Implementation of a single femtosecond optical frequency comb for rovibrational cooling

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Abstract

We show that a single femtosecond optical frequency comb may be used to induce two-photon transitions between molecular vibrational levels to form ultracold molecules, e.g., KRb. The phase across an individual pulse in the pulse train is sinusoidally modulated with a carefully chosen modulation amplitude and frequency. Piecewise adiabatic population transfer is fulfilled to the final state by each pulse in the applied pulse train providing a controlled population accumulation in the final state. Detuning the pulse train carrier and modulation frequency from one-photon resonances changes the time scale of molecular dynamics but leads to the same complete population transfer to the ultracold state. A standard optical frequency comb with no modulation is shown to induce similar dynamics leading to rovibrational cooling.
An optical frequency comb has been recognized as a new and 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 \[1-4\]. Owing to its broad-band spectrum, the frequency comb may efficiently interact with the medium inducing one-photon, two-photon and multi-photon resonances between finely structured energy levels. A unique ability of the frequency comb is provided by the presence of about a million of optical modes in its spectrum with very narrow bandwidth and exact frequency positions \[5\]. During last years, the investigations have been carried out on implementation of a femtosecond frequency comb to manipulate ultracold gases. The pioneering works in quantum control in ultracold temperatures include the two-photon excitation of specific atomic levels forming four-level diamond configuration in cold $^{87}\text{Rb}$ using a phase modulated, femtosecond optical frequency comb, \[6\], and a theory on piecewise stimulated Raman adiabatic passage performed with two coherent pulse trains possessing the pulse-to-pulse amplitude and chirped phase variation, \[7, 8\]. In these papers, the authors reported on creation of ultracold KRb molecules from Feshbach states, the highest excited vibrational states of the ground electronic state, using the pump-dump stepwise technique that coherently accumulates population in the ground vibrational state. Experimentally, a dense quantum gas of ultracold KRb polar molecules was produced from the Feshbach molecules using the STIRAP scheme with two $\mu$s pulses, \[9\]. Coherent population transfer was demonstrated to rovibrational ground state of the triplet and singlet electronic ground potential.

In this paper, we demonstrate that a single, phase modulated optical frequency comb may be used to control population dynamics aiming creation of deeply bound ultracold molecules from Feshbach states. We investigate the KRb rovibrational cooling, that involves the interaction of loosely bound KRb molecules with a single femtosecond optical frequency comb resulting in population flow from the Feshbach state to the ultracold state. The population dynamics takes place via the two-photon Raman transitions that involve primary three energy states separated in THz region. The efficient Raman transitions may occur resonantly and also when the carrier frequency and the modulation frequency of the laser field are detuned off resonance with the one-photon transition frequency between electronic-vibrational states. Coherent accumulation of population in the cold KRb state with a negligible population of the excited state is accomplished by a well defined number of sequential pulses with zero carrier-envelope phase and within the lifetime of the Feshbach KRb molecules, which
is known to be about 100 ms, [10, 11].

A semi-classical model of two-photon Raman transitions, induced by a femtosecond optical frequency comb in a three-level \( \lambda \) system, describes the cooling process of the internal degrees of freedom initiated in Feshbach molecules. Feshbach molecules can be created by association of ultracold atom pairs performed by magnetic field sweep across zero-energy resonance between the diatomic vibrational bound state and the threshold for dissociation into an atom pair at rest, [12]. They are the key intermediates in the process of creation of deeply bound molecules in the ground electronic configuration.

A frequency comb known to be characterized by two key parameters, the radio-frequency \( f_r \), determined by the pulse repetition rate and specifying the spacing between modes, and by the carrier-envelop-offset frequency \( f_0 = f_r \Delta \phi_{ce}/(2\pi) \), here \( \Delta \phi_{ce} \) is the carrier-envelope phase. Both parameters, the \( f_0 \) and the \( f_r \), were efficiently used to manipulate dynamics in, e.g., [6], to resonantly enhance two photon transitions in cold \( ^{87}Rb \) atoms. Here, the control scheme is simplified by not involving \( f_0 \). We make \( f_0 \) equal to zero by implying zero carrier-envelope phase in each pulse in the pulse train.

