Feshbach-to-ultracold molecular state Raman transitions via a femtosecond optical frequency comb

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We report on a theoretical study of implementation of a single femtosecond optical frequency comb to perform an efficient transfer of Feshbach molecules to the ground electronic and rovibrational state. A standard optical frequency comb is considered with zero offset frequency and a sinusoidally phase modulated frequency comb, having modulation frequency in the IR region. A stepwise population transfer to the ultracold molecular state is observed, with a negligible population of the excited state in the case of the sinusoidally modulated frequency comb.

Keywords: optical frequency comb; Feshbach molecule; ultracold state; Raman transition

Magnetically tunable Feshbach resonances are used to create loosely bound Feshbach molecules by association of atom pairs in cold temperatures. The methodology is based on magnetic field sweep across zero-energy resonance between the diatomic vibrational bound state and the threshold for dissociation into an atom pair at rest [1]. The technique was applied to produce diatomic molecules constituted of identical atoms, e.g. K₂ [2], Li₂ [3], Cs₂ [4], or of different species of atoms, resulting in the creation of polar molecules, e.g. the KRb [5,6]. The permanent dipole moment associated with polar molecules may be used for studies of long-range interactions in many-body quantum systems. Optically induced transitions in Feshbach molecules is an efficient root to reach the fundamentally lowest state in molecules. In [7], using two narrow-band pulses applied to weakly bound KRb molecules in accordance with the STIRAP scheme, the ultracold gas of polar KRb molecules in the ground singlet or triplet electronic configuration and rovibrational state was created. Recently, a theory on piecewise stimulated Raman adiabatic passage involving Feshbach states was performed using two coherent pulse trains [8,9]. The ultracold KRb molecules were created using the pump-dump stepwise technique that coherently accumulated population in the ground electronic-vibrational state.

An optical frequency comb is a unique tool for high-resolution spectroscopic analysis of internal energy structure and dynamics as well as for controlling ultrafast phenomena in atomic and molecular physics [10–13]. Owing to its broadband spectrum, the frequency comb may efficiently interact with the medium inducing one-photon, two-photon and multi-photon resonances in atoms and molecules. A unique ability of the frequency comb is provided by the presence of about a million optical modes in its spectrum with very narrow bandwidth and exact frequency positions [14]. In this paper, aiming to create deeply bound ultracold molecules from Feshbach states, the control of population dynamics in a molecular system is investigated using a single optical frequency comb with zero offset frequency. The frequency comb is generated by a phase-locked femtosecond pulse train having unmodulated or sinusoidally modulated phase across an individual pulse. We investigate the dynamics of rovibrational cooling on an example of the KRb molecule, which involves the interaction of loosely bound Feshbach KRb molecules with the femtosecond optical frequency comb resulting in the molecular transfer to the ground electronic and rovibrational state. The population dynamics takes place via two-photon Raman transitions between three energy levels separated by THz region frequencies. Coherent accumulation of population in the ultracold KRb state with a negligible population (for the case of the sinusoidally modulated optical frequency comb applied) of the excited state is accomplished by a series of sequential pulses with zero carrier-envelope phase and within the lifetime of the Feshbach KRb molecules, which is about 100 ms [6,15].

A semi-classical model of two-photon Raman transitions induced by a femtosecond optical frequency
efficient stepwise population accumulation in the final state in the λ-system.

The evolution of the density matrix of the λ-system is investigated via a set of coupled differential equations obtained from the Leoville von Neumann equation

\[
\dot{\rho}_{11} = 2 \text{Im}[H_{12}\rho_{21}], \\
\dot{\rho}_{22} = 2 \text{Im}[H_{21}\rho_{12} + H_{23}\rho_{32}], \\
\dot{\rho}_{33} = 2 \text{Im}[H_{32}\rho_{23}], \\
\dot{\rho}_{12} = -iH_{12}(\rho_{22} - \rho_{11}) + iH_{32}\rho_{13}, \\
\dot{\rho}_{13} = -iH_{12}\rho_{23} + iH_{23}\rho_{12}, \\
\dot{\rho}_{23} = -iH_{23}(\rho_{13} - \rho_{22}) - iH_{21}\rho_{13}.
\] (2)

Two nonzero matrix elements of the interaction Hamiltonian are \(H_{ij} = \Omega_R(t - T)[\exp\{-i((\omega_L + \omega_R) \times (t - T))\} + \exp[i((\omega_L - \omega_R)(t - T))]\], here, \(i, j\) are the indexes of the basis set, \(i = 1,2\) and \(j = i+1\), \(\Omega_R(t - T) = \Omega_R \exp\{-(t - T)^2/(2\tau^2)\}\) is the Rabi frequency, and \(\Omega_R\) is the peak value of the Rabi frequency. Calculations are done beyond the rotating wave approximation.

