Harmonic spectral modulation of an optical frequency comb to control the ultracold molecules formation

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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 sine modulation of the spectral phase of the comb leads to a creation of a quasi-dark dressed state. An insignificant population of the excited state manifold in this dark state provides an efficient way of 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.

Quantum control of ultracold molecules is an exciting new field that has developed in response to recent progress in the formation and manipulation of ultracold molecules. This progress has enabled the preparation of ultracold systems with internal degrees of freedom and exhibiting long-range dipole-dipole interactions [1]. A search for methods to create ultracold molecules nevertheless continues in order to provide viable substitutes to well established control scheme of stimulated Raman adiabatic passage (STIRAP) [2–4]. A crucial factor of efficient control is the ability to mitigate decoherence. 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 and molecular gases and to control open quantum systems. 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 no longer a limiting factor, as opposed to room temperatures [5, 6]. In this letter, we propose a quantum control method for preparation of molecules in an ultracold state using two-photon Raman transitions from the Feshbach state, induced by a spectrally modulated ultrafast pulse train. This method

FIG. 1: A seven-level Λ system modeling the Feshbach state $|1\rangle$, the excited state manifold $|2\rangle$ and the final state $|3\rangle$ in a molecule interacting with a phase-locked pulse train. The transitional frequencies used in calculations are $\omega_{21} = 340.7$ THz, $\omega_{32} = 410.7$ THz, and $\omega_{31} = 70.0$ THz [10]. The $\Delta\omega = 0.1 \omega_{31}$.

was specifically designed to achieve a desired quantum yield in ultracold systems in a time scale faster than spontaneous decay [7, 8].

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 Λ 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 lying molecular 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 manifold of rovibrational states is considered in order to evaluate the effect of dense rovibrational structure on population dynamics induced by a broadband frequency comb. This is in contrast to STIRAP scheme, where a reduction to a three-level system is sufficient.

The sinusoidal modulation of the spectral phase of the incident field is introduced in the

frequency domain as $M(\omega) = e^{-iA \sin(\omega T_0 + \phi)}$ [9]. The spectral profile of the field is

$$E(\omega) = \sqrt{\pi/2} E_0 \tau (e^{-\frac{1}{2}(\omega - \omega_0)^2 \tau^2} + e^{-\frac{1}{2}(\omega + \omega_0)^2 \tau^2}) \cdot e^{-iA \sin(\omega T_0 + \phi)} \sum_{k=0}^N e^{i\omega k T} \quad (1)$$

Here E_0 is the peak field amplitude, τ is the pulse duration, T is the pulse train period, ω_0 is the carrier frequency, A is the modulation amplitude, T_0^{-1} is the modulation frequency, ϕ 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 $M(t) = \sum_{n=-\infty}^{\infty} J_n(A) e^{-in\phi} \delta(t - nT_0)$. The pulse train with such phase modulation reads

$$E(t) = E_0 \sum_{n=-\infty}^{\infty} J_n(A) e^{-in\phi} \sum_{k=0}^N e^{-(t - nT_0 - kT)^2 / (2\tau^2)} \cdot \cos(\omega_0(t - nT_0 - kT)). \quad (2)$$

In Eqs.(1,2), the ϕ governs the parity of the chirp. The real and imaginary components of the field $E(\omega)$ 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. Different symmetry causes different interference pattern in the molecular system leading to a quasi-dark state formation in the case of the odd chirp as opposed to the dynamics induced by the even chirp. The Wigner diagram for the sine and cosine spectrally modulated optical frequency comb, Fig.(3), manifests a qualitatively different pattern depending on the parity of the chirp. It also shows that modulation broadens the spectrum: The spectral bandwidth of modulated comb has ~ 700 THz bandwidth, while unmodulated comb bandwidth is ~ 300 THz in the case of a single pulse duration of 3 fs.

For a description of the time evolution of the seven-level system we refer to the Liouville-von Neumann equation $i\hbar\dot{\rho} = [H_{int}, \rho]$ with relaxation terms that take into account spontaneous decay and elastic collisions. Five excited vibrational states are uncoupled since radiationless 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 Λ systems.

FIG. 2: The real and imaginary components of the field $E(\omega)$, Eq (1), for $\phi = 0$ (a,b) and $\phi = \pi/2$ (c,d).

FIG. 3: The Wigner diagram of the sine (a) and cosine (b) modulated optical frequency comb.

