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Formation of a molecular ion by photoassociative Raman processes

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Abstract
We show theoretically that it is possible to form a cold molecular ion from a pair of colliding atom and ion at low energy by photoassociative two-photon Raman processes. We explore the possibility of stimulated Raman adiabatic passage from the continuum of ion–atom scattering states to an ionic molecular state. We provide physical conditions under which coherent population transfer is possible in stimulated Raman photoassociation. Our results are important for experimental realization of PA in ion–atom cold collisions.

Keywords: photoassociative Raman process, STIRAP, ion–atom cold collision, molecular ion, dressed continuum, dark state

(Some figures may appear in colour only in the online journal)

1. Introduction

In recent years, a number of schemes has been used to synthesize cold molecules from cold atoms in the presence of external fields. One of the most important methods in this regard is the photoassociation (PA) of cold atoms [1–4]. One- or two-color PA has been successfully employed to produce neutral cold molecules in excited or ground-state electronic potentials, respectively. Another important method is magnetoassociation [5, 6] by which exotic Feshbach molecules [7–9] are produced in the presence of an external magnetic field. Although the formation of cold molecular ion by PA between a cold atom and a cold ion had been theoretically predicted [10] and analyzed [11] over the last few years, the experimental realization of ion–atom PA is yet to be achieved. The difficulty in experimental ion–atom PA mainly stems from the fact that it is difficult to obtain sufficiently low temperature (down to sub-milliKelvin) in a hybrid ion–atom system [12–16] as required for ion–atom PA process. As ions

are usually trapped and cooled by radio-frequency fields, the major hindrance in cooling trapped ions arises from the trap-induced micro-motion of the ions. But this difficulty can be overcome by using optical traps [17] where both ions and atoms can be trapped and cooled. Experimental progress towards optical trapping of ions is underway [18–22] and so one can expect that ion–atom PA will soon be realized in an optical trap.

In this paper, we present and analyze a theoretical method for creating cold molecular ion from a colliding pair of alkali ion and alkali atom by photoassociative Raman processes. Our model is schematically shown in figure 1. For illustration, we consider $(LiCs)^+$ system, but our theoretical formulation is most general and can be applied to any other ion–atom system. We consider molecular level structure in Λ-type configuration involving one continuum of ion–atom scattering states and two molecular bound levels. As depicted in the inset of figure 1, the continuum of states $|E\rangle$ in the electronic ground-state manifold of the ion–atom system is
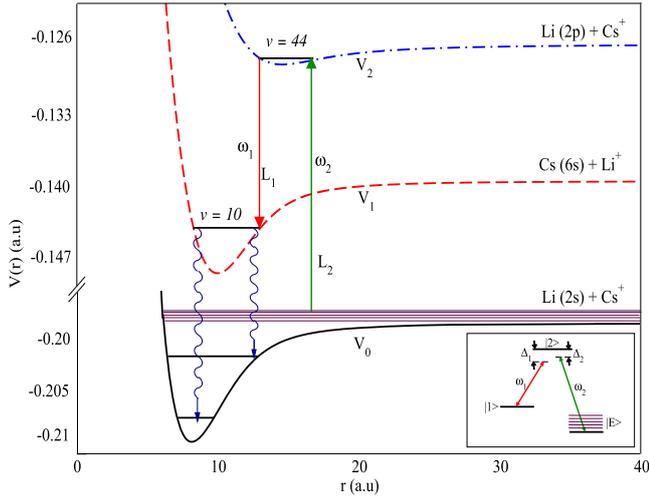
Figure 1. Adiabatic potential energy curves of the three lowest electronic states of (LiCs)$^+$ ion. The asymptotic states of separated ions and atoms are indicated. Three states can be considered like a Λ type system as indicated above. The band of horizontal lines above the threshold of the lowest potential indicate the continuum of states of low energy collision between the closed-shell Cs$^+$ ion and ground-state Li atom. The inset shows a three-level Λ-like structure with one lower ground-state sub-level in discrete Λ-system being replaced by the continuum. Three potentials are in $^2\Sigma^+$ electronic state.

coupled to the excited ro-vibrational bound state $|2\rangle$ in the electronic potential $V_2(r)$ by PA laser L_2 and $|2\rangle$ is coupled to the ro-vibrational bound state $|1\rangle$ in a lower potential $V_1(r)$ by laser L_1. The resulting configuration looks like a three-level Λ system with one of the lower ground-state sub-levels being replaced by the continuum. Here, we are interested in transferring population from $|E\rangle$ to $|1\rangle$ via the intermediate state $|2\rangle$ by a two-photon Raman process. One can consider two possible processes—incoherent and coherent Raman processes to accomplish the state transfer. In the incoherent process, one laser photon from L_2 excites the system to make an upward transition $|E\rangle \rightarrow |2\rangle$ followed by bound–bound downward transition $|2\rangle \rightarrow |1\rangle$ by spontaneous emission of a photon. The net result is the formation of the bound state $|1\rangle$. The success of such incoherent Raman process in molecular systems relies on the availability of favorable Franck–Condon (FC) overlap between the continuum and the bound state and also between the two bound states. Nevertheless, the efficiency of transfer is in general limited in the case of incoherent Raman processes. In a recent paper Côté and coworkers [23] have theoretically discussed the possibility of forming molecular ion by incoherent Raman photoassociation.

An efficient way for state transfer is the coherent method of stimulated Raman process using two laser pulses. When the two slowly varying time-dependent laser pulses are applied in a counter-intuitive way, that is, if laser L_1 is applied first between the target ($|1\rangle$) and the intermediate state ($|2\rangle$) which are initially empty, and then after a time delay the laser L_2 is applied between the initial and the intermediate state, the resultant process of state transfer is popularly known as stimulated Raman adiabatic process (STIRAP) [24–30] subject

to the fulfillment of an adiabatic condition. In the case of discrete three-level Λ systems driven by two Raman lasers, associated with a host of coherent processes including STIRAP is the existence of a dark state (DS) which is a special eigenstate of the system. The coupling of the excited intermediate state with the DS is zero. An adiabatic evolution of the DS can be executed by slowly varying the intensity of the applied lasers in a proper sequence, allowing nearly lossless population transfer between two ground-state sub-levels.