For the λ-system, we apply a set of parameters that correspond to data on molecular cooling of loosely bound KRb molecules from the Feshbach states presented in [7]. Experimental schemes used in [7] involve a transition to the ground electronic triplet or the ground electronic singlet state with zero rovibrational quantum number. These states are achieved by implementing the STIRAP control scheme with a pair of pulses having a narrow bandwidth and being in resonance with the transitions between three states involved in the process. These are the Feshbach state, the 2Σ electronically excited state, and the triplet or singlet ground electronic state with zero rotational and vibrational quantum number. In our model we implement parameters that correspond to the experiment involving the singlet ground electronic state thus addressing a fundamentally cold molecule. The parameters of the λ-system are \(\omega_{13} = 309.3\ \text{THz}, \ \omega_{23} = 434.8\ \text{THz}\), making the frequency of two-photon transition \(\omega_{13}\) to be equal to 125.5 THz. The carrier frequency of the pulse train \(\omega_L\) is chosen to be in resonance with the one-photon transition frequency \(\omega_{32}\) in the λ-system, and the multiples of the radio frequency provide two-photon resonances when the condition \(\omega_L - n\omega_r = \omega_{31}\) is satisfied, where \(n\) is an integer number. The peak Rabi frequency is \(\Omega_R = 0.10\omega_{31} = 1.255\ \text{THz}\), the pulse duration \(\tau\) is 3 fs, and the pulse train period is \(T = 6.4 \times 10^5\ \tau\), (2 ns), giving the radio frequency 500 MHz. For given peak Rabi frequency, the evaluated peak field amplitude is \(10^8 - 10^9\ \text{V cm}^{-1}\).
The system levels addressed in [9] include the final state as the ground electronic singlet state with vibrational quantum number $v = 22$ which suggests the following parameters for the $\lambda$-system: the transitional frequency from the initial to the excited electronic state $\omega_{31} = 340.7$ THz, and the transition frequency from the excited electronic state to the final state $\omega_{32} = 410.7$ THz. It makes the initial-to-final state frequency difference $\omega_{31} = 70$ THz. The carrier frequency and the modulation frequency of the pulse train

\[ E(t) = \sum_{k=0}^{N-1} E_0 \exp\left(-\left(t - kT^*\right)^2/(2\tau^2)\right) \cos(\omega_0(t - kT)) + \Phi_0 \sin(\Omega(t - kT)). \]

Here, $\Omega$ is the modulation frequency, and $\Phi_0$ is the modulation amplitude. The carrier-envelope phase is zero in the pulse train resulting in zero offset frequency of the generated optical frequency comb. The time-dependent phase across each pulse in the form of the $\sin$ function enriches the frequency comb power spectrum with new peaks compared to a standard optical frequency comb discussed above. More specifically, laser frequency $\omega_L$ determines the center of the frequency comb, and the sinusoidal modulation forms the sidebands at multiples of $\Omega$ with the amplitude dictated by $\Phi_0$.

Numerical analysis of the population dynamics in the $\lambda$-system was done using Equations (2) with the Hamiltonian having two nonzero matrix elements $H_f = \Omega_0(t - T)[\exp\{-i(\omega_1 + \omega_0)(t - T) + M(t - T))\} + \exp[i(\omega_1 - \omega_0)(t - T) + M(t - T)]$, here $i = 1, 2$ and $j = i + 1$, and $M(t - T) = \Phi_0 \sin(\Omega(t - T))$ is the phase modulation in a single pulse. The carrier frequency of the pulse train $\omega_L$ is chosen to be equal to the $\omega_{32}$, and the modulation frequency $\Omega$ to the $\omega_{31}$, so, that the field frequencies are in resonance with the one-photon transitions in the three-level $\lambda$-system which automatically satisfies the condition for the two-photon Raman resonance. Additionally, the modes that are multiples of the radio frequency $f_r$ provide pairs of frequencies that differ by exactly the transition frequency $\omega_{31}$. These lead to an efficient stepwise population accumulation in the final state in the $\lambda$-system. Parameters of the $\lambda$-system used in these calculations were taken from the paper [9].