The nearest excited states energy separation is $\Delta\omega$. With respect to the lowest excited state, the separation increases with q as $(q - 1)\Delta\omega$, where q is a state number from 1 to 5. The interaction Hamiltonian reads as $\hat{H}_{int} = \sum_{q=1}^5 \hat{H}_{int}^q$, here $\hat{H}_{int}^q$ describes a single three-level Λ system. The $\hat{H}_{int}^q$ contains terms proportional to the spectral sinusoidal modulation of the field and, in the rotating wave approximation, has two nonzero matrix elements $\hat{H}_{ij}^q = \Omega_R(t)[\exp\{i((\omega_0 - \omega_{ji}) - (q - 1)\Delta\omega)t\}]$, here, i, j are the indexes of the basis set, $i = 1, 2$ and $j = i + 1$, and the Rabi frequency is $\Omega_R(t) = -\mu/\hbar \sum_n J_n(A) E_0 e^{-(t-nT_0)^2/(2\tau_0^2) - in\omega_0 T_0}$, with the peak value Ω_R . The Liouville-von Neumann equation gives a set of coupled differential

equations for the density matrix elements

$$\begin{aligned}
\dot{\rho}_{11} &= 2Im[H_{12}\rho_{21}] \\
\dot{\rho}_{22} &= 2Im[H_{21}\rho_{12} + H_{23}\rho_{32}] \\
\dot{\rho}_{33} &= 2Im[H_{32}\rho_{23}] \\
\dot{\rho}_{12} &= -iH_{12}(\rho_{22} - \rho_{11}) + iH_{32}\rho_{13} \\
\dot{\rho}_{13} &= -iH_{12}\rho_{23} + iH_{23}\rho_{12} \\
\dot{\rho}_{23} &= -iH_{23}(\rho_{33} - \rho_{22}) - iH_{21}\rho_{13}
\end{aligned} \tag{3}$$

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

$$\begin{aligned}
\dot{\rho}_{11})_{sp} &= \gamma_1\rho_{22} & \dot{\rho}_{12})_{sp,col} &= -(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} + \Gamma_1)\rho_{12} \\
\dot{\rho}_{22})_{sp} &= -\gamma_1\rho_{22} - \gamma_2\rho_{22} & \dot{\rho}_{13})_{sp,col} &= -\Gamma_3\rho_{13} \\
\dot{\rho}_{33})_{sp} &= \gamma_2\rho_{22} & \dot{\rho}_{23})_{sp,col} &= -(\frac{\gamma_1}{2} + \frac{\gamma_2}{2} + \Gamma_2)\rho_{23}.
\end{aligned} \tag{4}$$

The decay from the excited states to the initial and the final state is at the rate γ_1 and γ_2 , respectively. Collisions are considered between the molecules in any state within the model and the buffer gas, they are described by three dephasing rates, Γ_1 , Γ_2 and Γ_3 related to dephasing between the excited state manifold and the initial state, that and the final state and the initial state and the final state, respectively. Dephasing 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.

Spectrally sine modulated pulse train, when interacts with the seven-level system, Fig.(4), produces a piecewise 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. Within these states, population is distributed proportionally to the state number q in $\hat{H}_{ij}^q$ and decreases when q increases. 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 the 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 the same as the two-photon transitional frequency ω_{31} , a *complete adiabatic population transfer* occurs in 32 pulses. The system parameters are $\omega_{21} = 340.7$ THz, $\omega_{32} = 410.7$ THz, and $\omega_{31}=70.0$ THz [10], the $\Delta\omega = 0.1\omega_{31}$. The

FIG. 4: Population dynamics in the seven-level Λ system induced by the resonant Raman transitions using a *sine modulated* pulse train described by Eq.(2), ($\phi = 0$.) The values of the system parameters are $\omega_{32} = 410.7$ THz, $\omega_{21} = 340.7$ THz, $\omega_{31} = 70$ THz and $\Delta\omega = 0.1\omega_{31}$ [10]. The carrier frequency is $\omega_0 = \omega_{32}$, the spectral modulation parameter is $T_0 = 1/\omega_{21}$, the modulation amplitude is $A = 4$, the peak Rabi frequency is $\Omega_R = 7$ THz, a single pulse duration is $\tau = 3$ fs, and the pulse train period is $T = 22$ ps.

carrier frequency ω_0 is in the one-photon resonance with the ω_{32} transitional frequency and the reciprocal of spectral modulation parameter T^{-1} is equal to the transitional frequency ω_{21} . The $\omega_0 - T^{-1}$ satisfies the two-photon resonance condition and equals to ω_{31} . Two complementary mechanisms of the resonant, two-photon adiabatic passage take place: The first by frequency difference $\omega_0 - T^{-1}$ and the second by difference of the pairs of optical frequencies in the comb. Detuning of ω_0 and T^{-1} off the one-photon resonance reduces the impact of the first mechanism and results in greater population of the excited state manifold. The second mechanism strongly depends on the period of the pulse train, since the repetition rate determines the one-photon detuning of the optical frequencies [11, 12]. When $\omega_0 - T^{-1} = \omega_{31}$, dynamics occurs within a quasi-dark state with the excited state manifold insignificantly populated. The system response resembles the one in the STIRAP control scheme [3].