The possibility of STIRAP via an intermediate continuum of states is discussed by different groups [31–34]. Besides, taking continuum as an initial or final target state, state transfer is theoretically studied [35, 36]. STIRAP in a PA system is a subject of such category of state transfer. Indeed, the possibility of STIRAP in such systems is a matter of intense theoretical debate [37–40]. Photoassociative adiabatic passage in ultra-cold rubidium atomic gas has been studied [41, 42]. In 2005, Lett and coworkers experimentally observed EIT-like spectral features in two-color PA of ultra-cold sodium [43]. Experimental signatures of a dark resonance has also been found in ultra-cold meta-stable helium by Cohen-Tannoudji's group [44] and in ultra-cold strontium atoms by Killian and co-workers [45]. Although, these findings indicate to the possibility of STIRAP from cold atomic gas to cold molecules, such process is not yet clearly established theoretically or experimentally. Particularly, the connection of a dark resonance to the anticipated STIRAP in continuum systems is not known. Furthermore, STIRAP involving a structured continuum in the presence of a magnetic Feshbach resonance has been addressed in a few recent papers [46–49], where the initial state is chosen to be a quasi-bound state embedded in the continuum. However, the population transfer in such systems with Feshbach resonance does not occur directly from or to the continuum. Because, on elimination of the continuum, such systems effectively reduce to Λ-type discrete three-level systems where one can explore the standard STIRAP. In contrast, our work differs in an important way from the previous studies in that we have considered the continuum as an initial or target state for efficient state transfer. In our work, we address the question whether STIRAP from or to a continuum state can be accomplished via a short-lived molecular bound state.

In this paper, we adopt the Fano diagonalisation method [50] to obtain a dressed continuum state and find the physical conditions under which STIRAP involving the continuum is possible. We show that the dressed-continuum can be treated as a quasi-DS (which we defined later) under certain physical conditions. We explore the regime where the quasi-DS can be adiabatically evolved to perform a STIRAP-like process to transfer the atomic population from scattering continuum to molecular population in electronic ground-state. We use a strongly dipole allowed PA transition to an excited molecular ionic state which, at long separation, corresponds to an ion in s-state and a neutral atom in p-state. This excited state is short-lived. Here we use this molecular state as an intermediate state in performing STIRAP. We use two laser pulses in a counter-intuitive way. We have shown that effective coherent population transfer is possible.

This paper is organized in the following way. In section 2, we describe our theoretical model and analyze the condition for formation of DS. Numerical results are presented and discussed in section 3 In last section we make some concluding remarks.

2. The theoretical formulation

2.1. The model

Here we present our theoretical formulation using a model system of $(LiCs)^+$ as depicted in figure 1. The adiabatic potential data of $(LiCs)^+$ system are calculated using pseudopotential method and described elsewhere [51]. Initially the system is in the continuum states $|E\rangle$ of the ground-state adiabatic potential $V_0(r)$. The potential $V_0(r)$ asymptotically corresponds to the heavier alkali element in the ionic state (Cs^+) and the lighter one in the charge-neutral state (Li). The potential $V_1(r)$ asymptotically corresponds to the charge-exchanged state of that of $V_0(r)$. For both $V_0(r)$ and $V_1(r)$ potentials, the ion and the neutral atom are in electronic ground-state (S) at long separations. Therefore, the molecular electric dipole coupling between the electronic states of these two potentials vanishes at large separations while it is finite but small at short separations only. As a result, the ro-vibrational states supported by $V_1(r)$ are expected to be long-lived or meta-stable. However, accessing these states via free-bound transitions from the continuum of $V_0(r)$ is extremely difficult due to the poor FC overlap integral at short separations. In contrast, the transition dipole moment between the electronic states of $V_0(r)$ and $V_2(r)$ is large and constant at large separations where the potential $V_2(r)$ correspond to the lighter element in the excited P state while the heavier one remains in the ground ionic state as in the case of $V_0(r)$. Therefore, the electric dipole transition between the electronic states of $V_0(r)$ and $V_2(r)$ is similar to the strongly allowed electric dipole $E1$ transition. On the other hand, the ro-vibrational states in $V_2(r)$ are short-lived. So, from the practical point of view, it will be difficult to produce or probe the molecular states in $V_2(r)$ by photoassociation. Our primary purpose here is to form a meta-stable bound state in $|1\rangle$ in the potential $V_1(r)$ via two-photon coherent Raman process using the lossy state $|2\rangle$ as an intermediate state. Once a molecular ion is formed in the potential $V_1(r)$, the molecular ion in the ground-state potential $V_0(r)$ can be created either by spontaneous or stimulated bound–bound emission. Interestingly, since the ro-vibrational state in $V_1(r)$ is meta-stable, population inversion between two molecular levels in $V_0(r)$ and $V_1(r)$ may be achieved. At low temperatures (milli or sub-milliKelvin regimes), depending on the rotational quantum number of the excited molecular state that is coupled by PA laser, only one or two or a few partial waves of the ion–atom scattering state can contribute to the free-bound FC factor. This allows us to select a narrow range of energy for

continuum-bound coupling as elaborated in the next section.

