High-fidelity formation of deeply bound ultracold molecules via non-Hermitian shortcut to adiabaticity

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Abstract
Stimulated Raman adiabatic passage allows robust transfer between two ends of a three-state quantum system and has been employed to transfer weakly bound Feshbach molecules into their deeply bound rovibrational ground state. However, the efficient transfer remains to be explored. Here we propose a possible alternative route, based on a recently developed non-Hermitian shortcut to adiabaticity method. It is able to realize single-step transfer efficiencies up to 100% even in the presence of a decaying excited level, surpassing all the previous methods. We also prove that our scheme is robust against the external field parameter fluctuations and is expected to be applicable for abundant molecular species.

1. Introduction
Creation of quantum degenerate gases of ultracold deeply-bound molecules is an essential task in many branches of fundamental physics and chemistry [1–3]. Such samples promise myriad possibilities since their translational, vibrational and rotational degrees of freedom can be manipulated with great accuracy [4]. The potential applications range from few-body collisional physics [5], ultracold chemistry [6, 7], precision measurements [8] to quantum computation [9] and quantum simulation [10].

Strategies toward the creation of translational ultracold molecules (100 nk–1 mk) generally follow binary association of precooled atoms via either the light-assisted photoassociation [11] or magnetically induced Feshbach association [12]. While the latter is effective, reversible and adiabatic, and inherits the phase-space density from the initial atomic gases [13], it is typically restricted to leave molecules in weakly bound vibrational levels near the dissociation limit [14, 15]. For collisional and radiative stability, these molecules must be quickly moved to their rovibronic ground state \(|\nu'' = 0, J'' = 0\rangle\), i.e. the lowest energy level of the electronic ground state. As a general method, Stimulated Raman adiabatic passage has been used to step weakly bound Feshbach molecules down the vibrational ladder [16], as demonstrated for molecular species like homonuclear Rb₂ [17–19], Cs₂ [20, 21], Sr₂ [22], and heteronuclear KRb [4, 23–25], RbCs [26–29], NaK [14, 30–33], RbSr [34], NaRb [35, 36], NaLi [37]. In particular, molecules can be loaded in a collision-free environment of a 3D optical lattice to prevent reactive and inelastic collisions during state preparation or manipulation.

STIRAP as a special coherent optical transfer scheme, in principle, allows lossless transfer via an adiabatic evolution of the dark state [38], and avoids any losses that would otherwise occur from spontaneous light scattering [13]. Despite the advantages, adiabatic following requires a long time, and hence can result in significant molecular loss due to undesirable interactions [29]. The transfer efficiency of Feshbach molecules to their rovibrational ground state is reported hitherto typically of about 90% [27, 28, 30], below the efficiency potentially of STIRAP. A way to avert this detriment of adiabatic following is based on an alternative scheme characterized as shortcut to adiabaticity (STA) [39–49], for instance, counter-diabatic driving (equivalently, the transitionless quantum algorithm) [39–41, 50]. The basic idea is to drive faster system at the same final state as the slow adiabatic evolution. STA has been widely applied in designing control scheme for closed-system scenarios since its inception [51]. In practice, however,
ultracold molecular transfer is inevitably subject to interactions with its surroundings \[52\], especially for the spontaneous emission from intermediate excited levels. Such a system admits an effective non-Hermitian (NH) description \[53\]. Recently, the success of STA has prompted its extension to perform for several versions of NH system \[52, 54–57\].

In this paper, we develop a possible alternative scheme to transfer weakly bound Feshbach molecules to their deeply bound state, based on a recently developed non-Hermitian shortcut to adiabaticity method. We consider the spontaneous emission from the intermediate excited molecular electronic vibrational state and employ an NH Hamiltonian to quantitatively describe the details of adiabatic transfer. We demonstrate that our scheme achieves complete molecular transfer with robustness against the external field parameter fluctuations, since the modulated pulses eliminate the nonadiabatic effect. Considering the specific technical limitation, we further simplify the auxiliary laser fields.

