Molecular cooling via Raman transitions and a harmonic spectral modulation of an optical frequency comb: The role of the parity of the chirp in quantum control

S. A. Malinovskaya* and G. Liu1

1Department of Physics and Engineering Physics, Stevens Institute of Technology, Hoboken, NJ 07030
*Corresponding author: smallinov@stevens.edu

Compiled December 15, 2015

A method for creation of ultracold molecules by stepwise adiabatic passage from the Feshbach state to the fundamentally ground state using an optical frequency comb is presented within a semiclassical multilevel model. The sinusoidal modulation of the spectral phase of the comb is implemented that leads to a creation of a quasi-dark dressed state having an insignificant population of the excited state manifold and, thus, efficiently mitigating decoherence in the system. In contrast, the cosine modulation does not lead to the quasi-dark state formation. The results demonstrate the importance of the parity of the spectral chirp in quantum control.

© 2015 Optical Society of America

OCIS codes: (320.7110) Ultrafast nonlinear optics; (270.0270) Quantum optics.

http://dx.doi.org/10.1364/ao.XXXXXX

1. INTRODUCTION

Ultracold control has originated on the base of latest developments in the field of ultracold gases, that opened up possibilities to generate ultracold molecular systems that offer internal structure and exhibit long-range, dipole-dipole interactions [1]. The development of methods for molecular cooling continues to be an actual objective owing to diversified internal structure in molecules limiting the implementation of a well established stimulated Raman adiabatic passage (STIRAP) [2]. Quantum control methods have demonstrated high potential to produce ultracold molecules and open up a wide range of opportunities to operate on a time scale much faster than the spontaneous decay [3].

Decoherence is inherently present in ultracold dynamics and reveals itself in quantum measurements. A study of decoherence is of particular importance for the development of methods to manipulate ultracold atomic gases, create and control ultracold molecules, for the advancements in ultracold chemistry, quantum computation science, etc. At ultracold temperatures, decoherence acquires new features while occurs in matter systems free from thermal motion. Under these conditions, the dephasing due to collisions is not longer a limiting factor, [4], in contrast to this effect at room temperatures [5]. The central idea to success of the control scenarios in systems with decoherence is maintenance of the phase information contained both in matter and light. Understanding of the impact of decoherence, thus, is crucial for experimental realizations.

In this letter, we propose a quantum control method for a preparation of molecules in an ultracold state, e.g., the ground electronic, rovibrational, spin-singlet state, starting from the Feshbach state by means of two-photon Raman transitions induced by a spectrally modulated ultrafast pulse train. We will analyze decoherence effects that lead to the loss of the quantum phase and have detrimental effects on controllability.

2. THEORETICAL FRAMEWORK

We consider a semiclassical model of a seven-level quantum system interacting with a classical phase-locked pulse train. Each pulse in the train is spectrally modulated in a form of a sinusoidal function. The model aims to describe a controllable internal dynamics in molecules initiated from the Feshbach state and resulting in molecular stepwise transition into the ultracold state. The seven-level system has a \( \Lambda \) configuration formed by the initial Feshbach state, five transitional, electronically excited rovibrational states and the final ultracold state, Fig(1). The electronically excited rovibrational states are equally separated in accordance with the one-dimensional harmonic oscillator solution, which is a good approximation for the low laying internal states. The frequency separation \( \Delta \omega \) between these states corresponds to the vibrational frequency of the totally symmetrical mode in the KRb molecule.

The sinusoidal modulation of the spectral phase of the incident field is introduced in the frequency domain as \( M(\omega) = \)
The pulse train with such phase modulation reads

\[ e^{-iA \sin(\omega T_0 + \phi)} \]  

[7]. The spectral profile of the field is

\[ E(\omega) = \frac{\sqrt{2 \pi E_0 T}}{e^{\frac{1}{2}(\omega - \omega_0)^2 \tau^2} + e^{\frac{1}{2}(\omega + \omega_0)^2 \tau^2}} \sum_{k=0}^{N} c_k e^{i k T} \]  

(1)

Here \( E_0 \) is the peak field amplitude, \( \tau \) is the pulse duration, \( T \) is the pulse train period, \( \omega_0 \) is the carrier frequency, \( A \) is the modulation amplitude, \( T_0^{-1} \) is the modulation frequency, \( \phi \) is the phase, which may be chosen accordingly to address the parity of the chirp.

