Efficient formation of ground-state ultracold molecules via STIRAP from the continuum at a Feshbach resonance

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Abstract. We develop a theoretical description of photoassociative stimulated Raman adiabatic passage (STIRAP) near a Feshbach resonance in a thermal atomic gas. We show that it is possible to use low-intensity laser pulses to directly excite pairs of atoms in the continuum near a Feshbach resonance and to transfer most of the atomic cloud to the lowest rovibrational level of the molecular ground state. For a broad resonance, commonly found in several mixtures of alkali atoms, our model predicts a transfer efficiency up to 97\% for a given atom pair, and up to 70\% when averaged over an atomic ensemble. The laser intensities and pulse durations needed for such an optimal transfer, $10^2$–$10^3$ W cm$^{-2}$ and several microseconds, are easily achievable experimentally. A single pair of STIRAP pulses converts an estimated fraction $f \sim 10^{-6}$--$10^{-4}$ of atoms in an atomic ensemble, leading to the production of 10–1000 molecules in a large sample of $10^7$ atoms. A total of $\sim 10^4$–$10^6$ pulse pairs are thus required to transfer most atoms into molecules. Such an efficiency compares with or surpasses currently available techniques for creating stable diatomic molecules, and the versatility of this approach simplifies its potential use for many molecular species.
1. Introduction

The realization of rovibrationally stable dense samples of ultracold diatomic molecules remains one of the major goals in the field of atomic and molecular physics. Though obtaining diatomic alkali molecules was seen as a logical next step following the optical cooling of atoms, many of the possible applications currently under investigation extend beyond atomic and molecular physics. Testing fundamental symmetries based on high-precision spectroscopy of ultracold molecules [1]–[3] or the attempts to detect the time variation of fundamental constants [4] are examples of such applications. Another one is ultracold chemistry, where controlling reactive processes could be accomplished using coherent quantum superpositions of interacting species and products [5]. Important insights about new phases of matter could be gained from the strong anisotropic dipole–dipole interaction between ultracold dipolar molecules [6]. Finally, ultracold polar molecules could also represent an attractive platform for quantum computation [7, 8]. Many of those applications require dense samples of ultracold polar molecules in the lowest rovibrational state that makes them collisionally stable and long-lived.

Translationally ultracold (100 nK–1 mK) molecules are produced from an ultracold atomic gas by photoassociation (PA) [9] or magnetoassociation (MA) [10]. In a typical PA scheme, a pair of colliding atoms is photoassociated into a bound electronically excited molecular state that spontaneously decays, forming molecules in the electronic ground state. In MA, a magnetic field is adiabatically swept across a Feshbach resonance, converting two atoms from a scattering state into a bound molecular state. Both techniques produce weakly bound molecules in highly excited vibrational states of the ground electronic potential. Such molecules have to be rapidly transferred to deeply bound vibrational states before they are lost from the trap due to inelastic collisions.

Stimulated Raman adiabatic passage (STIRAP) [11] has recently attracted significant interest as an efficient way of producing deeply bound molecules, starting from Feshbach molecules [12, 13]. It allows us to realize high transfer efficiency and preserve the high phase-space density of an initial atomic gas. In STIRAP, the laser pulses, coupling an initial and a final state to an intermediate excited state, are applied in a counter-intuitive sequence where a pump pulse is preceded by a Stokes pulse. During the transfer, the system stays in a ‘dark’ state, i.e. a coherent superposition of initial and final states, preventing any losses that would otherwise occur from the excited state. By adiabatically changing the amplitudes of the laser...
pulses, the ‘dark’ state evolves from the initial to the final state, resulting in nearly 100% transfer efficiency [11].

Feshbach molecules are, however, usually short-lived because of inelastic collisions with background atoms or other molecules. This is especially true for those produced from bosonic or mixed bosonic/fermionic atoms, for which collisions are not suppressed by the Fermi statistics at ultralow temperatures. In a dense atomic gas of density $n_{at} \approx 10^{13} – 10^{14}$ cm$^{-3}$, the collisional decay rate can be up to $\sim 10^4$ s$^{-1}$ (with inelastic rate coefficient $K_{\text{inel}} \sim 0.5 – 1.0 \times 10^{-10}$ cm$^3$ s$^{-1}$ for Feshbach molecules [14, 15]). The STIRAP transfer must therefore be fast enough to avoid losing molecules by inelastic decay. To alleviate this problem, we propose to start the STIRAP process directly from the scattering continuum without first forming Feshbach molecules. Using this approach with many STIRAP pulses and a fast repetition rate would also allow the conversion of nearly an entire atomic ensemble into ground state molecules, not only those atoms that were first transferred to a Feshbach molecular state.

Efficient adiabatic passage from the continuum requires laser pulses shorter than the coherence time of the continuum [16]–[18]. The adiabaticity condition of STIRAP, $\Omega T \gg 1$, where $T$ is the transfer time, therefore implies a large effective Rabi frequency $\Omega$ for the pulses. In addition, dipole matrix elements between the continuum and the bound state are usually small, and so the pump pulse that couples the continuum and the excited state would require a very high intensity, which proves impractical. Thus the previous STIRAP experiments [12], being restricted by the very short coherence time of the continuum, used a Feshbach molecular state as an initial state.

The small continuum-bound dipole matrix elements can be dramatically increased by photoassociating atoms in the vicinity of a Feshbach resonance. It has been shown, both theoretically and experimentally, that the PA rate increases in the presence of a Feshbach resonance by several orders of magnitude [19]–[22]. This can be explained by considering that delocalized scattering states acquire some bound-state character due to admixture of a bound level associated with a closed channel, resulting in a large increase of the Franck–Condon factor between the initial scattering state and the final excited state. The recently proposed Feshbach optimized photoassociation (FOPA) technique [22] relies on this enhancement to directly reach deeply bound ground state vibrational levels from the scattering continuum. Consequently, photoassociation in the vicinity of a Feshbach resonance is expected to increase the molecular formation rate up to $10^6$ molecules s$^{-1}$ [22].

In the present work, we combine the approach used in FOPA with STIRAP for reducing the required pulse intensity. We predict highly efficient transfer of most of an atomic ensemble into the lowest rovibrational level in the molecular ground state. We note that a proposal, namely the admixing of a short-range potential to a longer range excited electronic potential, was recently suggested for improving a two-color pump–dump photoassociation scheme [23]. The scattering continuum states have good overlap with the long-range potential, while the admixed short-range potential provides a good overlap with tightly bound vibrational levels of the ground electronic state, greatly improving the efficiency of photoassociation.