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

FIG. 5: Dynamics of the dressed state energies in the three-level Λ system induced by the resonant Raman transitions using a pulse train Eq.(2) with (a) $\phi = 0$ (sin) and (b) $\phi = \pi/2$ (cos) modulation. A pulse duration τ is 3 fs, the peak Rabi frequency is equal to the transition frequency between the Feshbach and the ultracold state, $\Omega_R=70$ THz.

all excited states are 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

$$\hat{H}_d = \begin{bmatrix} 0 & R(t)e^{i(\omega_{31}t+M^-)} & 0 \\ R(t)e^{-i(\omega_{31}t+M^-)} & 0 & R(t)e^{iM^+} \\ 0 & R(t)e^{-iM^+} & 0 \end{bmatrix}. \quad (5)$$

Here $R(t)e^{iM^\pm} = -\frac{\mu}{\hbar}E_0 \sum_{n=-\infty}^{\infty} J_n(A)e^{-\frac{(t-nT_0)^2}{2\tau^2}}e^{in(\pm\omega_0T_0+\phi)}$, $\phi = 0$ for the sine and $\pi/2$ for the cosine modulation. The time dependence of the dressed state energies is obtained numerically for 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 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 $T = 22$ ps. The three-level system parameters are $\omega_{21} = 340.7$ THz, $\omega_{32} = 410.7$ THz, and $\omega_{31}=70.0$ THz [10]. For the *sine modulation*, Fig.(5(a)), dynamics occurs within a single dressed state $|I\rangle$ (blue) performing the adiabatic passage from the initial bare state $|1\rangle$ (dashed blue) to the final bare state $|3\rangle$ (dashed black). Meantime, 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 formation. In the dressed state analysis, Fig.(5(b)), the adiabatic passage is also performed within a single dressed state $|I\rangle$ (blue), but this dressed state contains a significant contribution of the excited state. The energy of dressed state

FIG. 6: 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 $\Delta\omega = 0.1\omega_{31}$ [10]. The carrier frequency is $\omega_0 = \omega_{32}$, the spectral modulation parameter is $T_0 = 1/\omega_{21}$, the modulation amplitude is $A = 4$, the peak Rabi frequency is $\Omega_R = 7$ THz, a single pulse duration is $\tau = 3$ fs, and pulse train period is $T = 22$ ps.

$|I\rangle$ (blue) initially coincides with the bare state $|1\rangle$ (dashed blue), then with the excited bare state $|2\rangle$ (dashed green), and at the end of pulse duration it coincides with the final bare state $|3\rangle$ (dashed black). The oscillations in dressed state energies are due to the $\pi/2$ -phase shift in $R(t)e^{iM^\pm}$ in Eq.(5). *The involvement of all three bare states in the dressed state dynamics manifests itself in the exact solution, Fig.(6), where all the excited states are populated under the conditions of cosine modulation.* The transfer efficiency in this case is 95% during the first Rabi cycle and reduces with time due to population accumulation in the excited state manifold.

In the presence of decoherence, population dynamics in the seven-level system is shown in Fig.(7) induced by the *sine* modulated pulse train. The spontaneous decay rate is $\gamma_1 = \gamma_2 = \gamma = 10^8 \text{ s}^{-1}$ and the collision rate is $\Gamma_i = \Gamma = 10^4 \text{ s}^{-1}$, ($i=1,2,3$), relevant for the ultracold temperature regime [5]. The figure demonstrates a complete population transfer to the ultracold state in controllable way in a time scale much faster than the spontaneous decay. It is accomplished within 700 ps in the framework of the first Rabi cycle.

In summary, a new method for controlled formation of molecules in the ultracold state is proposed using a single optical frequency comb with the spectral modulation. A sequence of pulses that form the frequency comb induce coherent dynamics leading to the complete

FIG. 7: Population dynamics in the seven-level Λ system induced by a sine (a) and cosine 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 $\Delta\omega = 0.1\omega_{31}$ [10]. The carrier frequency is $\omega_0 = \omega_{32}$, the spectral modulation parameter is $T_0 = 1/\omega_{21}$, the modulation amplitude is $A = 4$ and the peak Rabi frequency is $\Omega_R = 70$ THz. A single pulse duration is $\tau = 3$ fs, and pulse train period is $T = 22$ ps. Spontaneous decay rate is $\gamma_{1,2} = 10^8 \text{ s}^{-1}$ and the collision rate is $\Gamma_{1,2} = 10^4 \text{ s}^{-1}, \Gamma_3 = 0$.

population transfer of molecules to the ultracold state. This dynamics is reversible showing Rabi oscillations vanishing due to decoherence within tens of nanoseconds. The method proposes to create ultracold molecules during the first Rabi cycle, in less than one nanosecond, much faster than the spontaneous decay time. The following laser parameters may be used for, e.g., KRb molecule: A single pulse duration 3 fs, the pulse train period 22 ps, the peak Rabi frequency 7 THz, the modulation amplitude $A=4$, the spectral modulation 1/340.7 THz and the carrier frequency 410.7 THz. The parity of the spectral chirp shown to play a key role in the stepwise adiabatic passage to the ultracold state - the odd chirp leads to a quasi-dark state formation providing a foundation to efficiently mitigate decoherence, as opposed to the effect of the even chirp. The incentive for using the proposed method is that vibrational states with different dipole moments are addressed by the comb simultaneously, while the spectral modulation of the comb mitigates decoherence.

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