The Fourier transform of the modulation function gives the temporal variation of the phase of the field in the form of a series of Bessel functions \( M(t) = \sum_{n=-\infty}^{\infty} J_n(\omega t) e^{-i n \phi} \delta(t - n T_0) \).

The pulse train with such phase modulation reads

\[ E(t) = E_0 \sum_{n=-\infty}^{\infty} J_n(\omega t) e^{-i n \phi} \sum_{k=0}^{N} c_k e^{-(t-nT_0-kT)^2/(2\tau^2)} \cdot \cos(\omega_0(t-nT_0-kT)) \]  

(2)

For the description of the time evolution of the seven-level system we refer to the Leoville-von Neumann equation \[ i\dot{\rho} = [H_{\text{int}}, \rho] \] with relaxation terms that take into account spontaneous decay and elastic collisions.

Five excited vibrational states are uncoupled since radiative transitions between vibrational states within a single electronic state are not included. Thus, the system may be treated as a superposition of five three-level \( \Lambda \) systems with the excited states differing by \((q - 1)\Delta \omega\), where \( q \) is an integer from 1 to 5. The interaction Hamiltonian reads \[ \hat{H}_{\text{int}} = \sum_{q=1}^{5} \hat{H}_{\text{int}}^q, \]

with \( \hat{H}_{\text{int}}^q \) describing a single three-level \( \Lambda \) system from a set of five of them. The \( \hat{H}_{\text{int}}^q \) contains terms proportional to the spectral sinusoidal modulation of the field and, in the rotating wave approximation, has two nonzero matrix elements \[ \hat{H}_{ij}^q = \Omega_R(t)(\cos(\omega t) - (q - 1)\Delta \omega) \delta(i,j), \] where \( i,j \) are the indexes of the basis set, \( i = 1, 2, \) and \( j = i + 1 \), and the Rabi frequency \( \Omega_R(t) = \mu/\hbar \sum_{n} J_n(\omega t)e^{-[(nT_0)^2/(2\tau^2)]} \cdot \cos(\omega_0 t) \), with the peak value \( \Omega_R \). The Leoville-von Neumann equation gives a set of coupled differential equations for the density matrix elements

\[ \rho_{11} = 2i[H_{12}\rho_{21}] \]
\[ \rho_{22} = 2i[H_{21}\rho_{12} + H_{23}\rho_{32}] \]
\[ \rho_{33} = 2i[H_{32}\rho_{23}] \]
\[ \rho_{12} = -iH_{12}(\rho_{22} - \rho_{11}) + iH_{23}\rho_{13} \]
\[ \rho_{13} = -iH_{12}\rho_{23} + iH_{23}\rho_{12} \]
\[ \rho_{23} = -iH_{23}(\rho_{33} - \rho_{22}) - iH_{23}\rho_{13} \]  

(3)

The decoherence terms are taken into account through the reduced density matrix elements

\[ \rho_{11}\text{sp} = \gamma_1\rho_{22} \]
\[ \rho_{12}\text{sp, col} = -(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} + \Gamma_1)\rho_{12} \]
\[ \rho_{22}\text{sp} = -\gamma_1\rho_{22} - \gamma_2\rho_{22} \]
\[ \rho_{13}\text{sp, col} = -\Gamma_3\rho_{13} \]
\[ \rho_{23}\text{sp, col} = -(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} + \Gamma_2)\rho_{23}. \]  

(4)

The decay from the excited states to the initial and the final state is at the rate \( \gamma_1 \) and \( \gamma_2 \), respectively, and that from the initial to the final state is at the rate \( \gamma_3 \). Collisions are considered between the molecules in any state within the model and the buffer gas, they are described by three dephasing rates, \( \Gamma_1, \Gamma_2 \), and \( \Gamma_3 \) related to dephasing between the excited state manifold and the initial state, that and the final state and the initial state, respectively. Dепhasing due to collisions between molecules in different vibrational states of the electronically edited states is not taken into account. The reduced density matrix elements in Eqs.(4) were added to Eqs.(3), which were solved numerically.