We have organized the paper as follows. In section 2, we derive a theoretical model of a combined atomic and molecular system. Fano theory is used to describe the interaction of a bound molecular state with the scattering continuum, represented as closed and open channels, respectively. The resulting continuum states are coupled by two laser fields to the vibrational target state in the ground state via the intermediate excited molecular electronic vibrational state. Next, we describe STIRAP-assisted conversion of a pair of colliding atoms into a deeply bound
molecule. In section 3, we present the results of numerical solutions of the model described in section 2 and, in the appendix, using typical parameters of alkali dimers. We find optimal Rabi frequencies and profiles of STIRAP pulses. In section 4, we average the pair-of-atoms STIRAP transfer efficiency over a thermal atomic ensemble, calculate a fraction of atoms that can be converted into molecules by one STIRAP sequence and the number of pulses and overall time required to convert a large fraction of a cloud of atoms into molecules. Finally, we conclude in section 5.

2. Model

We consider a three-level system plus a continuum as shown in figure 1, representing scattering states of two colliding atoms and bound states of a molecule. The ground level labeled $|1\rangle$ is the final product state to which a maximum of population must be transferred. Typically, this level will be the lowest vibrational level ($v'' = 0, J'' = 0$) of a ground molecular potential. This ground level is coupled to an excited bound level $|2\rangle$ of an excited molecular potential via a ‘Stokes’ pulse depicted by the blue down-arrow in figure 1. This level $|2\rangle$ is itself coupled via a pump pulse (red up-arrow) to an initial continuum of unbound scattering states $|\Psi_\epsilon\rangle$ of energies $\epsilon$ (grey area in figure 1). If we denote by $C_1(t)$, $C_2(t)$ and $C(\epsilon, t)$ the time-dependent amplitudes associated with the final, intermediate and initial states $|1\rangle$, $|2\rangle$ and $|\Psi_\epsilon\rangle$, respectively, then the total wave function $|\Phi\rangle$ of the system is given by

$$|\Phi\rangle = C_1(t)|1\rangle + C_2(t)|2\rangle + \int d\epsilon \ C(\epsilon, t)|\Psi_\epsilon\rangle.$$ (1)
We assume that the levels associated with states $|1\rangle$ and $|2\rangle$ are well isolated and that there are no off-resonant laser couplings to other levels: this ensures sufficient accuracy of the three-state model (see e.g. [12, 13]).

No restriction applies to the definition of the continuum state $|\Psi_\epsilon\rangle$ as it can be associated with either a single-channel or a multichannel scattering state. In this work, we consider the multichannel case in which a bound level $|b\rangle$ associated with a closed channel is embedded in the continuum of scattering states $|\epsilon\rangle$ of an open channel. When the energy of $|\epsilon\rangle$ coincides with that of $|b\rangle$, a Feshbach resonance [24] occurs. These are common in binary collisions of alkali atoms due to hyperfine mixing and the tuning of the Zeeman interaction by an external magnetic field, hence the possibility of controlling interatomic interactions with a magnetic field. Following the Fano theory presented in [25], the scattering state $|\Psi_\epsilon\rangle$ can be expressed as

$$|\Psi_\epsilon\rangle = a(\epsilon)|b\rangle + \int d\epsilon' b(\epsilon, \epsilon')|\epsilon\rangle,$$

(2)

with

$$a(\epsilon) = \sqrt{\frac{2}{\pi \Gamma(\epsilon)}} \sin \Delta$$

(3)

and

$$b(\epsilon, \epsilon') = \frac{1}{\pi} \sqrt{\frac{\Gamma(\epsilon')}{\Gamma(\epsilon)}} \frac{\sin \Delta}{\epsilon - \epsilon'} - \cos \Delta \delta(\epsilon - \epsilon').$$

(4)

Here, $\Delta = -\arctan(\frac{\Gamma(\epsilon')}{\Gamma(\epsilon) - \epsilon'})$ is the phase shift due to the interaction between $|b\rangle$ and the scattering state $|\epsilon\rangle$ of the open channel. We assume $\Delta \in [-\pi/2, \pi/2]$. The width of the Feshbach resonance, $\gamma = 2\pi |V(\epsilon)|^2$, is weakly dependent on the energy, while $V(\epsilon)$ is the interaction strength between the open and closed channels. The position of the resonance, $\epsilon_F = E_b + P \int \frac{|V(\epsilon)|^2 d\epsilon'}{\epsilon - \epsilon'}$, includes an interaction-induced shift from the energy of the bound state $E_b$.

If we label $E_i$ the energy of the state $|i\rangle$, the total Hamiltonian $H$ is given by

$$H = \sum_{i=1,2} E_i |i\rangle \langle i| + \int d\epsilon |\Psi_\epsilon\rangle \langle \Psi_\epsilon| + V_{\text{light}}.$$

(5)

The light–matter interaction Hamiltonian $V_{\text{light}}$ takes the form

$$V_{\text{light}} = -\left[\vec{\mu}_{21} |2\rangle \langle 1| + \text{H.c.}\right] \left(\vec{\xi}_p + \vec{\xi}_S + \text{c.c.}\right) - \int d\epsilon \left[\vec{\mu}_{2\Psi} |2\rangle \langle \Psi_\epsilon| + \text{H.c.}\right] \cdot \left(\vec{\xi}_p + \vec{\xi}_S + \text{c.c.}\right),$$

(6)

where $\vec{\xi}_{p,S} = \hat{e}_{p,S} e_p \exp(-i\omega_{p,S} t)$ are the pump and Stokes laser fields of polarization $\hat{e}_{p,S}$, respectively, while $\vec{\mu}_{21}$ and $\vec{\mu}_{2\Psi}$ are the dipole transition moments between the states $|2\rangle$ and $|1\rangle$, and $|2\rangle$ and $|\Psi_\epsilon\rangle$, respectively. In this form, the Hamiltonian already takes into account the mixing between the bound state of the closed channel and the scattering states of the open channel. The Schrödinger equation describing the STIRAP conversion of two atoms into a molecule gives

$$i\hbar \frac{\partial C_1}{\partial t} = E_1 C_1 - \vec{\mu}_{21} \cdot \vec{\xi}_S C_2,$$

(7)

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Equations (the rotating wave approximation with the electronic potential with respect to the state of the bound states: New Journal of Physics where we introduced a spontaneous decay term \( \gamma \). Equations (7)–(9) then become

\[
i \hbar \frac{\partial C_1}{\partial t} = -\Omega_S c_2,
\]

\[
i \hbar \frac{\partial C_2}{\partial t} = \delta c_2 - \Omega_S c_1 - \int_{\epsilon_{th}}^{\infty} d\epsilon \, \Omega_\epsilon c(\epsilon, t),
\]

\[
i \hbar \frac{\partial c(\epsilon, t)}{\partial t} = \Delta_\epsilon c(\epsilon, t) - \Omega_\epsilon^* c_2,
\]

where \( \delta = E_2/\hbar - \omega_S \), \( \Delta_\epsilon = \epsilon/\hbar - (\omega_S - \omega_P) \) and \( \epsilon_{th} \) is the dissociation energy of the ground electronic potential with respect to the state \( |1\rangle \). The Rabi frequencies of the fields are \( \Omega_S = \vec{\mu}_{21} \cdot \vec{E}_S/\hbar \) (assumed real), \( \Omega_\epsilon = \vec{\mu}_{2\psi_\epsilon} \cdot \vec{E}_P/\hbar \).

