_3 (r) \Omega_3 (r) \phi_2\) is the bound–bound Rabi coupling and \(V_{23} = \int d r' \phi_2 (r) V(r) \text{Re} \{G_E (r, r')\}\). \(\Omega(r) \phi_3 (r)\) represents an effective interaction between the two bound states mediated through their couplings with the common continuum. Note that \(q\) is independent of the laser intensity. In the limit \(k \to 0\), both \(\Omega_3\) and \(V_{23}\) become energy independent while \(\Omega_E V_{23} \sim k\). Thus at low energy, \(q \sim 1/k\). The detailed derivation of the dressed state is given elsewhere [23], but for completeness we reproduce the derivation in appendix.
3. Analytical results

The loss of atoms due to the decay of the excited state into the decay channels (which is modelled as an artificial scattering channel) is described by the PA rate

\[
K_{PA} = \langle |\langle \psi_{art}| V_{art} |\psi \rangle |^2 \rangle = \frac{1}{\hbar QT} \int dE \frac{\hbar^2 \gamma \Gamma \exp(-E_d/\hbar B_T)}{(E_k - \Delta E + \hbar \delta_p)^2 + \hbar^2 (\gamma + \Gamma)^2/4},
\]

where \( \langle \cdots \rangle \) means thermal averaging over the collision energy and \( \hbar \delta_p = \hbar \delta - (E_3 + E_{th}^0 - E_0) \) is the detuning parameter. Here \( E_{th} \) is the threshold of the open channel. Here \( QT = (2\pi \mu \hbar B_T / \hbar^2) \), \( E_k = E - E_0 \) and \( B_T \) is the Boltzmann constant.

The extra shift caused by MFR is given by

\[
\Delta E = \frac{1}{2} \left[ \frac{(q^2 - 1)\beta - 2q}{\beta^2 + 1} \right] \Gamma_{PA}.
\]

(6) shows that \( \Gamma \) depends on a nonlinear function \( f(q, \beta) \) of \( q \) and \( \beta \). Note that when \( \beta \to \pm \infty \), that is, far away from MFR, \( \Gamma \to \Gamma_{PA} \). It is to be further noted that when \( \beta = -q \), we have \( \Gamma = 0 \) and \( \Delta \Gamma = 0 \) at which PA ceases to occur. The Fano minimum is given by \( \beta_{min} = -q \) or equivalently the corresponding magnetic field \( B_{min} \). Thus \( \Gamma \) can be made arbitrarily small by tuning \( \beta \) close to \( -q \). It is possible to suppress power broadening at increased laser intensities by tuning the magnetic field \( B \) close to \( B_{min} \).

Next, we discuss the weak-coupling limit of (6) and (7) when laser intensity is low. For this we first find the dressed continuum state in the limit \( \Gamma_{PA} \to 0 \). As \( \Gamma_{PA} \to 0 \), \( \Lambda_{CC} \) can be expressed as

\[
\Lambda_{CC} = -\frac{2}{\pi \Gamma_f} \exp \left( i\eta_0 + \eta_{res} \right) \sin \eta_{res}
\]

and \( \psi_E(r) \) becomes

\[
\psi_E(r) = \exp \left( i\eta_0 + \eta_{res} \right) \left[ \psi_{E,reg} \cos \eta_{res} + \psi_{E,irr} \sin \eta_{res} \right].
\]

So in the limit of \( \Gamma_{PA} \to 0 \), at low energy the state \( |\psi_E\rangle \) of (1) reduces to

\[
|\psi_E\rangle_0 = \frac{1}{r} \left[ A_{CC} \phi_2(r)[2] + \int b_E^* \psi_E(r) dE' [1] \right].
\]

Taking \( b_E^* = \delta(E - E') \), the stimulated linewidth \( \Gamma^{weak} \) in the weak coupling limit is given by the Fermi golden rule expression

\[
\Gamma^{weak} = 2\pi \int r \phi_{VM}(r)[3|\Omega_1|r]|\psi_E\rangle_0 dE |^2
\]

\[
= \Gamma_{PA} |C_1 + C_2 \sin \eta_{res} + C_2 \tan \eta_{res}^2|,
\]

where \( C_1 = \Omega_{1E}^0 / \Omega_3E \) and \( C_2 = ( - \sqrt{2} ) \Omega_{1E} / \Omega_3E \). Here \( \Omega_{1E} = \{3|\Omega_1|\psi_{E,irr}^0 \} \). Expression (11) is in agreement with (6) of [5]. When \( \eta_{res} \to \pi/2 \), \( \Gamma^{weak} \) diverges and hence (11) is not valid near \( \eta_{res} = \pi/2 \). In other words, (11) is not applicable close to the Feshbach resonance.