The rest of this paper is organized as follows. Section 2 describes the model and NH Hamiltonian, and derive the formula of auxiliary driving fields accordingly, which aims to nullify the nonadiabatic coupling applied to transfer process. Numerical simulation results are presented in section 3. The physical implementation is given in section 4. Finally, section 5 is devoted to conclusion and outlook.

2. Molecular level and Hamiltonian

The idea of this work can be described using a simple open three-level molecular system, as illustrated in figure 1. On the premise that the hyperfine structure is fully distinguishable, the states \(|\bar{i}\rangle\) and \(|\bar{g}\rangle\) are vibrational levels of the ground electronic molecular state, while \(|e\rangle\) is vibrational state of the excited electronic molecular state. Here \(|\bar{i}\rangle\) and \(|\bar{g}\rangle\) are assumed to be molecular Feshbach and a well-defined deeply-bound rovibrational ground state, respectively. The optical fields \(\Omega_P\) and \(\Omega_S\) couple the molecular ground state levels \(|\bar{i}\rangle, |\bar{g}\rangle\) to a common excited level \(|e\rangle\), leading to a two-photon Raman transfer. The excited level \(|e\rangle\) is off single-photon resonance and decays out of the system by the spontaneously decays. Here we neglect the finite lifetime of the Feshbach and deeply-bound vibrational states due to the fact that they are long on the time scale of the STIRAP transfer. Noting that the identification of a suitable excited level serves as a bridge for the transfer is of crucial importance, one should have favourable transition dipole moments and good Frank–Condon overlaps with both \(|\bar{i}\rangle\) and \(|\bar{g}\rangle\) vibrational wavefunctions.

2.1. STIRAP protocol

First we consider the standard STIRAP protocol for coherent population transfer between two molecular internal quantum states. The total molecular wave function can be expanded as

\[
|\Phi(t)\rangle = c_i(t)|\bar{i}\rangle + c_e(t)|e\rangle + c_g(t)|\bar{g}\rangle,
\]

the vector \(c_i(t), c_e(t),\) and \(c_g(t)\) are the probability amplitudes of the three levels \(|\bar{i}\rangle, |e\rangle,\) and \(|\bar{g}\rangle\), respectively. The evolution is then governed by the time-dependent Schrödinger equation:

\[
i\hbar \frac{\partial}{\partial t} c(t) = H(t)c(t).
\]
Here, \( c(t) \) is a three-component column matrix with the elements \( \{ c_i(t), c_e(t), c_g(t) \} \), and \( P_{e\rightarrow g} = |c_{e\rightarrow g}(t)|^2 \) are the corresponding probability. Under the rotating wave approximation, this system can be quantitatively described in terms of a three-level NH Hamiltonian \((\hbar = 1)\) [17, 58],

\[
H(t) = \frac{\hbar}{2} \begin{pmatrix}
0 & \Omega_p(t) & 0 \\
\Omega_p(t) & 2\Delta_p - i\Gamma & \Omega_g(t) \\
0 & \Omega_g(t) & 2(\Delta_p - \Delta_g)
\end{pmatrix}.
\] (3)

Here \( \Omega_p(t) \) and \( \Omega_g(t) \) are Rabi frequencies of pump and Stokes laser fields, shown in figure 1. The quantities \( \Delta = (E_e - E_i)\hbar - \omega_p, \Delta_g = (E_e - E_g)\hbar - \omega_S \) stand for the one- and two-photon detunings of the pump and Stokes pulses from their respective transitions, respectively. \( E_{e \rightarrow g} \) denote bare-state basis energies [59].

The imaginary term \( \Gamma \) is introduced here to describe the inevitable spontaneous emission from state \(|e\rangle\), based on the above assumption that this decay dominates all other loss mechanisms [60]. Thus \( P_i + P_e + P_g \leq 1 \), and the equal sign implies that the total particle number is conserved, when \( \Gamma = 0 \).