In the expression for the external field Eq.(2), the \( \phi \) governs the parity of the chirp. The real and imaginary components of the field \( E(\omega) \), Eq. (1), are shown in Fig.(2) for the sine and cosine modulation and manifest different phase relationship between different components of the field. For the sine modulation (odd chirp), the imaginary part is asymmetric with respect to zero frequency while the real part is symmetric and has components dominantly negative in amplitude. In the case of the cosine modulation (even chirp), both the real and imaginary parts of the spectral field amplitude are symmetric. In any case, a pair of frequencies from the positive and the negative region of the spectrum provides the resultant Raman field having the amplitude that depends on the amplitude difference of two interfering field components. Odd chirp causes cancellation of the field amplitudes being \( \pi \) -shifted with respect to each other, meantime, the even chirp provides the addition of the imaginary components.

3. NUMERICAL RESULTS AND DISCUSSIONS

Spectrally sine modulated pulse train, when interacts with the seven-level system, Fig.(3), produces a peace-wise adiabatic passage of population from the initial Feshbach state and the final ultracold state by a series of coherent pulses. The excited state manifold is insignificantly populated transitionally. For each incident pulse, except for the first one, the “initial condition” of the seven-level system is the one, prepared by the previous pulse. It implies nonzero population of the Feshbach state and all other states. Each pulse in the pulse train transfers a fraction of population from the initial to the final state. For the peak Rabi frequency same as the two-photon transitional frequency \( \omega_{31} \), a complete adiabatic population transfer occurs in 32 pulses. The
The system response resembles the one in the STIRAP control peak Rabi frequency equal to the transition frequency between the photon resonance condition and equals to $\omega$. The dependence of the dressed state energies is obtained numerically. This phenomenon caused qualitatively different response of the seven-level system: population dynamics is shown in Fig.(5) with a significant population accumulation in the transitional, excited bare state manifold insignificantly populated. The system response resembles the one in the STIRAP control scheme \[8\]. To gain insight into dynamics of a stepwise population transfer to the final state within the dark state by the spectrally sine modulated pulse train, we performed the dressed state analysis. We approximated the excited state manifold by a single vibrational state of the excited electronic state for a clear picture of the passage. This is a feasible action because the excited states are all similarly populated according to the exact solution of the Schrödinger equation for the seven-level system. The Hamiltonian of a single pulse interaction with the three-level system in the field interaction representation reads

$$
\hat{H}_d = \begin{bmatrix}
0 & R(t) e^{i(\omega t + M(t))} & 0 \\
R(t) e^{-i(\omega t + M(t))} & 0 & R'(t) e^{iM'(t)} \\
0 & R'(t) e^{-iM'(t)} & 0 
\end{bmatrix}.
$$

Here $R(t) e^{iM(t)} = -\frac{\mu}{\hbar} E_0 \sum_{n=-\infty}^{\infty} J_n(A) e^{-i\omega t_0} e^{-i(\omega t_0)^2/(2\tau^2)} e^{-im\omega t_0}$ and $R'(t) e^{iM'(t)} = -\frac{\mu}{\hbar} E_0 \sum_{n=-\infty}^{\infty} J_n(A) e^{-i\omega t_0} e^{-i(\omega t_0)^2/(2\tau^2)} e^{-im\omega t_0}$. The time dependence of the dressed state energies is obtained numerically for the carrier frequency $\omega_0 = \omega_1$, the spectral modulation parameter $T_0 = 1/\omega_2$, the modulation amplitude $A = 4$ and the peak Rabi frequency equal to the transition frequency between the Feshbach and the ultracold state. A single pulse duration is $\tau = 3$ fs and the three-level pulse train period is $T = 19.2$ ns. The three-level system parameters are $\omega_1 = 309.3$ THz, $\omega_2 = 434.8$ THz, and $\omega_3 = 125.5$ THz \[2\].