The previous system of three equations can be reduced into a two-equation system by eliminating the continuum amplitude \( c(\epsilon, t) \) in equation (12). Introducing a solution in the form of \( c(\epsilon, t) = s(\epsilon, t) \exp(-i\Delta_\epsilon t) \) into equation (12), we get

\[
s = i \int_0^t dt' \Omega_\epsilon^* (t') c_2(t') e^{i\Delta_\epsilon t'} + s(\epsilon, t = 0),
\]

where \( t = 0 \) is some moment before the collision of the two atoms. The resulting continuum amplitude is

\[
c(\epsilon, t) = i \int_0^t dt' \Omega_\epsilon^* (t') c_2(t') e^{i\Delta_\epsilon (t'-t)} + s(\epsilon, t = 0) e^{-i\Delta_\epsilon t}.
\]

Inserting this result into equation (11), we obtain a final system of equations for the amplitudes of the bound states:

\[
i \hbar \frac{\partial c_1}{\partial t} = -\Omega_S c_2,
\]

\[
i \hbar \frac{\partial c_2}{\partial t} = (\delta - i\gamma) c_2 - \Omega_S c_1 - \int_{\epsilon_{th}}^{\infty} d\epsilon \Omega_\epsilon (t) s(\epsilon, t = 0) e^{-i\Delta_\epsilon t} + i \int_{\epsilon_{th}}^{\infty} d\epsilon \Omega_\epsilon (t) \int_0^t dt' \Omega_\epsilon^* (t') c_2(t') e^{i\Delta_\epsilon (t'-t)}
\]

\[
\equiv (\delta - i\gamma) c_2 - \Omega_S c_1 - S + T,
\]

where we introduced a spontaneous decay term \( \gamma c_2 \) in equation (16).

The third term of equation (16), labeled \( S \), corresponds to the source function, whereas the last term, labeled \( T \), corresponds to the ‘back-stimulation’ term (or back-conversion), which accounts for the transfer of the bound molecules back into the continuum. The initial amplitude of the continuum wave function \( s(\epsilon, t = 0) \) appearing in the source term has been discussed in
is given by

\[ s(\epsilon, t = 0) = \frac{1}{(\pi \delta_c^2)^{1/4}} \exp \left[ -\frac{(\epsilon - \epsilon_0)^2}{2\delta_c^2} + \frac{i}{\hbar} (\epsilon - \epsilon_0)t_0 \right], \tag{17} \]

where \( t_0 \) is the moment of the collision and \( \epsilon_0 \) is the central energy of the wavepacket.

Furthermore, the Rabi frequency of the field coupling continuum states \( |\Psi_\epsilon\rangle \) to the state \( |2\rangle \) is given by \[ \Omega_\epsilon = \frac{\vec{\mu}_{2\epsilon} \cdot \hat{e}_p \epsilon_p}{\hbar} \frac{g}{\sqrt{(\Gamma/2)^2 + (\epsilon - \epsilon_F)^2}} \text{sgn}(\epsilon - \epsilon_F), \tag{18} \]

where \( \vec{\mu}_{2\epsilon} \) is the dipole matrix element between an unperturbed scattering state \( |\epsilon\rangle \) and the state \( |2\rangle \), and \( g \) is the Fano parameter, expressed as

\[ g = \frac{(\vec{\mu}_{2\epsilon} \cdot \hat{e}_p) + P \int \frac{V(\epsilon)|\vec{\mu}_{2\epsilon} \cdot \hat{e}_p|\epsilon}{\epsilon - \epsilon_F} \, d\epsilon}{\pi V^2(\epsilon)|\vec{\mu}_{2\epsilon} \cdot \hat{e}_p|^2}, \tag{19} \]

where \( \hat{e}_p \) is the polarization vector of the pump field, and \( \vec{\mu}_{2b} \) is the dipole matrix element between bound states \( |2\rangle \) and \( |b\rangle \). The \( g \) factor is essentially the ratio of the dipole matrix elements from state \( |2\rangle \) to the bound state \( |b\rangle \) (modified by the continuum) and to an unperturbed continuum state \( |\epsilon\rangle \). This factor can be made much larger than unity, and as will be shown below, the total dipole matrix element from the continuum can be enhanced by this factor in the presence of the resonance. The magnitude of \( g \) can be controlled by the choice of the vibrational state \( |2\rangle \). Selecting a tightly bound excited vibrational state will increase the bound–bound and decrease the continuum-bound dipole matrix elements, resulting in a larger \( g \), whereas choosing a highly excited state close to a dissociation threshold will decrease \( g \).

Using the expressions given in equations (17)–(19) for the initial amplitude of the continuum wavefunction, the Rabi frequency between the continuum state \( |\Psi_\epsilon\rangle \) and the excited bound state \( |2\rangle \), and the Fano parameter, respectively, we obtain the following complete expression for the source term:

\[ S = S_0 \int_{\epsilon_{th}}^{\infty} d\epsilon \, g(\epsilon, \epsilon) \text{sgn}(\epsilon - \epsilon_F) \exp \left[ -\frac{(\epsilon - \epsilon_0)^2}{2\delta_c^2} + \frac{i(\epsilon - \epsilon_0)t_0}{\hbar} \right] e^{-i\Delta_c \tau}, \tag{20} \]

with \( S_0 = \frac{\vec{\mu}_{2\epsilon} \cdot \hat{e}_p \epsilon_p}{\hbar}(\pi \delta_c)^{1/4} \), and where the function \( g(\epsilon, \epsilon) \) is defined as

\[ g(\epsilon, \epsilon) = \frac{q + (2/\Gamma)(\epsilon - \epsilon_F)}{\sqrt{1 + (4/\Gamma^2)(\epsilon - \epsilon_F)^2}}. \tag{21} \]