An essential condition for the conventional STIRAP procedure is the two-photon resonance, defined as \( \Delta_p = \Delta_g = \Delta \), then the Hamiltonian (3) can be written as

\[
H_0(t) = \frac{\hbar}{2} \begin{pmatrix}
0 & \Omega_p(t) & 0 \\
\Omega_p(t) & 2\Delta - i\Gamma & \Omega_g(t) \\
0 & \Omega_g(t) & 0
\end{pmatrix}.
\] (4)

We introduce the so-called adiabatic basis to understand the mechanism of the population transfer in STIRAP and the limitation of adiabaticity. It is straightforward to verify that the following linear combinations of the bare states \(|i\rangle, |e\rangle, \) and \(|g\rangle\) are eigenstates of \( H_0(t) \):

\[
\begin{align*}
|\Phi_0(t)\rangle &= \cos \theta |i\rangle - \sin \theta |g\rangle, \\
|\Phi_+(t)\rangle &= \sin \theta \sin \phi |i\rangle + \cos \phi |e\rangle + \cos \theta \sin \phi |g\rangle, \\
|\Phi_-(t)\rangle &= \sin \theta \cos \phi |i\rangle - \sin \phi |e\rangle + \cos \theta \cos \phi |g\rangle.
\end{align*}
\] (5)

The eigenvalues of \( H_0(t) \) are

\[
E_0(t) = 0, \quad E_\pm(t) = \frac{\hbar}{2}(\Delta - i\Gamma/2) \pm \sqrt{(\Delta - i\Gamma/2)^2 + \Omega^2(t)},
\] (6)

where \( \Omega(t) = \sqrt{\Omega_p(t)^2 + \Omega_g(t)^2} \), and the term of time-dependent mixing angles are respectively defined as

\[
\theta = \arctan \frac{\Omega_p(t)}{\Omega_g(t)}, \quad \phi = \frac{1}{2} \arctan \frac{\Omega(t)}{\Delta - i\Gamma}.
\] (7)

For a counterintuitively ordered pulse sequence, i.e. the Stokes pulse precedes but overlaps with the pump pulse, there comes a change in the mixing angle \( \theta(t) \), i.e.

\[
\lim_{t \to -\infty} \theta(t) \to 0, \quad \lim_{t \to +\infty} \theta(t) \to \frac{\pi}{2}.
\] (8)

During this process, the molecules always remain in a dynamically changing dark state superposition of long-lived system eigenstates \( |\Phi_0(t)\rangle \), while being decoupled from the short-lived excited state \(|e\rangle\), and thus it is robust against the spontaneous emission from the excited state \(|e\rangle\). This means that if the excitation is adiabatic and the system is initially prepared in state \(|i\rangle\), at the end of the process all of the population in dark state will project onto the final deeply-bound state \(|g\rangle\) [61, 62]. We have the relation

\[
|\Phi_0(-\infty)\rangle = |i\rangle, \quad |\Phi_0(\infty)\rangle = -|g\rangle.
\] (9)

However, efficient molecular transfer requires that STIRAP must meet adiabatic conditions, i.e. \( |\langle \Phi_0(t) | \Phi_+(-\infty) \rangle| \ll |E_0(t) - E_+(-\infty)| \). This generally requires a long time to evolve the system, and therefore the system has a long time to interact with the environment leading to molecular losses or decoherence [63, 64].