For the particular system $(LiCs)^+$ under consideration, we find that vibrational state $v_2 = 44$ in $V_2(r)$ has significant FC factor with the continuum states of $V_0(r)$ at energies ranging from microKelvin to milliKelvin regime. We notice that the inner turning point of $v_2 = 44$ lies at almost same separation of the outer turning point of vibrational state $v_1 = 10$ in $V_1(r)$ potential. This means that, according to FC principle, these two vibrational states are the most useful or favorable for our purpose. The state $|1\rangle$ is coupled to $|2\rangle$ via laser pulse L_1 with frequency ω_1 and electric field $\mathbf{E}_{L1}$. $|2\rangle$ is coupled to the continuum of states $|E\rangle$ via laser pulse L_2 having frequency ω_2 and field $\mathbf{E}_{L2}$. We denote E_{v_1} (E_{v_2}) as the energy of the bound state $|1\rangle$ $(|2\rangle)$. The Hamiltonian of the system under rotating wave approximation (RWA) can be expressed as $H = H_0 + H_I$, where

$$H_0 = \hbar (\Delta_1 - \Delta_2)|1\rangle\langle 1| + (-\hbar\Delta_2|2\rangle\langle 2|) + \int E'|E'\rangle\langle E'|\mathrm{d}E' \tag{1}$$

is the free part and

$$H_I = \int \Lambda_{E'2}|E'\rangle\langle 2|\mathrm{d}E' + \hbar G_1|1\rangle\langle 2| + \mathrm{H.c.} \tag{2}$$

is the interaction part of the Hamiltonian. Here $G_1 = \frac{1}{\hbar}\langle 1|\mathbf{D}.\mathbf{E}_{L1}|2\rangle$ is the bound–bound Rabi frequency and $\Lambda_{2E} = \langle 2|\mathbf{D}.\mathbf{E}_{L2}|E\rangle$ is the free-bound coupling parameter, $\mathbf{D}$ being the molecular electric dipole moment. The detuning parameters introduced above are defined as $\Delta_1 = \omega_1 - (E_{v_2} - E_{v_1})/\hbar$ and $\Delta_2 = \omega_2 - E_{v_2}/\hbar$. The limits of the energy integrals are $[0, \infty[$ and remain the same throughout the paper. Following the Fano diagonalisation method the dressed eigenstates of H can be written as

$$|E\rangle_{\mathrm{dr}} = B_{1E}|1\rangle + B_{2E}|2\rangle + \int F_E(E')|E'\rangle\mathrm{d}E', \tag{3}$$

where B_{1E}, B_{2E}, $F_E(E')$ are expansion coefficients as defined in the appendix. Since $|E\rangle_{\mathrm{dr}}$ is energy normalized we must have $\int_{\mathrm{dr}}\langle E'|E\rangle_{\mathrm{dr}}\mathrm{d}E = 1$ which gives

$$\int |B_{1E}|^2\mathrm{d}E + \int |B_{2E}|^2\mathrm{d}E + \int\int |F_E(E'')|^2\mathrm{d}E\mathrm{d}E'' = P_1 + P_2 + P_C = 1, \tag{4}$$

where P_1, P_2 and P_C denote the probability that an atom-pair is in the state $|1\rangle$, $|2\rangle$ and the continuum, respectively.

2.2. DS condition

As stated earlier, for the systems involving a continuum, the existence of a DS is not obvious, and perhaps an exact DS is not possible. We search both analytically and numerically for a condition for which the contribution of the excited intermediate state to the dressed state equation (3) is small for a wide range of collisional energy. The purpose is to use the dressed state continuum as DS that can be used for reasonably efficient STIRAP. The expression for B_{2E} (for derivation, see the appendix) is given by

$$B_{2E} = \frac{[E + \hbar\,(\Delta_2 - \Delta_1)][E + \hbar\,\Delta_2 - E_1^{sh} - i\hbar\,\Gamma_{2E}/2]\Lambda_{2E}}{[E + \hbar\,(\Delta_2 - \Delta_1) - E_2^{sh}][(E + \hbar\,\Delta_2 - E_1^{sh})^2 + (\hbar\,\Gamma_{2E}/2)^2] + i\hbar^3\Gamma_{2E}G_1^2/2}. \tag{5}$$

Clearly, this cannot be zero for all energies. For a particular set of Δ_1 and Δ_2 (such that $\Delta_1 > \Delta_2$), the exact two-photon resonance condition is satisfied for a particular energy $E = \hbar\,(\Delta_1 - \Delta_2)$. Let us denote this energy as $\bar{E} = \hbar\,(\Delta_1 - \Delta_2)$. In case of a discrete three-level Λ system, DS condition is exactly fulfilled only at two-photon resonance. For our model with one continuum, the two-photon resonance condition has changed due to continuously varying energy of one of the lower state. Since $E \geqslant 0$, the two-photon resonance condition for $\bar{E}$ can only be fulfilled if $\Delta_1 > \Delta_2$, implying that the bound–bound one-photon detuning Δ_1 must be greater than the one-photon continuum-bound detuning Δ_2 which is defined as $\Delta_2 = \omega_2 - (E_{\nu_2} - E_{th})/\hbar$, where E_{th} is the threshold value of the open (continuum) channel.

Thus for $E = \bar{E} = \hbar\,(\Delta_1 - \Delta_2)$ the coefficient B_{2E} vanishes. As a result, the dressed continuum of equation (3) reduces to the form

$$|\bar{E}\rangle_{dr} = \bar{B}_{1E}|1\rangle + \int \bar{F}_E(E')|E'\rangle\,dE'. \tag{6}$$

If the coupling of this state with the excited state $|2\rangle$ vanishes then we can regard this as a DS. This means that

$$\langle 2|H_I|\bar{E}\rangle = 0. \tag{7}$$

This condition may be fulfilled by adjusting the phase of the two lasers. So, a DS for the entire range of continuum energy cannot be achievable. However, if we consider that the system is kept at very low temperature so that the kinetic energy distribution of the free atoms are very narrow about $\bar{E}$ then most of the atoms lie very near to $\bar{E}$.