We assume that the unperturbed continuum is structureless and that the corresponding Rabi frequency \( \vec{\mu}_{2\epsilon} \cdot \hat{e}_p \epsilon_p / \hbar \) depends only weakly on the energy. We also extend \( \epsilon_{th} \) to \(-\infty\) to have the initial continuum wavefunction normalized to unity\footnote{Extension of \( \epsilon_{th} \) to \(-\infty\) in the source term (20) can be justified by the sharp reduction of the Gaussian term \( \exp(-((\epsilon_{th} - \epsilon_0)^2/2\delta_c^2)) \) for \( \epsilon_0 - \epsilon_{th} > \delta_c \). For \( \epsilon_0 \) close to \( \epsilon_{th} \), this approximation is less accurate, but as will be shown in section 4, these low energies give negligible contribution to transfer efficiency averaged over an atomic ensemble due to their small weight in the Maxwell–Boltzmann distribution.}: \[ \int_{-\infty}^{\infty} d\epsilon |C(\epsilon)|^2 = 1. \]
We can as well obtain a complete expression for the back-stimulation term \( T \). We have

\[
T = \left\| \frac{\hat{\mu}_{2e} \hat{c}_p}{\hbar} \right\|^2 E_p(t) \int_{\epsilon_{th}}^{\infty} d\epsilon \, g^2(\epsilon, \epsilon) \int_0^t dt' c_2(t') E_p(t') e^{i\Delta_p(t'-t)}.
\]  

Extending the lower integration limit\(^5\) allows for an analytical solution for the integrals over energy and time, leading to the following expression for the back-stimulation term:

\[
T = \left\| \frac{\hat{\mu}_{2e} \hat{c}_p}{\hbar} \right\|^2 \left[ \pi \hbar E_p^2(t) c_2(t) + \frac{\pi \Gamma}{2} (q - \tau)^2 E_p(t) \int_0^t dt' c_2(t') E_p(t') e^{i\Delta_p/(\pi/2h)} \right].
\]

3. Results of STIRAP transfer for a pair of atoms

In this work, we consider two different cases: first, when \( \Gamma \gg \delta_e \), i.e. when the width \( \Gamma \) of the Feshbach resonance is much larger than the thermal energy spread \( \delta_e \) of the colliding atoms, and second, when \( \Gamma \ll \delta_e \). By considering these two limiting cases of broad and narrow resonances, more practical expressions for both the source term \( S \) and the back-stimulation term \( T \) can be found. The derivation of the final system of equations used in numerical solutions is given in appendix \( A \). Here, we describe the solutions of these systems for both broad and narrow resonances.

We note that during the transfer an initial incoherent mixture of atomic scattering states is converted into a pure internal state, which seems to decrease the entropy of the system. However, the entropy is transferred to the center-of-mass motion of the created molecules, which can lead to a slight translational heating of the sample.

Using equations (A.3)–(A.4) and (A.7)–(A.8) with the parameters listed in table 1 for a broad (\( \Gamma = 1 \) mK) and a narrow (\( \Gamma = 1 \) \( \mu \)K) Feshbach resonance, we obtain the results for the STIRAP transfer of an atom pair, depicted in figure 2. Here the left column corresponds to the broad resonance, and the right column to the narrow resonance. The top row shows the variation of the Rabi frequencies over the time period required for the population transfer along with population in the intermediate state \( |2\rangle \) (middle row) and final state \( |1\rangle \) (bottom row).

For the broad case, we considered a Feshbach resonance with a width \( \Gamma = 1 \) mK (typical for broad resonances), and a thermal atomic ensemble with an energy bandwidth \( \delta_e = 10 \) \( \mu \)K. We see that the transfer efficiency can reach \( \sim 97\% \) of the continuum state into the target state \( |1\rangle \) (see figure 2(c)). The parameters of the Gaussian laser pulses used (optimized Rabi frequencies, durations and delays of laser pulses) are given in table 1: the peak intensities of the Stokes and pump fields were calculated from Rabi frequencies as \( I_S = cE_S^2/8\pi = c(\Omega_p^0/\hbar)^2/8\pi \mu_{21}^2 \) and \( I_p = cE_p^2/8\pi = c(\Omega_p^0/\hbar)^2/32\pi^{3/2} \mu_{2e}^2 \), where we use equation (19) to estimate the continuum-bound dipole matrix element \( \mu_{2e} \approx \mu_{2b}/q\pi V(\epsilon) = \sqrt{2} \mu_{2b}/q\sqrt{\pi \Gamma} \), resulting in \( I_p = g^2(\Omega_p^0)^2 \delta_e/64\sqrt{\pi \mu_{2b}^2} \).
Table 1. Parameters of the Stokes and pump photoassociating pulses providing optimal population transfer for a pair of atoms shown in figure 2. We use $q = 10$, $\gamma = 10^{8} \text{s}^{-1}$, and $\mu_{2b} = \mu_{21} = 0.1 \text{ D}$ ($1 \text{ D} = 10^{-18} \text{ esu cm} = 0.3934 \text{ eA}_0$).

Rabi frequencies are modeled by Gaussians $\Omega_{1S} = \Omega_{0S} \exp(-(t - t_0 \pm \tau_{S,p})^2 / T_{S,p}^2)$, where $\pm$ refers to the Stokes and pump pulses, respectively.

| Resonance | $\delta_e$ ($\mu K$) | $\Gamma$ ($\mu K$) | $\Omega_{1S}^0$ $(10^8 \text{s}^{-1})$ | $I_S$ (W cm$^{-2}$) | $I_p$ (W cm$^{-2}$) | $T_S$ ($\mu s$) | $T_p$ ($\mu s$) | $\tau_S$ ($\mu s$) | $\tau_p$ ($\mu s$) |
|-----------|----------------------|------------------|----------------|-----------------|-----------------|--------------|--------------|--------------|--------------|
| None      | 10                   | –                | 0.72           | 62              | $4 \times 10^5$ | 1.5          | 3            | 0.75         | 1.0          |
| Broad     | 10                   | 1000             | 0.74           | 65              | 4000            | 1.4          | 3.4          | 0.65         | 1.0          |
| Narrow    | 100                  | 1                | 2.24           | 600             | 400             | 0.157        | 0.3          | 0.1          | 0.207        |

When comparing the results for a broad resonance with that of the unperturbed continuum (i.e. far from the resonance), we find that the source term $S$ is enhanced by the factor $g(q, \epsilon_0)$ (see equation (A.1) in appendix A):

$$g(q, \epsilon_0) = \frac{q + \frac{2}{\Gamma} (\epsilon_0 - \epsilon_F)}{\sqrt{1 + \frac{4}{\Gamma^2} (\epsilon_0 - \epsilon_F)^2}}.$$  \hfill (24)

This factor has a maximum at $2(\epsilon_0 - \epsilon_F)/\Gamma = 1/q$, with the corresponding maximum value $\sqrt{1 + q^2} \approx q$ for $q \gg 1$: hence, the source amplitude is enhanced $q$ times. In this limit, all populated continuum states experience the same transition dipole matrix element enhancement factor to the state $|2\rangle$, so that the system essentially reduces to the case of a flat continuum with a uniformly enhanced transition dipole matrix element. One thus expects that in this limit, the adiabatic passage should be efficient, requiring less pump laser intensity when compared with the unperturbed (i.e. without resonance) scattering continuum. This is clearly demonstrated in figure 2 (left column, dashed lines): to reach the same $\sim 97\%$ transfer efficiency achieved with the broad resonance, a very large pump laser intensity ($\sim 100$ times larger) is required if there is no resonance in the continuum (figure 2(a)), while the Stokes laser intensity is basically the same. Considering the intensity used in this particular example, this would lead to intensities in the range of $5 \times 10^5 \text{ W cm}^{-2}$, making STIRAP from the continuum technically impossible to achieve without a resonance. This is consistent with the analysis of photoassociative adiabatic passage from an unstructured continuum [18], and the above prediction that in the presence of a wide resonance the required pump laser intensity is reduced by a factor of $\sim 1/q^2$.