### 2.2. NH-STA protocol

In the general nonadiabatic regime, there exists nonadiabatic coupling between the adiabatic states (5), which causes the STIRAP not to follow exactly the path laid down by the dark state \( |\Phi_0(t)\rangle \), leading to reduced molecular transfer efficiency [16, 38]. To overcome this, one can apply the transitionless driving algorithm, also referred to as counterdiabatic driving [39–41, 50], to eliminate the unwanted nonadiabatic...
system subjects to the total Hamiltonian, dipole transition when the electric dipole is forbidden \[64–66\]. For atomic and molecular systems, which can be physically implemented by a magnetic coupling. A supplementary Hamiltonian, in principle, should be attached to original Hamiltonian \[52\]. It thus generate the desired \( |i\rangle \rightarrow |g\rangle \) adiabatic population transfer in the three-state molecular system. The system subjects to the total Hamiltonian,

\[
H(t) = H_0(t) + H_1(t)
\]

with the correction

\[
H_1(t) = -i\hbar \sum_{a,b} \left\{ \frac{\langle \Phi_a(t)|\hat{H}_0|\Phi_b(t)\rangle}{E_b(t) - E_a(t)} \right\}, \quad (a, b = 0, +, -),
\]

which is determined by the instantaneous eigenstates of \( H_0(t) \): \( |\Phi(t)\rangle \equiv (|\Phi_0(t)\rangle, |\Phi_+(t)\rangle, |\Phi_-(t)\rangle) \) and \( H_1(t) \): \( |\Phi(t)\rangle = (|\Phi_0(t)\rangle, |\Phi_+(t)\rangle, |\Phi_-(t)\rangle) \).

Under the standard procedure, the supplementary Hamiltonian \( H_1(t) \) is further formulated into

\[
H_1(t) = i\hbar \begin{pmatrix} 0 & \Omega_p(t) & \Omega_g(t) \\ -\Omega_p(t) & 0 & -\Omega_e(t) \\ -\Omega_g(t) & \Omega_e(t) & 0 \end{pmatrix}
\]

with the matrix elements given by \( \Omega_p(t) = \sin \theta(t)\dot{\phi}(t) \), \( \Omega_q(t) = \dot{\theta}(t) \), \( \Omega_e(t) = \cos \theta(t)\dot{\phi}(t) \), where the dot represents derivative with respect to time, and the time-dependent auxiliary parameters \( \theta(t) \) and \( \phi(t) \) satisfy \( \dot{\theta}(t) = \frac{[\Omega_p(t)\Omega_q(t) - \Omega_q(t)\Omega_p(t)]}{\Omega_p(t)} \), and \( \phi(t) = \left[ \Omega(t)(\Delta - i\Gamma/2) - \Omega(t)(\Delta - i\Gamma/2)/2 \right] \Omega_p(t) + (\Delta - i\Gamma/2)^2 \right] \), respectively. Note the finite \( \Gamma \) constitutes an NH contribution to the Hamiltonian. Then population transfer from \( |i\rangle \) to \( |g\rangle \) will be forced to follow the adiabatic path defined by \( H_0(t) \). If the system initial state is in the dark one, the system will remain in that dark state at all times.

This induces the additional transition lifetimes. There new lasers fields are needed to implement this Hamiltonian. \( \Omega_p(t) \) and \( \Omega_q(t) \) represent the addition to the pump and Stokes pulses. The \( \Omega_p(t) \) couples directly the initial and the final state, which is totally defined by (the shape of) \( \Omega_p(t) \) and \( \Omega_q(t) \). Upon most occasions, we stress that the transitions \( |i\rangle \leftrightarrow |e\rangle \) and \( |e\rangle \leftrightarrow |g\rangle \) are allowed while the transitions \( |i\rangle \leftrightarrow |g\rangle \) is forbidden. But for atomic and molecular systems, which can be physically implemented by a magnetic dipole transition when the electric dipole is forbidden \[64–66\].
Figure 3. Contour plots of the transfer efficiency as a function of $\Omega_0$, $\Gamma$, and $\Delta$ for the STIRAP (left column) and the NH-STA (right column) protocols. Parameters: $t_f = 30$ ns, $T = t_f / 6$, $\tau = t_f / 10$, $\Omega_0 = 2 \pi \times [200, 800]$ MHz, $\Gamma = 2 \pi \times [0, 1000]$ MHz, $\Delta = 2 \pi \times [0, 1000]$ MHz.