Assuming the condition as mentioned above prevails, we define the DS condition as $P_2 = \int |B_{2E}|^2\,dE \ll 1$ as this would lead to small contribution of $|2\rangle$ to the dressed state. We call this state as a quasi-DS. As shown in the appendix, the expressions for P_1 and P_2 are

$$P_1 = \frac{\hbar^3}{2}|G_1|^2 \int Z^{-1}\Gamma_{2E} \\ \times (E + \hbar\,\Delta_2 - E_1^{sh})^2\,dE \\ + \frac{\hbar^5}{8}|G_1|^2 \int Z^{-1}\Gamma_{2E}^3\,dE \tag{8}$$

and

$$P_2 = \frac{\hbar}{2} \int Z^{-1}[E + \hbar\,(\Delta_2 - \Delta_1)]^2 \\ \times (E + \hbar\,\Delta_2 - E_1^{sh})^2\Gamma_{2E}\,dE \\ + \frac{\hbar^3}{8} \int Z^{-1}[E + \hbar\,(\Delta_2 - \Delta_1)]^2\Gamma_{2E}^3\,dE, \tag{9}$$

where

$$Z = [E + \hbar\,(\Delta_2 - \Delta_1) - E_2^{sh}]^2 \\ [(E + \hbar\,\Delta_2 - E_1^{sh})^2 + (\hbar\,\Gamma_{2E}/2)^2]^2 \\ + \hbar^6\Gamma_{2E}^2 G_1^4/4]. \tag{10}$$

E_1^{sh} and E_2^{sh} are shift quantities whose expressions are given in the appendix. These quantities are usually small and can be neglected. We consider that the energy-dependence of the free-bound coupling is very weak and can be assumed to be constant which is calculated at some energy in the energy regime of interest near $\bar{E}$. This approximation is commonly known as slowly varying continuum approximation [52]. Now, for computational convenience we scale all the energy terms of equation (8) and equation (9) by G_1 and rewrite them as

$$P_1 = \int \frac{[(\epsilon + \delta_2)^2 + \beta^2]\beta}{(\epsilon + \delta_{12})^2 + [(\epsilon + \delta_2)^2 + \beta^2]^2 + \beta^2}\,d\epsilon \tag{11}$$

and

$$P_2 = \int \frac{(\epsilon + \delta_{12})^2[(\epsilon + \delta_2)^2 + \beta^2]\beta}{(\epsilon + \delta_{12})^2[(\epsilon + \delta_2)^2 + \beta^2]^2 + \beta^2}\,d\epsilon, \tag{12}$$

where $\delta_{12} = \hbar\,(\Delta_2 - \Delta_1)/G_1$, $\delta_2 = \hbar\,\Delta_2/G_1$, $\beta = |\Lambda_{2\bar{E}}|^2/\hbar G_1$. So, in terms of scaled parameters, the two-photon resonance condition is $\bar{\epsilon} + \delta_2 = 0$ where $\epsilon = E/G_1$. Now, for the $\epsilon \neq \bar{\epsilon}$, the system will be off-resonant and as a consequence there is no DS.

2.3. Adiabatic regime

In order to develop an intuitive understanding how the well-known discrete Λ configuration can be compared with Λ-like configuration with a continuum as far as DS and its application to STIRAP are concerned, we first recall the DS condition in the discrete Λ system. If the continuum state $|E\rangle$ in the inset of figure 1 is replaced by a bound state $|3\rangle$, it will be a three-level discrete Λ configuration for which the total Hamiltonian under RWA can be written as 3×3 matrix. On diagonalising the matrix, one obtains three eigenstates [24, 25]

$$|a^0\rangle = \cos\theta|3\rangle - \sin\theta|1\rangle \tag{13}$$

$$|a^+\rangle = \sin\phi\sin\theta|3\rangle + \cos\phi|2\rangle + \sin\phi\cos\theta|1\rangle \tag{14}$$

$$|a^-\rangle = \cos\phi\sin\theta|3\rangle - \sin\phi|2\rangle + \cos\phi\cos\theta|1\rangle \tag{15}$$

with eigenvalues $\alpha_0 = \Delta_2$, $\alpha_\pm = \frac{1}{2}[\Delta_2 \pm (\Delta_2^2 + G_1^2 + G_2^2)^{1/2}]$ respectively. The continuum-bound coupling is replaced by another bound–bound coupling parameter G_2. The angles θ and ϕ are defined as

$$\tan\theta = \frac{G_2}{G_1}, \tag{16}$$

$$\tan \phi = \frac{(G_1^2 + G_2^2)^{1/2}}{(\Delta_2^2 + G_1^2 + G_2^2)^{1/2} - \Delta_2}. \quad (17)$$

The degree of slowness or adiabaticity is assured when the rate of nonadiabatic coupling is small compared to the separation of the corresponding eigenvalues i.e

$$|\langle a^{\pm}|\frac{d}{dt}|a^0\rangle|^2 \ll |\alpha_0 - \alpha_{\pm}|^2 \quad (18)$$

which implies [24, 25]

$$\left|\langle a^+|\frac{d}{dt}|a^0\rangle\right|^2 + \left|\langle a^-\left|\frac{d}{dt}\right|a^0\rangle\right|^2 = \left|\frac{d\theta}{dt}\right|^2. \quad (19)$$

Thus, the adiabaticity is maintained when the rate of change of the mixing angle (θ) is sufficiently small, i.e when

$$|\dot{\theta}| \ll |\alpha_0 - \alpha_{\pm}|. \quad (20)$$

In the case of Λ-like configuration with a continuum, as mentioned earlier, a quasi-DS like situation can be achieved at ultra-cold temperature. We can set $\bar{E}$ to the most probable kinetic energy of a thermal distribution by adjusting $\Delta_1 - \Delta_2$. Since T is a measure of spread of the thermal energy distribution, all atoms having energy within $\bar{E} - k_b T/2$ to $\bar{E} + k_b T/2$ can be approximately said to be in a DS, k_b being the Boltzmann constant. Eventually, for this case, the energy difference between a DS and a bright state is $\sim k_b T$. Thus, the adiabaticity condition can be written as

$$\left|\frac{d\beta}{dt}\right| \ll k_b T/\hbar. \quad (21)$$

The condition equation (21) should be maintained in conformity with the energy- or temperature-dependence of the FC factor and $\bar{E} = 2k_b T = \hbar(\Delta_1 - \Delta_2)$. The free-bound coupling or free-bound FC factor is only significant at low energy. This means $T = \frac{\hbar}{2k_b}(\Delta_1 - \Delta_2)$ should be small enough so that FC factor is large. However, as we discuss in the next section, the rotational selection rule allows us to restrict the energy range for effective free-bound coupling.