Results of adiabatic passage for a pair of atoms in a narrow resonance limit are shown in figure 2 (right column). We considered the typical values for a narrow resonance width $\Gamma = 1 \mu K$ and the ensemble energy bandwidth $\delta_e = 100 \mu K$. Again, we give the parameters providing the optimal transfer in table 1. In this limit, the transfer efficiency is lower: in the specific case analyzed here, it does not exceed $47\%$. The reason for this lower efficiency compared with a wide resonance is the destructive quantum interference which leads to electromagnetically induced transparency [27] in the transition from the continuum to the excited state. It can be explained using the following argument (see figure 3). The limit of a narrow Feshbach resonance corresponds to a weak coupling between the bound Feshbach state and the scattering continuum, and thus can be neglected in this simplified explanation. The system can then be viewed as consisting of bound and continuum states $|b\rangle$ and $|c\rangle$ having the
Figure 2. Time-dependence of the Stokes and pump pulses (top row) and population in state $|2\rangle$ (middle row) and target state $|1\rangle$ (bottom row) for the STIRAP transfer of a pair of atoms within the center of the thermal distribution. The left column is for a broad Feshbach resonance, while the right column is for a narrow resonance (see table 1 for values of the parameters used). The dashed blue lines in the left column are the results obtained without resonance, when the parameters are adjusted to obtain the same overall transfer efficiency as for the broad resonance. The Stokes Rabi frequency is in units of $10^6$ s$^{-1}$, while the pump Rabi frequency is in dimensionless units $(16\pi/\delta \epsilon)^{1/4} \bar{\mu}_{2\epsilon} \bar{E}_p \epsilon_p$ in the broad resonance limit and $(2\pi/\Gamma)^{1/2} \bar{\mu}_{2\epsilon} \bar{E}_p \epsilon_p$ in the narrow resonance limit. Note that the scale for the Rabi frequencies in the narrow resonance case is 40 times the scale for the broad resonance, and the magnitude of the pump Rabi frequency is enlarged 10 times for better visibility.

same energy, which are coupled by the pump field to a molecular state $|2\rangle$, itself coupled to the state $|1\rangle$ by the Stokes field. Assuming that initially all the population is in the state $|c\rangle$, due to the small interaction strength between $|b\rangle$ and $|c\rangle$, we can eliminate the state $|b\rangle$, taking into account its coupling to $|2\rangle$ by the pump laser as the formation of ‘dressed’ states $|\pm\rangle = (|2\rangle \pm |b\rangle)/\sqrt{2}$. If the dipole matrix element of the $|b\rangle \rightarrow |2\rangle$ transition is much larger than that of the $|c\rangle \rightarrow |2\rangle$ transition, the detuning of the ‘dressed’ states satisfies $|\Delta_\pm| = \Omega_{p}^{2b} \gg \Omega_{p}^{2c}$, $\Omega_S$. As a result, the
one-photon coupling of \(|c\rangle\) to the excited state, as well as two-photon coupling to \(|1\rangle\) vanishes, preventing the adiabatic transfer. This mechanism is similar to the Fano interference effect, the difference is that the continuum is initially populated. One can therefore view it as an inverse Fano effect. The effective dipole matrix element of the \(|c\rangle \rightarrow |2\rangle\) transition is \(\mu_{2c} \sim \mu_{2b}/q \sqrt{\xi}\).

In the case we analyzed, \(q = 10, \xi = \Gamma/\sqrt{2}\delta_{c} = 0.01\) and \(\mu_{2c} \approx \mu_{2b}\), which gives \(\sim 50\%\) transfer efficiency.

The transfer efficiency increases if the Feshbach state is far detuned from the populated continuum. Our calculations show that for a Feshbach state detuning \(\epsilon_{F} = \bar{h}(\omega_{S} - \omega_{p})\gg |\Omega_{2b}|^{2}/\gamma\), the transfer efficiency reaches 70% using the laser pulse parameters in table 1. We note that the smaller intensity of the pump pulse used for the narrow resonance, as compared with the broad resonance, is due to the fact that we used the same \(q = 10\) and assumed \(\mu_{2b} = 0.1\) D for both resonances. From the definition of \(q\), it means that the continuum-bound dipole matrix element \(\mu_{2c}\) is higher in the narrow than in the broad resonance we considered. This explains the smaller resulting pump pulse intensity. The overall conclusion for a narrow resonance is that, as opposed to a broad resonance, the presence of the Feshbach resonance prevents one from realizing high transfer efficiencies. It should be noted, however, that the destructive quantum interference effect is based on negligible interaction between the Feshbach and continuum states during the transfer time, since \(T < \delta_{c}^{-1} \ll \Gamma^{-1}\). This argument shows that already for \(\Gamma \gg \delta_{c}\), there is enough interaction to neutralize the effect of destructive interference. Therefore, we expect that the broad resonance limit can be extended down to \(\Gamma \sim \delta_{c}\), making it applicable to a wide variety of atomic species.

4. Conversion of atomic ensembles into ground state molecules

The results of figure 2 were obtained for a pair of atoms having a specific mean collision energy \(\epsilon_{0} = \hbar(\omega_{S} - \omega_{p})\). Such a situation could be realized in very tight traps, e.g. in tight optical lattices. For a system with a wider energy distribution, one would like to find an
ensemble averaged transfer efficiency, and thus one needs to calculate the transfer probability $P(\epsilon_0) = |c_1|^2$ for all central wavepacket energies $\epsilon_0$ within the thermal spread of energies, and perform the averaging as

$$P_{\text{avg}} = \frac{2}{\sqrt{\pi} (k_B T)^{3/2}} \int_0^\infty e^{-\epsilon_0/k_B T} \sqrt{\epsilon_0} P(\epsilon_0) d\epsilon_0,$$

where we assume a Maxwell–Boltzmann energy distribution, the pump laser resonant with the center of the distribution at $\langle \epsilon \rangle = 3/2 k_B T$, and set the bandwidth of the distribution at $\delta_\epsilon = \sqrt{\langle (\Delta \epsilon)^2 \rangle} = \sqrt{3/2} k_B T$. The results are shown in figure 4: while the maximum transfer efficiency in the broad resonance case is $\sim 70\%$, it can be achieved with lower laser intensities than in the case of a pair of atoms of figure 2.