The results of population transfer are presented in Figure 2. A stepwise adiabatic population transfer is observed from the initial |1⟩ (black) to the final |3⟩ state (green) via the transitional state |2⟩ (red) which gets populated to up to 45%. Each pulse brings a fraction of population to the final state and contributes to the accumulative effect. Total population transfer occurs in 460 ns after 242 sequential pulses, which is within the lifetime of the Feshbach KRb molecules. Next 242 pulses return population to the initial state, the system returns to the initial conditions and, then, the dynamics repeats. Within the lifetime of the Feshbach states, it is possible to transform the medium from highly vibrationally excited molecules to the ultracold state and back. Notably, the detuning of the carrier frequency off resonance with the $\omega_{32}$ gives very similar dynamics of population transfer to the ultracold molecular state and back as in the resonance case with a difference being some reduction in the number of pulses needed to accomplish this transfer. The increase in the strength of the electric field decreases the number of pulses and the overall time duration needed for the full population transfer, preserving the quality of the dynamics picture. Decreasing the pulse repetition rate elongates the duration of the coherent accumulation dynamics, however, this does not change the number of required pulses (given other field parameters are preserved). Since a single pulse duration determines the bandwidth of the comb, the shorter it is the more efficient excitation of the Raman transitions takes place.

We performed the phase modulation across an individual pulse in the pulse train in the form of a sinusoidal function. In previous works, the sinusoidal modulation in the THz region was applied to the carrier frequency, e.g., to study absorption resonances in $I_2$ with high precision in [17]. A general form of a phase modulated pulse train reads

\[ E(t) = \sum_{k=0}^{N-1} E_0 \exp\left(-\left(t - kT^*\right)^2/(2\tau^2)\right) \cos(\omega_0(t - kT)) + \Phi_0 \sin(\Omega(t - kT)). \]

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train are in resonance with $\omega_{32}$ and $\omega_{21}$, respectively; the peak Rabi frequency is equal to $\omega_{31}$, $\Phi_0$ is equal to 4 (the discussion of this choice will be given below), a single pulse duration $\tau$ is 3 fs and the pulse train period $T$ is $6400 \tau$ (about 20 ps). The modulation of the carrier frequency having value 410.7 THz is to be done at frequency 340.7 THz. To achieve the needed modulation, an approach, described in [18], on efficient generation of a Raman-type optical frequency comb in an enhancement cavity, may be applied. The technique provides the whole comb bandwidth covering 300–900 THz.

The results of the numerical calculations, Figure 3, show a smooth stepwise population transfer from the initial to the final state with a negligible population of the excited state, in analogy to the STIRAP stepwise scheme proposed by Shapiro et al. [9], however, performed with a single optical frequency comb. In our opinion, making use of a single pulse train, in contrast to the two-pulse train scheme in [9], significantly simplifies the experimental conditions for cooling of rovibrational degrees of freedom in molecules. Total population transfer occurs after 109 sequential pulses and, thus, is accomplished in 2.5 ns.

Notably, there is a strong dependence of the efficiency of population transfer on the value of the amplitude $\Phi_0$ of sinusoidal modulation of the phase across an individual pulse in the pulse train. For the resonant excitation, population dynamics was calculated using different values of the parameter $\Phi_0$, however, for the values of $\Phi_0$ from 1 to 9, only $\Phi_0 = 4$ gave the desired population transfer to the cold state. To get insight into the mechanism of the frequency comb–system interaction leading to a successful cooling, we made a Fourier transform both analytically and numerically to reveal its spectral properties linking to the molecular system resonances.

The Fourier transform reads

$$E(\omega) = (E_0 \tau) \left/ \sum_{n} J_n(\Phi_0) \exp \left(-1/2(\omega_L + n\Omega - \omega)^2 \tau^2\right) \times \sum_{k} \exp(iokT) \right.$$

where $J_n(\Phi_0)$ is the Bessel function of the order $n$ and $\Phi_0$ is the modulation index. When multiplied by $\exp(-1/2(\omega_L + n\Omega - \omega)^2 \tau^2)$, it determines the shape of the power spectrum of the optical frequency comb.