For the sine modulation, Fig.(4(a)), dynamics occurs within a single dressed state $|1\rangle$ (blue) performing the adiabatic passage from the initial bare state $|1\rangle$ (dashed blue) to the final bare state $|3\rangle$ (dashed black). In contrast, the cosine modulation of the pulse train does not induce destructive interference of the spectral components actively involved into two-photon Raman transitions, resulting in no quasi-dark excited state generated. This phenomenon caused qualitatively different dynamics of the seven-level system: population dynamics is shown in Fig.(5) with a significant population accumulation in the transitional, excited state manifold. In the dressed state analysis Fig.(4(b)), a generalized adiabatic passage is also performed by a single dressed state $|1\rangle$ (blue). However, the energy of the dressed state $|1\rangle$ (blue) initially goes along with the bare state $|1\rangle$ (dashed blue), then with the excited bare state $|2\rangle$ (dashed black).
Fig. 5. Population dynamics in the seven-level Λ system induced by a cosine-modulated pulse train described by Eq.(2), \( \phi = \pi/2 \). The values of the system parameters are \( \omega_{32} = 410.7 \) THz, \( \omega_{21} = 340.7 \) THz, \( \omega_{31} = 70 \) THz and \( \Delta \omega = 0.1 \omega_{31} \) [6]. The carrier frequency \( \omega_0 = \omega_{32} \), the spectral modulation parameter \( T_0 = 1/\omega_{21} \), the modulation amplitude \( A = 4 \) and the peak Rabi frequency \( \Omega_R = 7 \) THz. A single pulse duration \( \tau = 3 \) fs, pulse train period is \( T = 19.2 \) ns.

Fig. 6. Population dynamics in the seven-level Λ system induced by a sine-modulated (a) and cos-modulated (b) pulse train in the presence of decoherence. The values of the system parameters are \( \omega_{32} = 410.7 \) THz, \( \omega_{21} = 340.7 \) THz, \( \omega_{31} = 70 \) THz and \( \Delta \omega = 0.1 \omega_{31} \) [6]. The carrier frequency \( \omega_0 = \omega_{32} \), the spectral modulation parameter \( T_0 = 1/\omega_{21} \), the modulation amplitude \( A = 4 \) and the peak Rabi frequency \( \Omega_R = 7 \) THz. A single pulse duration \( \tau = 3 \) fs, pulse train period is \( T = 19.2 \) ns.

4. FUNDING INFORMATION
This research is supported by the National Science Foundation under Grant No. PHY-1205454.

REFERENCES
1. T. Lahaye, C. Menotti, L. Santos, M. Lewenstein, and T. Pfau, “The physics of dipolar bosonic quantum gases,” Rep. Prog. Phys. 72, 126401 (2009).
2. K.-K. Ni, S. Ospelkaus, M.H.G. de Miranda, A. Pe’er, B. Neyenhuis, J.J. Zirbel, S. Kotochigova, P.S. Julienne, D.S. Jin, J. Ye, “A high phase-space-density gas of polar molecules”, Science 322, 231 (2008).
3. C. Corder, B. Arnold, X. Hua, H. Metcalf, “Laser cooling without spontaneous emission using the dichromatic force,” J. Opt. Soc. America B 32, B75 (2015).
4. Femtosecond Optical Frequency Comb: Principle, Operation, and Applications, edt. J. Ye, S.T. Cundiff, Springer, (2005).
5. L. Allen, J.H. Eberly, Optical Resonance and Two-Level Atoms, Dover Publications, Inc., New York, 1987.
6. E.A. Shapiro, A. Pe’er, J. Ye, M. Shapiro, “Piecewise adiabatic population transfer in a molecule via a wave packet”, Phys. Rev. Lett., 101, 023601 (2008).
7. T. Bayer, M. Wollenhaupt, H. Braun, T. Baumert, “Ultrafast and Efficient Control of Coherent Electron Dynamics via Spods,” Adv. Chem. Phys. 1 (2014).
8. U. Gaubatz, P. Rudecki, S. Schiemann, K. Bergmann, “Population transfer between molecular vibrational levels by stimulated Raman scattering with partially overlapping laser fields. A new concept and experimental results,” J. Chem. Phys. 92, 5363 (1990).
9. W. Shi, S. Malinovskaya, “Implementation of a single femtosecond optical frequency comb for molecular cooling”, Phys. Rev. A 82, 013407 (2010).
10. S.A. Malinovskaya, S.L. Horton, “Rovibrational cooling using optical frequency combs in the presence of decoherence,” J. Opt. Soc. Am. B 30, 482 (2013).
11. D. Goswami, “Laser pulse modulation approaches towards ensemble quantum computing”, Phys. rev. Lett. 88, 177901 (2002).