3. Results and discussion

To select the two most suitable bound states of (LiCs)$^+$ system for our purpose, we first calculate the scattering-state and a large number of bound state wave functions by solving the partial wave Schrödinger equation given by

$$\left[\frac{d^2}{dr^2} + k^2 - \frac{2\mu}{\hbar^2}V(r) - \frac{\ell(\ell+1)}{r^2}\right]\psi_\ell(kr) = 0, \quad (22)$$

where r is the inter-nuclear separation of the ion and atom. k is the wave number related to collisional energy E and μ is the reduced mass of the ion–atom pair. $V(r)$ is the isotropic interaction potential between the ion and atom. The wave function $\psi_\ell(kr)$ has the asymptotic form $\psi_\ell(kr) \sim \sin[kr - \ell\pi/2 + \eta_\ell]$, η_ℓ being the phase shift for ℓth partial waves. We solve equation (22) numerically by standard re-normalized Numerov–Cooley method [53]. The

method involves calculation of wave function using a three-point recursion relation. The second derivatives are provided by the Schrödinger equation itself. To calculate a scattering state one has to start with an initially small internuclear separation r_0 and propagate it to asymptotically large separation r_a where the wave function corresponds to a free particle state. With the help of standard scattering boundary conditions, the asymptotic solutions are matched to deduce scattering S-matrix. By verifying the unitarity of the S-matrix, the correctness is justified. With the help of estimated theoretical behavior of the wave function at small r, one has to set the values of the wave function at two consecutive initial values of r. Then using the three-point recursion relation the value of wave function at the next (3rd) consecutive point is calculated. The 2nd and 3rd points are reset to initial points and by the same process the wave function is calculated at further next (4th) point. The process continues to propagate until the asymptotic boundary (r_a) is reached. The value of r_a can be calculated by setting the condition that the effective long range part of the potential, $V_{eff}(r) = -\frac{1}{2}\left(\frac{C_4}{r^4} + \frac{C_6}{r^6}\right) + \frac{\hbar^2}{2\mu}\ell(\ell+1)/r^2$ is much smaller than the collisional energy E (at least one tenth of E). For s-wave scattering the long-range part of potential is $-C_4/r^4$ for ion–atom pair, whereas, it is $-C_6/r^6$ for neutral atom pair. Thus, the value of r_a largely differs for the two systems. For higher partial waves, however, the dominating part of the long-range potential is the centrifugal barrier $\frac{\hbar^2}{2\mu}\ell(\ell+1)/r^2$ for both ion–atom and atom–atom systems and the value of r_a is same. For our system, we find that the value of r_a for the s-wave scattering has to be more than $4000a_0$ in order to get convergent results at microKelvin regime of collisional energy, where a_0 is the Bohr radius.

We have calculated the deeply bound states as well as bound states close to continuum of both the $V_1(r)$ and $V_2(r)$ potentials. To search for appropriate bound states for building our model, we have calculated 71 and 48 bound states of $V_1(r)$ and $V_2(r)$ potentials, respectively. To calculate the transition dipole elements between the continuum and the bound state or between the two bound states, we have used the transition dipole moment data of [51]. Molecular dipole transitions between two ro-vibrational states or between continuum and bound state is governed by FC principle. According to this principle, for excited vibrational (bound) states, bound–bound or continuum-bound transitions mainly occur near the turning points of bound states. In general, highly excited vibrational wave functions of diatomic molecules or molecular ions have their maximum amplitude near the outer turning points. Spectral intensity is proportional to the square of the FC overlap integral or FC factor. It implies that PA spectral intensity will be significant when the continuum state has a prominent anti-node near the separation at which outer turning point of the excited bound state lies. The value of free-bound FC factor for $v_2 = 44$ is found to be quite high since the maximum of the excited bound state wave function near the outer turning point coincides nearly with a prominent anti-node of the ground-state scattering wave
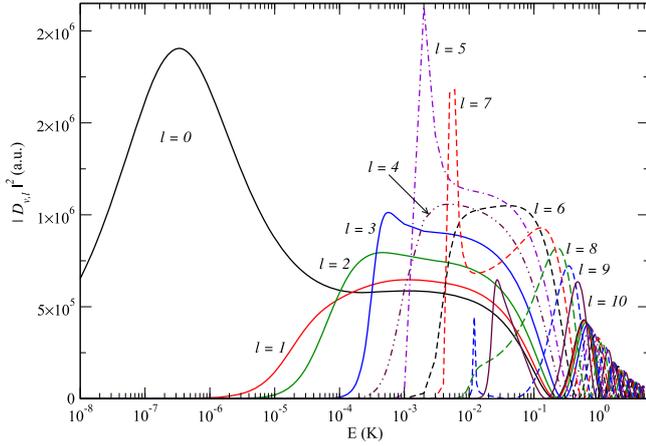
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Figure 2. The square of free-bound radial transition dipole moment in a.u for ground continuum sates with ℓ ranging from 0 to 10 and excited bound vibrational level with $v_2 = 44$ of $V_2(r)$ potential.

Figure 3. Solid line and dotted line represents the population of the final and intermediate state, respectively as a function of β for (LiCs)$^+$ ionic system. Here, $\beta = |\Lambda_{2\bar{E}}|^2/\hbar G_1$ is defined as the ratio of continuum-bound to bound–bound coupling.

function. We have also calculated the value of FC factor between the continuum of $V_1(r)$ potential and different bound states of the $V_2(r)$ potential and it is found to be very small.