Depending on the value of $\Phi_0$, the power spectrum has a different number of maxima as seen in Figure 4 which shows the envelope of the power spectrum for $\Phi_0 = 3, 4, 5, 8$. The increase in modulation index brings additional, intense peaks of modes into the spectrum and broadens it. These maxima are located at different frequencies for different values of $\Phi_0$ affecting the population dynamics in the $\lambda$-system.

The power spectrum of the pulse train with the modulation amplitude $\Phi_0 = 4$ has three maxima, the

![Figure 3. Population transfer in the three-level $\lambda$-system, achieved via the resonant Raman transitions using a phase-modulated optical frequency comb described by Equation (3). The values of the parameters are the carrier frequency $\omega_L = 410.7$ THz, the modulation frequency $\Omega = 340.7$ THz, the modulation amplitude $\Phi_0 = 4$, and the peak Rabi frequency $\Omega_R = 70$ THz. Stepwise, adiabatic accumulation of the population is observed in state [3] (green), which is the ultracold KRb state. The population of the Feshbach state [1] (black), comes gradually to zero, while the excited state manifold [2] (red) is slightly populated during the transitional time. Full population transfer is accomplished in 109 pulses. Time is given in the units of $[\omega^{-1}]$, where $\omega = \omega_{31} = 70$ THz. (The color version of this figure is included in the online version of the journal.)](image)

![Figure 4. The envelope of the power spectrum of the optical frequency comb described by Equation (3) (no dense radio frequency comb lines are included). The pulse train parameters are the effective Rabi frequency $\Omega_R = 70$ THz, the carrier frequency $\omega_L = 410.7$ THz, the modulation frequency $\Omega = 340.7$ THz, $\tau = 3$ fs, and the modulation amplitude $\Phi_0 = 3$ (red), $\Phi_0 = 4$ (green), $\Phi_0 = 5$ (blue) and $\Phi_0 = 8$ (black). (The color version of this figure is included in the online version of the journal.)](image)
highest one is located at \( \omega = 4.9 \) (which is in resonance with the \( \omega_{21} \)), making the pulse train with \( \Phi_0 = 4 \), an optimal one for coherent accumulation of the population in the final state of the \( \lambda \)-system. It provides full population transfer to the ground electronic-vibrational state and, thus, cooling the KRb Feshbach molecule. The numerical result of the fast Fourier transform of Equation (3) with \( \Phi_0 = 4 \), Figure 5, shows a power spectrum of the optical frequency comb with the fine structure of the radio-frequency spaced modes appearing as a solid color under the envelope. The parameters of the phase-locked pulse train, needed to accomplish molecular cooling from Feshbach states, have to be chosen based on the analysis of the power spectrum of the sine-phase modulated optical frequency comb and the energy levels involved in the dynamics of the molecular system. The offset frequency does not necessarily have to be zero. Its nonzero value does not affect the two-photon resonance condition, while, for one-photon transitions, it induces a detuning. The field carrier frequency detuned off resonance with the one-photon transition in the \( \lambda \)-system leads to the same quantum yield on a shorter time scale. Alternatively, the detuning may be compensated by adjusting the radio frequency to keep the frequency comb modes in resonance with the \( \lambda \)-system.

In conclusion, we have demonstrated a coherent population transfer from a loosely bound Feshbach state to the ultracold molecular state using a single femtosecond, optical frequency comb. We studied the phenomenon on an example of the KRb molecule, modelled by a three-level \( \lambda \)-system with the energy levels taken from [7,9]. Coherent accumulation of the population in the ground electronic and rovibrational state is achieved by applying a standard optical frequency comb with zero offset frequency, or an optical frequency comb generated by a pulse train with a phase modulation in the form of the sinusoidal function across an individual pulse. The mechanism of the accumulative effect leading to full population transfer is based on the excitation of the two-photon Raman resonances by pairs of optical frequency modes with the frequency difference matching to the Feshbach-to-ultracold molecular state transition. In the case of sinusoidally modulated optical frequency comb, the Raman transitions are stimulated by the optical frequency comb generated by a pulse train with the frequency difference matching to the Feshbach-to-ultracold molecular state transition. In the case of sinusoidally modulated optical frequency comb, the Raman transitions are stimulated by the optical frequency comb.

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