Here we show how the exact expressions of \( \Gamma \) and \( \Delta E \) as given by (6) and (7), respectively, do enable us to realize the possibility of suppression of power broadening with enhanced light shift at increased laser intensities. It is clear from (7) that \( \Delta E \) goes to zero as \( \beta \to \pm \infty \). From (6) and (7), we note that if the laser intensity \( I \) is increased by a factor \( M_I \), to suppress power broadening \( \beta \) is to be changed to \( \beta' \) such that \( f(q, \beta') = M_I f(q, \beta) \). For \( \beta \approx -q \) we have \( \Delta E \approx q \Gamma_{PA}/2 \) which is proportional to \( I \). Hence, when line broadening is suppressed by the tunability of MFR, the total
the absence of the coupling between open and closed channels. It is clear from the above expression that in continuum and the excited bound state and also between the open channel. In the limit \( \beta \rightarrow 0 \), \( \Gamma \rightarrow \Gamma_{\text{weak}} \).

Finally, we prove that line narrowing in one-photon PA is not possible in the absence of coupling between the open and closed channel. It can be noted that the PA laser can be tuned either near continuum-bound frequency in which case \( \hbar \delta_p \simeq E_k \) or near bound–bound transition frequency \( (E_3 - E_2)/h \) in which case \( -\hbar \delta_p = (E_3 - E_2) = \Delta_{\text{res}} \). Here \( E_1 = E_3 + E_{\text{shift}}^0 \) and \( E_2 = E_2 + \Delta E_2, \Delta E_2 \) being the shift of the closed channel bound state due to its coupling \( V \) with the open channel. In the limit \( \Gamma_{\text{r}} \rightarrow 0 \),

\[
K_{\text{PA}} \sim \frac{\hbar^2 \gamma \Gamma_{\text{PA}}}{\left( E - E_3 + E_{\text{shift}}^0 + \hbar \delta - \frac{k^2 \gamma^2 C_{\text{PA}}^2}{E - E_2} \right)^2 + \hbar^2 (\gamma + \Gamma_{\text{PA}})^2/4},
\]

which is in agreement with the expression of \( |S_{1,2}|^2 \) of [14] if we identify \( \Delta_1 \) and \( \Delta_2 \) of [14] with \(-\hbar \delta \) and \( E_2 \), respectively. In our case there is only one laser coupling between the continuum and the excited bound state and also between the two bound states. It is clear from the above expression that in the absence of the coupling between open and closed channels the narrowing of PA linewidth is not possible.

4. Numerical results and discussion

For numerical illustration, we consider a model system of two ground-state \( (S_{1/2}) \) \(^7\) Li atoms undergoing PA from the ground molecular configuration \( ^3\Sigma_u^+ \) to the vibrational state \( v = 83 \) of the excited molecular configuration \( ^1\Sigma_u^+ \) which correlates asymptotically to \( 2S_{1/2} + 2P_{1/2} \) free atoms [28, 29]. All the relevant parameters \( \gamma, E_{\text{shift}}^0, \Delta, \alpha_{\text{res}} \) and \( \Gamma_{\text{r}} \) are estimated from [28, 30, 31]. In figure 2, we have plotted \( \Gamma/\Gamma_{\text{PA}} \) and \( \Delta E/\Gamma_{\text{PA}} \) against \( 1/k_{\text{res}} \) for positive and negative \( q \) values. The maximum and minimum values of linewidth would be observed for \( \beta = 1/q \) and \( \beta = -q \), respectively. The magnitude of the change in shift due to PA in the presence of MFR is significant near \( \beta = -q \).

Figure 3 clearly shows that the stimulated linewidth in the weak coupling limit, represented by dashed lines, deviates appreciably from nonperturbative results as shown by solid lines. The deviations are the most prominent in the region \((k\alpha_{\text{res}})^{-1} \sim 0/(\alpha_{\text{res}} \simeq \pi/2)\). Furthermore, for lower \( q \) values these two results deviate most significantly. Figure 4 illustrates how to suppress power broadening by the appropriate tuning of the magnetic field near \( B_{\text{min}} \) and thereby to keep the total linewidth close to the natural linewidth. There are two values of \( \beta/(B) \) and correspondingly two values of \( \alpha_{\text{res}} \), where the linewidth \( \Gamma \) can be kept fixed at a small value at an increased laser intensity. In figure 5, we show how to vary \( \Gamma_{\text{PA}} \) (or laser
intensity) and the magnetic field in order to keep $\Gamma$ fixed at 0.04 MHz. The lower inset shows the variation of $\Delta E$ (in MHz) against $\Gamma_{PA}$ (in MHz) and the upper inset exhibits the variation $a_{int}$ (in nm) against $B$ (in Gauss) at the fixed $\Gamma = 0.04$ MHz.