Let $|\phi_{vJ}\rangle$ represents the ro-vibrational bound state in the potential $V_2(r)$ with vibrational and rotational quantum number v and J, respectively. The energy-normalized partial-wave scattering state in the potential $V_0(r)$ is denoted by $|\psi_{\ell,k}(r)\rangle$ where ℓ is the partial wave of the relative motion between the ion and the atom; and k is the wave number related to the collision energy by $E = \hbar^2 k^2/2\mu$ with μ being the reduced mass of the ion–atom pair. The free-bound transition dipole moment element $\mathcal{D}_{vJ,\ell}$ is defined by

$$\mathcal{D}_{vJ,\ell} = \langle\phi_{vJ}|\mathbf{D}(r) \cdot \mathbf{E}_{L_2}|\psi_{\ell,k}(r)\rangle = |E_{L_2}|^2 \eta_{J,\ell} D_{v,\ell}, \quad (23)$$

where $\mathbf{D}(r)$ is the molecular transition dipole moment that depends on the internuclear separation r, $\eta_{J,\ell}$ is an angular factor where $|\eta_{J,\ell}| \leqslant 1$.

$$D_{v,\ell} = \int dr \phi_{vJ}(r) D(r) \psi_{\ell,k}(r) \quad (24)$$

is the matrix element of radial part with $D(r)$ being the transition dipole moment, $\phi_{vJ}(r) = \langle r|\phi_{vJ}\rangle$ and $\psi_{\ell,k}(r) = \langle r|\psi_{\ell,k}\rangle$ represent the bound and the scattering wave functions. The quantity $D_{v,\ell}$ is weakly dependent on J and so we ignore the J-dependence of $D_{v,\ell}$. The photoassociative stimulated line width is given by $\Gamma(E) = 2\pi|\eta_{J,\ell}D_{v,\ell}|^2$. We plot the quantity $|D_{v,\ell}|^2$ as a function of energy E for a number of partial waves ℓ up to $\ell = 10$.

It is clear from figure 2 that the contributions to $|D_{v,\ell}|^2$ of higher partial waves are larger than that of s-wave for the energy above to 0.1 mK. At very lower energy ($E < 0.1$ mK), only s-wave makes finite contribution to the dipole transition. The higher partial-wave contributions in the sub-milliKelvin or milliKelvin regime can be attributed to the relatively stronger attractive nature of the long-range part of ion–atom interaction which goes as $\sim -1/r^4$ as $r \to \infty$. This is unlike for neutral atom–atom interaction that asymptotically goes as $\sim -1/r^6$.
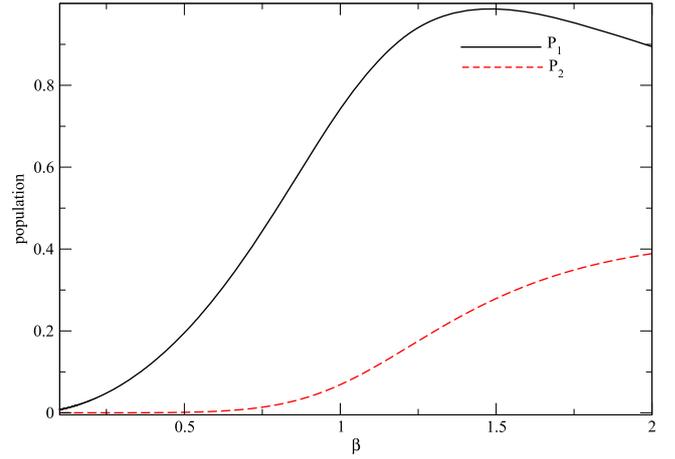
Therefore, compared to neutral atom–atom case where only a few low lying partial waves become important at such temperatures, a larger number of higher partial waves contribute significantly even in the sub-milliKelvin regime. Furthermore, the higher partial waves exhibit a prominent anti-node in the sub-milliKelvin or milliKelvin regime. For instance let us consider the contribution from $\ell = 3$. Its contribution is maximum at energy near 0.3 mK and $|D_{v,\ell=3}|^2$ remains more or less steady for energy ranging from 0.3 to 10 mK, but its contribution is negligible for energy lower than 0.1 mK.

For the sake of simplicity let us neglect the hyperfine interactions. This assumption can be well justified for our system. As per FC principle, PA transitions predominantly occur at the separation r near the outer turning point which is about 19 a_0 for $v_2 = 44$. At this separation the value of central potential C_4/r^4 is of the order of 1000 GHz. The hyperfine splitting in the ground-state of (LiCs)$^+$ is of the order of 100 MHz. So, in comparison to the central interaction, we can safely neglect hyperfine interaction. Since the total molecular angular momentum is given by $\mathbf{J} = \mathbf{S} + \mathbf{L} + \ell$, where $\mathbf{S}$ and $\mathbf{L}$ are the total electronic spin and orbital quantum number, respectively. In our model $\mathbf{L} = 0$ and $\mathbf{S} = \frac{1}{2}$ for ground-state potential $V_0(r)$. Now, if we tune the PA laser to the near-resonance for ro-vibrational state $\left(v = 44, J = \frac{9}{2}\right)$ then only $\ell = 3$ will be coupled by the PA laser due to selection rule $\Delta J = \pm 1$. Now, if we choose $\bar{E} = 0.3$ milliKelvin and accordingly adjust the parameters Δ_1 and Δ_2, then it is expected that the ion–atom pair will be in a quasi-DS for a range of collision energies near $\bar{E}$. For a thermal system of ion–atom mixture, we need to set the temperature $k_b T \simeq \bar{E}$ in order to bring a finite fraction of the atom–ion pairs in the quasi-DS. Now, under these conditions, if we slowly vary the ratio $\beta = |\Lambda_{2\bar{E}}|^2/\hbar G_1$ subject to the condition of equation (21), then we can accomplish an almost STIRAP process for transfer of cold atom–ion pairs into cold ionic molecules.