Given the adiabatic photoassociation probability $P(\epsilon)$ for two colliding atoms with relative energy $\epsilon$, we can calculate the number of atoms photoassociated during the time overlap $\tau$ of the Stokes and pump pulses. During this time, the atom with the energy $\epsilon = \mu v^2/2$, where $\mu$ is the reduced mass, will collide with atoms in the volume $\pi b^2 v \tau$, where $\pi b^2$ is the collision cross section. The impact parameter for the collision corresponding to a partial wave with angular momentum $\ell$ is $b = (\ell + 1/2) \hbar / p = (\ell + 1/2) \hbar / \sqrt{2 \mu \epsilon}$. The number of collisions that atoms with a relative energy in the interval $(\epsilon, \epsilon + d\epsilon)$ will experience during the transfer time

Figure 4. Same as figure 2, but for the energy averaged transfer. The parameters are listed in table 2.
is therefore \( N(\varepsilon) d\varepsilon = \pi b^2 v \tau \rho(\varepsilon) d\varepsilon \), where \( \rho(\varepsilon) = 2 \rho \exp(-\varepsilon/k_B T) \sqrt{\varepsilon}/\sqrt{\pi}(k_B T)^{3/2} \) is the spectral density of the atoms (\( \rho \) is the density of the sample). Finally, \( \ell = 0 \) for ultracold s-wave collisions, and the fraction of atoms in the energy interval \( (\varepsilon, \varepsilon + d\varepsilon) \) photoassociated by the two pulses is \( f(\varepsilon) = P(\varepsilon) N(\varepsilon) \), or

\[
f(\varepsilon) = \frac{\sqrt{2\pi} h^2}{4(\mu k_B T)^{3/2}} \tau \rho P(\varepsilon) \exp(-\varepsilon/k_B T).
\]

The total fraction of atoms photoassociated by a pair of pulses is \( f = \int_0^\infty d\varepsilon \ f(\varepsilon) \approx P_{\text{avg}} \rho \sqrt{2\pi} \tau h^2 / 4 \mu k_B T \), where we assumed that \( P(\varepsilon) \) does not significantly vary within the ensemble, and approximated it by the averaged value \( P_{\text{avg}} \). Considering as an example \(^6\text{Li} \) atoms at \( T = 100 \mu K \) with an atomic density \( \rho = 10^{12} \text{cm}^{-3} \), an overlap time \( \tau \approx 1 \mu s \), and assuming \( P_{\text{avg}} = 0.7 \), the fraction of atoms photoassociated by the Stokes and pump pulses is \( f \approx 2.5 \times 10^{-4} \): for heavier atoms, \( f \sim 10^{-6} \sim 10^{-5} \). It will therefore require \( \sim 10^4 \sim 10^6 \) pairs of pulses for converting a significant fraction of an atomic ensemble into deeply bound molecular levels.

While only a small fraction of atoms can be transferred to \( |1\rangle \) by a pair of STIRAP pulses, a train of pulse pairs can be applied to photoassociate most of the atomic ensemble. To prevent excitation of molecules in \( |1\rangle \) back to the continuum by subsequent pulses, they have to be removed before the next pair of pulses is applied. This could be realized by applying, after each pair of Stokes and pump pulses, a relatively long pulse resonant with a transition from \( |1\rangle \) to some other vibrational level in the excited electronic potential which decays spontaneously to a deep vibrational state in the ground electronic potential. This long pulse would optically pump molecules out of the state \( |1\rangle \) to deeper vibrational states in the ground electronic potential. It therefore has to be longer than the spontaneous decay time of the excited state. The excited state has to be chosen carefully so that it does not decay back into the scattering continuum. This would empty the \( |1\rangle \) state and deposit molecules into ground potential vibrational states according to Franck–Condon factors before the next pair of pulses arrives. Finally, after all atoms have been converted into molecules the recently demonstrated optical pumping for molecules method [28] can be applied, which would transfer molecules from all populated vibrational states into the ground level \( v = 0 \).

The optimal strategy is to actually choose an excited state that decays mostly to the \( v = 0 \) level. This would allow one to avoid storing molecules in unstable vibrational states and using the optical pumping method. If such a state cannot be directly reached from \( |1\rangle \), a four-photon STIRAP transfer can be applied [29], which provides efficient transfer to deeply

| Resonance | \( \delta_\varepsilon \) (\( \mu K \)) | \( \Gamma_\varepsilon \) (\( \mu K \)) | \( \Omega_\varepsilon^2 \) (\( 10^8 \text{s}^{-1} \)) | \( I_S \) (W cm\(^{-2} \)) | \( I_P \) (W cm\(^{-2} \)) | \( T_S \) (\( \mu s \)) | \( T_P \) (\( \mu s \)) | \( T_s \) (\( \mu s \)) | \( \tau_p \) (\( \mu s \)) |
|-----------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| None      | 10             | –              | 0.50           | 30             | \( 1.7 \times 10^5 \) | 1.5            | 3.3            | 0.75           | 1.3            |
| Broad     | 10             | 1000           | 0.60           | 40             | 2500           | 1.3            | 3.2            | 0.7            | 1.25           |
| Narrow    | 100            | 1              | 2.24           | 600            | 400            | 0.157          | 0.3            | 0.1            | 0.207          |
bound molecular states. It allows one to choose the final state $|\Gamma\rangle$, from which the excited state decaying predominantly to $v = 0$ can be easily reached. In this case rotational selectivity can also be preserved, since only $v = 0$, $J = 0$ and $v = 0$, $J = 2$ states will be populated.

The total time required to photoassociate most of the atomic ensemble and transfer it to the $v = 0$ level can be estimated as follows. As the numerical results show, the adiabatic passage requires $\sim 5 \mu s$, the follow-up pulse emptying state $|\Gamma\rangle$ can have a $\sim 100$ ns duration, if the excited state lifetime is tens of ns, resulting in the whole sequence $\sim 6 \mu s$. In the time interval between pulses, the atomic ensemble needs to equilibrate to fill the depleted energy states. In the vicinity of the Feshbach resonance, the elastic scattering length $a$ can be large enough to give a rapid thermalization and thus replenish the energy states, while keeping three-body losses under control. For example, for $^{87}\text{Rb}$ with $a = 10^3 a_0$ (where $a_0$ is Bohr radius), $T = 100 \mu K$ and $\rho = 10^{12}$ cm$^{-3}$, an elastic collision rate $\Gamma_{\text{elas}} \sim 4\pi a^2 v \rho \sim 10^4$ s$^{-1}$ can be realized (using the relative velocity $v = \sqrt{2\epsilon/\mu}$ with $\epsilon \sim 3 k_B T/2$). This means that a delay of a few hundreds of microseconds between pulse pairs will suffice to ensure thermalization. Then the train of $10^4$–$10^6$ pulse pairs will take $\sim 1$–$100$ s. The final step, optical pumping to the $v = 0$ level, requires $\sim 100 \mu s$. Given an illuminated volume $\sim 10^{-2}$–$10^{-3}$ mm$^3$ and an atomic density $\rho \sim 10^{12}$ cm$^{-3}$, the resulting production rate is expected to be $10^4$–$10^7$ molecules s$^{-1}$. This compares well with recent experiments on STIRAP production of ground state KRb molecules starting from the Feshbach bound state, where $\sim 3 \times 10^4$ ground state molecules are produced during the entire cycle, including creation of Feshbach molecules, taking $\sim 10$–$30$ s [12].