Figure 4. (a) Real and imaginary parts of $\Omega_{ie}(t)$ and $\Omega_{eg}(t)$. (b) Efficiency for population transfer from state $|i\rangle$ to state $|g\rangle$ driven by $\text{Im}(\Omega_{ie}(t))$ and $\text{Im}(\Omega_{eg}(t))$. Parameters are the same of those in figure 2.

3. Numerical simulations

We study now the laser-driven coherent transfer. For the simplicity of discussion, we take $\Gamma$ as a constant and assume that the pump and Stokes pulses use partially overlapped Gaussian envelopes,

$$\Omega_p(t) = \Omega_0 \exp \left[ -\left( \frac{t - \tau - t_f/2}{T} \right)^2 \right],$$

$$\Omega_S(t) = \Omega_0 \exp \left[ -\left( \frac{t + \tau - t_f/2}{T} \right)^2 \right].$$

Here $\Omega_0$ is the peak Rabi frequency, $T$ is the pulse duration, and $\tau$ is the delay between the pulses, $t_f$ is the entire interaction time of the system evolution or the so-called operation time.

For the STIRAP transfer, we display the counterintuitive pulse sequence in figure 2(a), while time dependence of the populations of the Feshbach state $|i\rangle$, intermediate excited state $|e\rangle$, and deeply bound states $|g\rangle$ are exhibited in figure 2(c). First only pulse $\Omega_S(t)$, which couples the levels $|g\rangle$ and $|e\rangle$ is switched
on. It has no effect on the number of Feshbach molecules. Then the second pulse \( \Omega_d(t) \) is switched on with some delay with respect to the first one, which induces the |\( i \rangle \leftrightarrow |e \rangle \) (pump) transition and the |\( e \rangle \leftrightarrow |g \rangle \) (Stokes) transition [67]. The population transfer efficiency, defined as the population of the deeply bound state at the final time, is only 55.53\% for pure STIRAP protocol because it violates the adiabatic condition. The conversion no longer follows the adiabatic pathway defined by dark state |\( \Phi_0(t) \rangle \) [68]. We can understand this behavior via a superposition of dark and other eigenstates (i.e. so called bright states):

\[
|\tilde{i} \rangle = [\Omega_3|\Phi_0(t)\rangle + \Omega_p(\sin \phi|\Phi_+\rangle(t) + \cos \phi|\Phi_-\rangle(t))]/\Omega(t).
\]  

This means that as soon as the lasers are switched on, the states |\( \Phi_+\rangle(t) \) and |\( \Phi_-\rangle(t) \) are quickly exposed to irreversible losses due to excitation to |\( e \rangle \), while the dark state remains. These vanished molecules may be excited to the intermediate excited level and then undergo spontaneous emission not only to rovibrational levels |\( i \rangle \) and |\( g \rangle \), but also to other multitude bound states of the electronic ground molecular potential, or to dissociate into free atoms with high kinetic energy.

The auxiliary laser fields are shown in figure 2(b), and the dynamics of NH-ST A control is illustrated in figure 2(d). With the modified pulses predicted from equation (12), we can find that the time-dependent rovibrational population |\( i \rangle \) decreases smoothly from 1 to 0, while |\( g \rangle \) increases smoothly from 0 to 1. A perfect population transfer efficiency of 100\% can be achieved through NH-ST A protocol since the nondiabatic transition is eliminated by adding the Hamiltonian \( H_1(t) \). The evolution will follow exactly the dark state, which is decoupled from the light in the sense that there is no excitation of the short-lived state |\( e \rangle \), which makes the process insensitive to its properties, e.g. spontaneous emission to other states.