Table 1. Rabi-frequency calculated between vibrational level $v_2 = 44$ and different vibrational levels v_1 of the potential $V_1(r)$ for the intensity of laser L_1 being 1 W cm^{-2}.

v_2	v_1	G_1(MHz)
	9	3.269
	10	3.649
44	11	3.176
	15	2.612
	18	2.374

In figure 3, we have shown the variation of P_1 and P_2 with β. Although, the final state probability approaches unity ($P_1 \simeq 1$) for all value of β, there is a significant probability of the intermediate state ($P_2 \simeq 0.4$). Hence possibility of formation of DS is very low for higher values of β. But in the lower range of β, we find that the probability of intermediate state is negligibly small whereas that of probability of final state rises gradually. So in this region where the value of G_1 is much higher, the system remains almost in the DS and population transfer efficiency is about 50%. Finally, the formation of meta-stable molecular ion takes place in $V_1(r)$ state. This meta-stable molecular ion will have longer lifetime. Therefore, the formation of ground-state molecular ion becomes more likely due to favorable level structure. To find coherent laser coupling between the two selected bound states, we have calculated Rabi frequency G_1 which is expressed as

$$\hbar G_1 = \left(\frac{I_{L_1}}{4\pi\epsilon_0 c} \right)^{\frac{1}{2}} \langle \phi_{vj} | \mathbf{D}(r) \cdot \hat{\epsilon} | \phi_{v'j'} \rangle |, \tag{25}$$

where $\hat{\epsilon}$ is the unit vector of laser polarization and $|\phi_{vj}\rangle$ and $|\phi_{v'j'}\rangle$ are the two bound states. In table 1, we have shown the calculated Rabi frequency between the different bound levels of $V_1(r)$ potential and bound state ($v = 44$) of $V_2(r)$ potential for laser intensity $I_{L_1} = 1$ W cm^{-2} of laser L_1. From the table 1, the maximum Rabi frequency corresponding to the bound–bound transition is found to be 3.649 MHz. Comparing this value with the calculated spontaneous line width $\gamma = 26$ MHz of the excited bound state of $V_2(r)$ potential by using standard formula [10], we infer that the lifetime of the photoassociated molecule in this excited molecular bound state is very small. Hence, it is difficult to form the molecular ion by two-photon incoherent Raman PA.

Now we discuss the possibility of the formation of ground-state molecular ion by three photon process. From the above discussion we find that the excited molecular bound states in $V_2(r)$ potential are lossy in nature. Here we use a two-photon scheme by which the contribution of this lossy state is effectively minimized. The energy dependence of $\Gamma_{2\bar{E}}$ arises from the FC factor. Now, taking the value of $\Gamma_{2\bar{E}} = 1.43$ MHz that corresponds to FC factor at an energy near 0.3 mK and intensity 6 W cm^{-2} of L_2. In our calculation, we vary G_1 from 0.01 MHz to 10 MHz which corresponds to the variation of β from 143 to 0.143. Adiabatic condition will

be maintained if the time rate of change of β is kept much less than 6.3 s^{-1}.

4. Conclusions

In conclusion, we have shown that for a continuum–bound–bound photoassociative ion–atom system resembling Λ-type configuration, creation of a quasi-DS at low energy is possible by driving stimulated Raman transitions with two lasers under certain physical conditions. By making use of this state, the initial population in the continuum of ion–atom scattering states can be partially and coherently transferred to a long-lived molecular bound state via an intermediate excited bound state in a STIRAP-like fashion. We have illustrated our proposed method by numerical simulation with a realistic model system of $(LiCs)^+$ system. We have developed our treatment based on Fano's method of diagonalisation of a continuum-bound coupled system. The quasi-DS is a dressed continuum which is an admixture of a long-lived bound state and the ground-state continuum. The DS becomes decoupled from the lossy state only for a particular collision energy. Therefore, complete population transfer is, in general, not possible in a continuum-bound system in thermal equilibrium. However, by a proper choice of temperature, laser detunings and rotational levels of the lossy state, one can optimize the population transfer. Since the PA transition in our system is strongly dipole allowed, it has been possible to obtain appreciable PA coupling at a relatively low laser intensity. However, for systems with weakly dipole allowed PA transitions, one can use the method of Feshbach resonance induced Fano effect in PA [54] to enhance or modify PA coupling [55] or to achieve Feshbach optimized PA [56] at low laser intensities. As a further study, one can consider two-photon Raman process involving a magnetic Feshbach-resonance induced structured continuum [57]. In such physical situations, it is possible to create a bound state in continuum that can be effectively decoupled from the continuum [58]. This opens up the possibility of creating an effective Λ system of three bound states involving an underlying continuum. Whether complete or more efficient coherent population transfer in such an effective Λ system is possible or not would be an interesting problem to pursue in future.

Acknowledgments

Appendix. Dressed continuum

We diagonalise the hamiltonian H given in equation (1) and equation (2) in two steps using Fano's method. First, we diagonalise the sub-system comprising the bare continuum of states $|E\rangle$ and the bound state $|2\rangle$ which are coupled by laser pulse L_2. At this stage, we define an intermediate dressed state $|E\rangle_{\mathrm{idr}}$ as

$$|E\rangle_{\mathrm{idr}} = A_{2E}|2\rangle + \int C_E(E')|E'\rangle\,\mathrm{d}E', \qquad (A.1)$$

where $A_{2E} = \frac{\Lambda_{2E}}{E + \hbar\Delta_2 - E_1^{\mathrm{sh}} - i\hbar\Gamma_{2E}/2}$, $C_E(E') = \frac{A_{2E}\Lambda_{E'2}}{E - E'} + \delta(E - E')$ and $\Gamma_{2E} = \frac{2\pi\,|\Lambda_{2E}|^2}{\hbar}$ is the energy-dependent stimulated line width for the free-bound PA transition. $E_1^{\mathrm{sh}} = \mathcal{P}\int\frac{|\Lambda_{2E'}|^2\,\mathrm{d}E'}{E - E'}$ is the light shift of the bound state, where $\mathcal{P}$ represents the Cauchy Principle value integral. In terms of