Alternatively, back-stimulation of formed molecules into the continuum by subsequent STIRAP pulses can be avoided by placing them in a moving optical lattice, holding molecules but not atoms [18]. Another way of avoiding back-stimulation, applicable to polar molecules, is to overlap the atomic trap with a gradient of a dc electric field. It will leave dipoleless atoms unaffected, while shifting molecules out of STIRAP laser beams.

5. Conclusion

Combining both photoassociation and coherent optical transfer to rovibrational levels of the ground electronic molecular potential can allow one to convert a significant fraction of an atomic ensemble into deeply bound molecular states, and to produce an ultracold molecular gas with high phase-space density. We analyzed photoassociative adiabatic passage in a thermal ultracold atomic gas near a Feshbach resonance. The presence of a bound state embedded in and resonant with scattering continuum states strongly enhances the continuum-bound transition dipole matrix element to an excited electronic state, thus requiring less laser intensity for efficient transfer. In the limit of a wide resonance, when compared with the thermal spread of collision energies, the dipole matrix element is enhanced by the Fano parameter $q$. By choosing a tightly bound excited vibrational state, $q$ can be made much larger than unity, resulting in the intensity of the pump pulse required for efficient adiabatic passage to be $\sim 1/q^2$ times smaller than in the absence of the resonance. We modeled the adiabatic passage using typical parameters of alkali dimers and found intensities and durations of STIRAP pulses providing optimal transfer. Intensities of the pump pulse, coupling the continuum to an excited state, were

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found to be a few kW cm\(^{-2}\), which is \(\sim\)100 times smaller than without resonance. Optimal pulse durations are several microseconds, resulting in energies per pulse \(\sim\)10 \(\mu\)J for a focus area of 1 mm\(^2\).

If the Feshbach resonance is narrow compared with the thermal energy spread of colliding atoms, adiabatic passage is hindered by destructive quantum interference. The reason is that electromagnetically induced transparency significantly reduces the transition dipole matrix element from the scattering continuum to an excited state in the presence of the bound Feshbach state. In the narrow resonance limit, photoassociative adiabatic passage is therefore more efficient if the resonance is far-detuned.

Due to low atomic collision rates at ultracold temperatures, only a small fraction of atoms can be converted into molecules by a pair of photoassociative pulses. To convert most of an atomic ensemble, a train of pulse pairs can be applied. We estimate that \(10^4\)–\(10^6\) pulse pairs will associate an atomic gas of alkali dimers with a density of \(10^{12}\) cm\(^{-3}\) in an illuminated volume of \(10^{-2}\)–\(10^{-3}\) mm\(^3\), resulting in extremely high production rates of \(10^4\)–\(10^7\) molecules s\(^{-1}\). High transfer efficiencies combined with low intensities of adiabatic photoassociative pulses also make the broad resonance limit attractive for quantum computation. For example, a scheme proposed in [30] can be realized, where qubit states are encoded into a scattering and a bound molecular states of polar molecules. To perform one- and two-qubit operations, this scheme requires a high degree of control over the system, which our model readily offers.

Finally, marrying FOPA and STIRAP is a very promising avenue to produce large amounts of molecules, for a variety of molecular species. In fact, although we described here examples based on magnetically induced Feshbach resonances, such resonances are extremely common, and can be induced by several interactions, such as external electric fields or optical fields. Even in the absence of hyperfine interactions, other interactions can provide the necessary coupling, such as in the case of the magnetic dipole–dipole interaction in \(^{52}\)Cr [31, 32].

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**Appendix A. Adiabatic passage in the limits of broad and narrow Feshbach resonances**

In this appendix, we discuss equations (15) and (16) for various relative widths of the Feshbach resonance \(\Gamma\) with respect to the thermal energy spread \(\delta_e\) of the colliding atoms. Firstly, we describe the case of a broad resonance, i.e. when the width of the Feshbach resonance greatly exceeds the thermal energy spread (\(\Gamma \gg \delta_e\)), and secondly, we consider the opposite situation of a narrow resonance (\(\Gamma \ll \delta_e\)). Finally, we briefly present the case where there is no resonance.

**A.1. Limit of a broad Feshbach resonance \(\Gamma \gg \delta_e\)**

The typical thermal energy spread for colliding atoms in photoassociation experiments with non-degenerate gases is \(\delta_e \sim 10–100\mu\)K. The broad resonance case occurs for resonances having a width of several Gauss (\(\sim\)1 mK), for which we have \(\Gamma/\delta_e \sim 10–100\). A wide variety of systems exhibit broad resonances. For instance, they can be found in collision of \(^6\)Li atoms at 834 G for the \(|f = 1/2, m_f = 1/2\rangle \otimes |f = 1/2, m_f = -1/2\rangle\) entrance channel.
\( \Gamma = 302 \text{ G} = 40 \text{ mK} \) and in \(^7\text{Li} \) at 736 G for the \(|f = 1, m_f = 1\rangle \otimes |f = 1, m_f = 1\rangle \) entrance channel \((\Gamma = 145 \text{ G} = 19 \text{ mK})\). We note here that these values of \( \Gamma \) are slightly different from the ‘magnetic’ \( \Delta B \) usually given and based on the modeling of the scattering length.