We further characterize the performance of the STIRAP and NH-ST A protocols for a wide range of external field parameters variations, including the single-photon detuning \( \Delta \), Rabi frequency \( \Omega_0 \), and the decay \( \Gamma \) from intermediate excited level |\( e \rangle \). All the other parameters are the same as those in figure 2. The results of the pure STIRAP are illustrated in figures 3(a) and (c). The transfer efficiency is close to 100\% when the intensities of pulses are sufficiently high, where the adiabatic condition is fully satisfied [38]. However, the STIRAP transfer efficiency in some parameter regions is quite low, or even close to zero. The dependence of transfer efficiency on parameter fluctuation can be qualitatively understood as follows: the dark state cannot adiabatically follow the STIRAP pulse, leading to reduce transfer efficiency [17]. Contrastively, as shown in figures 3(b) and (d), the transfer efficiency of NH-ST A can keep constant against parameter fluctuation since the nondiabatic transition is eliminated by modifying the shape of the pulses accordingly. The ultracold Feshbach molecules can be transferred to deeply-bound rovibrational ground state with high fidelity for broad range of protocol parameters, even when the amplitudes of applied pump and Stokes fields are very small.

4. Physical implementation

It should be pointed out that the NH-ST A presented above is strongly dependent on the accurate implementation of the auxiliary fields. Clearly, the (normalized) shortcut fields for the |\( i \rangle \leftrightarrow |e \rangle \) transition and |\( e \rangle \leftrightarrow |g \rangle \) transition have real and imaginary parts, see figure 4(a), which renders that physical realization of the Hamiltonian \( H_1(t) \) is not straightforward. Normally, the imaginary part can be realized by a complementary laser with orthogonal polarization [42]. The real parts are not the complex conjugate of each other unless the real part of becomes zero, so in general there is no simple laser interaction leading to equation (12). Nevertheless, the real part contributes very little to the dynamics. Neglecting the real part of (12) by setting \( \Omega_{exg}(t) \approx i \text{Im}[\Omega_{exg}(t)] \), we notice that provides essentially the same molecular dynamics, as shown in figure 4(b). Until a physical implementation of the real part is discovered, the application of this NH-ST A technique with spontaneous emission will therefore be limited to this approximate treatment.

5. Conclusions and outlook

In conclusion, we have proposed a scheme to transfer Feshbach state to deeply bound rovibrational ground state via a recently developed NH-ST A method. Modified pulses are used to drive the molecular internal quantum state transfer. The system evolution along the dark state and adiabatic population transfer |\( i \rangle \leftrightarrow |g \rangle \) take place with unit probability even in the presence of spontaneous emission in the excited state. Numerical analysis suggests our scheme is robust against the fluctuation of parameters. Given the limitations of experimental techniques, this present work further points out a relatively easy and reliable method by simplifying auxiliary laser fields.

Another issue is also worth mentioning. It has been proposed in reference [38] to transfer Feshbach molecules to the ground state through a series of intermediate vibrational states via a multi-state chainwise
STIRAP with transfer efficiency $>90\%$. Therefore, it can be expected to combine this multi-state STIRAP with our scheme to create deeply-bound molecules with high fidelity in multi-state molecular systems [63].

Finally, this work is expected to be an important step towards the high-fidelity realization of a Bose–Einstein condensate of deeply bound molecules. The applicable molecular species may be abundant. Especially for the molecular species composed of fermions [30], the Pauli blocking effect can suppress vibrational quenching and assure collisional stability [69]. Fermionic alkaline dimer Feshbach state exhibits strong stability with respect to collisions, and lifetimes of $\sim 1\ s$ have been observed [58]. For bosonic molecules, the Pauli blocking is absent, this result in a dramatic decrease of molecular lifetime in dense samples of weak-bound vibrational level [17]. But once molecules are shielded from each other in a collision-free environment of 3D optical lattice, the lifetimes can be strongly increased even for dimers of bosonic atoms [70].

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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