\[ \frac{h^2}{2\mu} \frac{d^2}{dr^2} + B_1(r) \Phi_3 + \left[ V_e(r) - \hbar \delta - E - i\hbar \gamma / 2 \right] \Phi_3 = -\Omega_1 \chi - \Omega_2 \Phi_2. \]  

(A.1)

Using Green’s functions $G_3(r, r')$, we find $\Phi_3 = A_{PA} \phi_3$, where

\[ A_{PA} = \frac{\int dr' [\Omega_1(r') \chi(r') + \Omega_2(r') \Phi_2(r')] \phi_3(r')}{\hbar \delta + E - E_3 + i\hbar \gamma / 2}. \]  

(A.4)

In a similar way, using $G_2(r, r')$ we can write $\Phi_2 = A_{CC} \phi_2$, where

\[ A_{CC} = \frac{[A_{PA} \Omega_3 \phi_2 + \int dr' \chi(r') \Omega_1(r') \phi_3(r')] E - E_2}{\hbar \delta + E - E_3 + i\hbar \gamma / 2}. \]  

(A.7)

Here $D = \hbar \delta + E - E_3 + i\hbar \gamma / 2$, $\tilde{V}_{2E} = \int dr \phi_2(r) V(r) \psi_E(r)$ and $\Omega_{3E} = \int dr \phi_3(r) \Omega_1(r) \psi_E(r)$. Using Green’s function $G_E(r, r')$, from equation (A.6) we obtain

\[ \psi_E = \exp(i\eta_0) \psi_E^{0, \text{reg}} + \int dr' G_E(r, r') \left[ \Omega_1^{(r')} \tilde{A}_{PA} \phi_3(r') \right] \]  

(A.8)

Now using this solution, we can calculate the probability amplitude of excitation in the following form:

\[ \tilde{A}_{PA} = \frac{\exp(i\eta_0) \Omega_{3E} + G V_{2E}^{*}}{D - \left| \frac{B_p + G B_f}{E - E_2} \right|^2}, \]  

(A.9)

where

\[ V_{2E} = \int dr \phi_2(r) V(r) \psi_E^{0, \text{reg}}(r) \]  

(A.10)

and

\[ \Omega_{3E} = \int dr \phi_3(r) \Omega_1(r) \psi_E^{0, \text{reg}}(r). \]  

(A.11)

The other parameters here are

\[ G = \frac{[\Omega_{3E} + V_{2E} - i\pi V_{2E} \Omega_{3E}]}{[E - E_2 - \Delta E_2 + i\pi |V_{2E}|^2]}, \]  

(A.12)

\[ B_p = E_{\text{shift}}^0 - i\pi \left[ \Omega_{3E}^2 \right] - \Omega_{3E} \frac{[V_{2E}^2 + i\pi V_{2E} \Omega_{3E}]}{[E - E_2]}. \]  

(A.13)
and

\[ B_f = V_{32} - i\pi V_{2E} \Omega_{3E} - \Omega_{32} \frac{[-\Delta E_2 + i\pi V_{2E}^2]}{(E - E_2)}. \]  

(A.14)

where

\[ E_{\text{shift}}^0 = \int \int dr' dr \phi_3(r) \Omega_1^* (r) V(r') \tilde{\Gamma}_2 \Omega_1 (r) \phi_3 (r'), \]  

(A.15)

\[ V_{32} = \int \int dr' dr \phi_2 (r) V(r') \tilde{\Gamma}_2 \Omega_1^* (r) \phi_3 (r'), \]  

(A.16)

\[ \Delta E_2 = \int \int dr' dr \phi_2 (r) V^* (r') \tilde{\Gamma}_2 \Omega_1 (r') \phi_2 (r'). \]  

(A.17)

Here, we introduce two new parameters \( \beta = (E - E_2 - \Delta E_2) / (\hbar \Gamma_2 / 2) \) and \( q = (\Omega_{32} + V_{32}) / (\pi \Omega_{3E} V_{2E}) \). Now after some algebra we can express (A.9) in the following form:

\[ \tilde{A}_\text{PA} = \frac{\exp \left( i \eta_0 \beta + q \right) \pi \Omega_{3E}}{\beta + i \left[ \Delta_\text{P} + i\hbar (\gamma + \Gamma_{\text{PA}}) / 2 \right] - \hbar \Gamma_{\text{PA}} (q - i)^2 / 2}. \]  

(A.18)

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