The source function can be readily calculated from equation (20) by noting that the Rabi frequency term can be set at \( \epsilon = \epsilon_0 \) corresponding to the maximum of the Gaussian function in the integrand. Using the function \( g(q, \epsilon) \) defined in equation (21), the result takes the form

\[
S_w = S_0 \sqrt{2\pi} \delta_\epsilon g(q, \epsilon_0) \text{sgn}(\epsilon_0 - \epsilon_F) e^{-(\tau - \tau_0)^2} / (\sqrt{2\pi} \hbar) \left[ \int_{-\infty}^{\infty} e^{-(\tau' - iD)^2} (I_1(\xi |\tau - \tau_0 - \tau'|) - iq (I_0(\xi |\tau - \tau_0 - \tau'|) - L_0(\xi |\tau - \tau_0 - \tau'|)) \text{sgn}(\tau - \tau_0 - \tau') d\tau' \right],
\]

where \( S_0 = \mu_2 \delta_{\epsilon p} e_p / \hbar \) is the continuum-bound Rabi frequency in the absence of resonance. We also added a spontaneous decay term \( \gamma c_2 \), assuming that the excited molecules dissociate into high-energy continuum states and the resulting atoms leave a trap. From equation (A.1), one can see that in a broad resonance case, the source amplitude is enhanced by the factor \( g(q, \epsilon_0) = \sqrt{2\pi} \hbar \) when compared with the unperturbed continuum case. This factor has a maximum at \( 2(\epsilon_0 - \epsilon_F) / \Gamma = 1/q \), with the corresponding maximum value \( g_{\text{max}} \approx 1 \).

\[ \text{A.2. Limit of a narrow Feshbach resonance } \Gamma \ll \delta_\epsilon \]

This situation occurs when the width of the resonance is of the order of a few micro-Gauss or less. Examples of narrow resonances include \(^6\text{Li}^{23}\text{Na} \) at 746 G for the \(|f_1 = 1/2, m_{f_1} = 1/2\rangle |f_2 = 1, m_{f_2} = 1\rangle \) channel \((\Gamma = 7.8 \text{ mG} = 1 \mu \text{K})\) [26], or \(^6\text{Li}^{87}\text{Rb} \) at 882 G for the \(|f_1 = 1/2, m_{f_1} = 1/2\rangle |f_2 = 1, m_{f_2} = 1\rangle \) channel (p-wave, \( \Gamma = 10 \text{ mG} = 1.3 \mu \text{K})\).

We note that the source term expressed in equation (20) can be rewritten in a time representation:

\[
S = S_0 \sqrt{2\pi} \delta_\epsilon e^{-i(\epsilon_0/\hbar - (\omega_S - \omega_p))t} \left[ e^{-(\tau - \tau_0)^2} + \xi e^{2iD - D^2} \int_{-\infty}^{\infty} e^{-(\tau' - iD)^2} (I_1(\xi |\tau - \tau_0 - \tau'|) - iq (I_0(\xi |\tau - \tau_0 - \tau'|) - L_0(\xi |\tau - \tau_0 - \tau'|)) \text{sgn}(\tau - \tau_0 - \tau') d\tau' \right].
\]
where we introduced the dimensionless variables \( \tau = t \delta \phi / \sqrt{2} h \), \( D = (\epsilon_F - \epsilon_0) / \sqrt{2} \delta \phi \), \( \xi = \Gamma / \sqrt{2} \delta \phi \); \( I_{0,1} \) and \( L_{0,-1} \) are modified Bessel and Struve functions. One can see from this expression that the source function is a sum of the pure source function of the unperturbed continuum, given by the first term in square brackets, and of the admixed bound state, given by the integral. The coefficient \( \xi = \Gamma / \sqrt{2} \delta \phi \), which is the ratio of the Feshbach resonance width to the width of the thermal energy spread, gives the ratio of contributions from the bound state and the unperturbed continuum, respectively.

It is then easier to note that in the limit of a narrow resonance, the Gaussian function in the integrand of equation (A.5) is much narrower than the Bessel and Struve functions, which change on the timescale \( \sim 1 / \xi \). Therefore the source term can be approximated as

\[
S_n = S_0 \sqrt{2 \pi} \delta \phi e^{-i(\epsilon_F - \epsilon_0) t} \left[ e^{-(\tau - t_0)^2} + \xi \sqrt{\pi} e^{2D^2 - D^2} (I_1(\xi |\tau - t_0|) - L_{-1}(\xi |\tau - t_0|) - i q I_0(\xi |\tau - t_0|) - L_0(\xi |\tau - t_0|) \sgn(\tau - t_0)) \right].
\]

(A.6)

Since \( \xi \ll 1 \), the real part of the source function is given by the first term in the square brackets, which is a pure continuum source function, while the imaginary part is due to the admixed bound state and its magnitude depends on the product \( \xi q \). Using asymptotic expansions of modified Bessel and Struve functions \( I_q(x) - L_q(x) \to -2/\pi x \), \( I_1(x) - L_{-1}(x) \to -2/\pi x^2 \), it is seen from equation (A.6) that the contribution to the source function from the bound state decays on the timescale \( |\tau - t_0| \sim 1 / \xi \), while the contribution from the unperturbed continuum decays on the timescale \( |\tau - t_0| \sim 1 \ll 1 / \xi \).

In the limit of a narrow resonance the system (15) and (16) becomes

\[
i \frac{\partial c_1}{\partial t} = -\Omega_s c_2,
\]

(A.7)

\[
i \frac{\partial c_2}{\partial t} = -\Omega_s c_1 - S_n + (\delta - i \gamma) c_2 - i \left[ \frac{\tilde{\mu}_{2e} \hat{e}_p}{\hbar} \right]^2 \left[ \pi h \mathcal{E}_p^2 c_2 + \frac{\pi \Gamma}{2} (q - i)^2 \mathcal{E}_p(t) \times \int_0^t dt' c_2(t') \mathcal{E}_p(t') e^{\Gamma(t' - t)/2 + i(\epsilon_F - \epsilon_0 - \delta \phi) (t' - t)} \right].
\]

(A.8)

A.3. Continuum without resonance

Finally, let us consider the case of a continuum without resonance. In this case, the continuum-bound Rabi frequency equation (18) is

\[\Omega_c = \Omega_\text{no-res} = \tilde{\mu}_{2e} \cdot \hat{e}_p \mathcal{E}_p / \hbar,\]

(A.9)

and the source function is

\[S_\text{no-res} = S_0 \sqrt{2 \pi} \delta \phi e^{-(\tau - t_0)^2 / 2 \hbar^2 - i(\epsilon_F - \epsilon_0 - \delta \phi) t}.\]

(A.10)

The back-stimulation term (23) reduces to

\[\left| \tilde{\mu}_{2e} \cdot \hat{e}_p / \hbar \right|^2 \pi \hbar \mathcal{E}_p^2 c_2 = \pi \hbar \Omega_\text{no-res}(t) \mathcal{E}_p^2 c_2,\]

(A.11)
and the system (15) and (16) takes the simple form

\[
\begin{align*}
\frac{i}{\hbar} \frac{\partial c_1}{\partial t} & = -\Omega_S c_2, \\
\frac{i}{\hbar} \frac{\partial c_2}{\partial t} & = -\Omega_S c_1 + (\delta - i\gamma) c_2 - i\pi \hbar |\Omega_{\mathrm{no-res}}(t)|^2 c_2 - S_{\mathrm{no-res}}.
\end{align*}
\]

(A.12)

(A.13)

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