$|E\rangle_{\mathrm{idr}}$, the hamiltonian can be written as

$$H = \hbar(\Delta_1 - \Delta_2)|1\rangle\langle 1| + \int |E\rangle_{\mathrm{idr\,idr}}\langle E|\,\mathrm{d}E \\ + (\hbar G_1|1\rangle\langle 2| + \mathrm{H.c.}). \qquad (A.2)$$

Using equation (A.1) and the completeness relation

$$|1\rangle\langle 1| + \int |E\rangle_{\mathrm{idr\,idr}}\langle E|\,\mathrm{d}E = 1 \qquad (A.3)$$

we can write

$$|2\rangle = \left(|1\rangle\langle 1| + \int |E\rangle_{\mathrm{idr\,idr}}\langle E|\,\mathrm{d}E\right)|2\rangle = \int A_{2E}^*|E\rangle_{\mathrm{idr}}\,\mathrm{d}E. \qquad (A.4)$$

Substituting equation (A.4) into equation (A.2) we get

$$H = \hbar(\Delta_1 - \Delta_2)|1\rangle\langle 1| + \int |E\rangle_{\mathrm{idr\,idr}}\langle E|\,\mathrm{d}E \\ + \left(\int \Lambda_{E1}|E\rangle_{\mathrm{idr}}\langle 1|\,\mathrm{d}E + \mathrm{H.c.}\right). \qquad (A.5)$$

The above Hamiltonian describes the one bound state $|1\rangle$ coupled to $|E\rangle_{\mathrm{idr}}$ by an effective coupling parameter $\Lambda_{1E} = \hbar G_1 A_{2E}$. In the second and final step we diagonalise

A.5. We define the final dressed state as

$$|E\rangle_{\mathrm{dr}} = B_{1E}|1\rangle + \int D_E(E'')|E''\rangle_{\mathrm{idr}}\,\mathrm{d}E'' \\ = B_{1E}|1\rangle + \int D_E(E'') \\ \times \left(A_{2E''}|2\rangle + \int C_{E''}(E')|E'\rangle\,\mathrm{d}E'\right)\mathrm{d}E'' \\ = B_{1E}|1\rangle + B_{2E}|2\rangle + \int F_E(E')|E'\rangle. \qquad (A.6)$$

The expression for B_{1E} is

$$B_{1E} = \frac{\Lambda_{1E}}{E + \hbar(\Delta_2 - \Delta_1) - E_2^{\mathrm{sh}} 215203 - i\hbar\Gamma_{1E}/2}, \qquad (A.7)$$

where $\Gamma_{1E} = 2\pi\hbar|G_1|^2|A_{2E}|^2$ is the effective stimulated linewidth of $|1\rangle$ and $E_2^{\mathrm{sh}} = \mathcal{P}\int\frac{|\Lambda_{2E}|^2\,\mathrm{d}E}{[(E + \hbar\Delta_2 - E_1^{\mathrm{sh}})^2 + \hbar^2\Gamma_{2E}^2/4](E - E')}$ is the laser induced shift of the same. Using the expression of A_{2E} as given above, the explicit form of B_{1E} is

$$B_{1E} = \frac{\hbar G_1\Lambda_{2E}(E + \hbar\Delta_2 - E_1^{\mathrm{sh}} + i\hbar\Gamma_{2E}/2)}{[E + \hbar(\Delta_2 - \Delta_1) - E_2^{\mathrm{sh}}]\left[(E + \hbar\Delta_2 - E_1^{\mathrm{sh}})^2 + \frac{\hbar^2\Gamma_{2E}^2}{4}\right] - \frac{i}{2}\hbar^3 G_1^2\Gamma_{2E}}. \qquad (A.8)$$

The other terms are $D_E(E'') = \frac{B_{1E}\Lambda_{E''1}}{E - E''} + \delta(E - E'')$ and $B_{2E} = \int D_E(E'')A_{2E''}\,\mathrm{d}E''$. The explicit form of B_{2E} can be written as

$$B_{2E} = \frac{[E + \hbar(\Delta_2 - \Delta_1)][E + \hbar\Delta_2 - E_1^{\mathrm{sh}} - i\hbar\Gamma_{2E}/2]\Lambda_{2E}}{[E + \hbar(\Delta_2 - \Delta_1) - E_2^{\mathrm{sh}}][(E + \hbar\Delta_2 - E_1^{\mathrm{sh}})^2 + (\hbar\Gamma_{2E}/2)^2] + i\hbar^3\Gamma_{2E}G_1^2/2}. \qquad (A.9)$$

Here $F_E(E') = \int D_E(E'')C_{E''}(E')\,\mathrm{d}E''$. Using equation (A.8) and equation (A.9) we finally obtain the expressions for P_1 and P_2 as

$$P_1 = \int |B_{1E}|^2\,\mathrm{d}E = \frac{\hbar^3}{2}|G_1|^2\int Z^{-1}\Gamma_{2E}(E + \hbar\Delta_2 - E_1^{sh})^2\,\mathrm{d}E \\ + \frac{\hbar^5}{8}|G_1|^2\int Z^{-1}\Gamma_{2E}^3\,\mathrm{d}E \qquad (A.10)$$

and

$$P_2 = \int |B_{2E}|^2\,\mathrm{d}E = \frac{\hbar}{2}\int Z^{-1}[E + \hbar(\Delta_2 - \Delta_1)]^2 \\ \times (E + \hbar\Delta_2 - E_1^{sh})^2\Gamma_{2E}\,\mathrm{d}E \qquad (A.11) \\ + \frac{\hbar^3}{8}\int Z^{-1}[E + \hbar(\Delta_2 - \Delta_1)]^2\Gamma_{2E}^3\,\mathrm{d}E,$$

where

$$Z = [E + \hbar (\Delta_2 - \Delta_1) - E_2^{\text{sh}}]^2$$
$$\times [(E + \hbar \Delta_2 - E_1^{\text{sh}})^2 + (\hbar \Gamma_{2E}/2)^2]^2$$
$$\times \hbar^6 \Gamma_{2E}^2 G_1^4/4]. \tag{A.12}$$

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