The beam of a cw ring dye laser, having a sinusoidal modulation at the MHz frequency, was implemented in [13] to study absorption resonances in \( I_2 \) with high precision. Following the idea of sin-phase modulation, we create a femtosecond pulse train, having the sinusoidal modulation at THz frequency across an individual pulse, aiming to induce two-photon Raman transitions. The general form of the pulse train reads

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

(1)

Here \( k \) is an integer number and \( T \) is the period of the pulse train. \( T \) is chosen to be much greater than a single pulse duration in order for the wings of each pulse to reduce to zero before next pulse arrives. The time-dependent phase across each pulse in the form of the \( \sin \) function enriches the frequency comb spectrum with new peaks compared to a standard frequency comb. More specifically, laser frequency \( \omega_L \) determines the center of the frequency comb, while sinusoidal modulation forms the sidebands at multiples of \( \Omega \) with the amplitude dictated by \( \Phi_0 \). The fine structure of the optical broadband comb is owing to the modes equally spaced by the radio frequency \( 2\pi T^{-1} \). We apply a single, sin-phase modulated optical frequency comb to induce Raman resonances in the three-level \( \lambda \)-system aiming full population transfer from the initial state \( |1> \) through intermediate state \( |2> \).
FIG. 1: The three-level $\lambda$-system modeling the molecular energy levels, involved into cooling dynamics. State $|1\rangle$ is the Feshbach state, state $|2\rangle$ is the transitional, electronic excited state, and state $|3\rangle$ is the cold molecular state.

to the final state $|3\rangle$, Fig. 1. We assume that state $|1\rangle$ is the Feshbach state, state $|2\rangle$ is the transitional, electronic excited state, or state manifold, and state $|3\rangle$ is the ultracold molecular state. For Feshbach molecules, the energy splitting is large enough to justify the validity of three-level model for the description of their interaction with the frequency comb aiming to coherently create ultracold molecules.

Parameters of the $\lambda$-system correspond to data on molecular rovibrational cooling of loosely bound KRb molecules from the Feshbach states, discussed in [8]. Coherent population accumulation was achieved in the deeply bound ground electronic state with vibrational quantum number $v=22$. The related parameters of the molecular system, implemented in our model, are $\omega_{21}=340.7$ THz, $\omega_{32}=410.7$ THz, and $\omega_{31}=70$ THz. In the discussion below, all frequency and time parameters will be given in the units of $[\omega_{31}]$ or $[\omega_{31}^{-1}]$ for convenience of interpretation, then, e.g., $\omega_{21} = 4.9$, and $\omega_{32} = 5.9$. The field frequencies are in resonance with the one-photon transitions in the three-level $\lambda$-system, the carrier frequency $\omega_L = \omega_{32}$, and the modulation frequency $\Omega = \omega_{21}$. The peak Rabi frequency is $\Omega_R = \omega_{31}$, the pulse duration is $\tau = 0.25$, (3 fs), and the pulse train period is $T = 6400\tau$, (20 ps), giving the radio frequency $f_r = 6.25 \cdot 10^{-4}$, (43 GHz). A single pulse area is estimated to be $0.2\pi$. The modulation of the carrier frequency having value 410.7 THz assumed to be done at frequency 340.7 THz. To achieve IR modulation, an approach, described in [14], on efficient generation of a Raman-type optical frequency comb in an enhancement cavity may be applied. The technique provides with the whole comb bandwidth covering 300-900 THz.
The evolution of the density matrix was investigated using the Leoville von Neuman equation with the interaction Hamiltonian having two nonzero matrix elements $H_{ij} = \Omega_R(t-T)[\exp\{-i((\omega_L + \omega_{ji})(t - T) + M(t - T))\} + \exp\{i((\omega_L - \omega_{ji})(t - T) + M(t - T))\}]$, here, i,j are the indexes of the basis set, $i = 1, 2$ and $j = i + 1$, $M(t - T) = \Phi_0 \sin \Omega(t - T)$ is the modulation of the phase, $\Omega_R(t - T) = \Omega_R \exp \left(\frac{-(t - T)^2}{(2\tau^2)}\right)$ is the Rabi frequency, and $\Omega_R$ is the peak value of the Rabi frequency. Calculations were done beyond the rotating wave approximation, that allows for flexible values of the transition frequencies relative to the laser carrier frequencies to be considered. To get zero value of the carrier-envelope phase, the temporal variation is taken in the form of (t-T), where T is the pulse train period. It guarantees the envelope maximum to coincide with the peak value of the amplitude of the electric field. The condition for the Raman resonance is satisfied by making the difference of laser field frequency components ($\omega_L - \Omega$) equal to the two-photon transition frequency $\omega_{31}$. Additionally, the modes that are multiples of the radio frequency $f_r$ provide with pairs of optical 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. The results of population transfer are presented in the Fig. for the parameters: The peak Rabi frequency $\Omega_R=1$, the carrier frequency $\omega_L=5.9$, the modulation frequency $\Omega=4.9$, the pulse duration $\tau = 0.25$, and the modulation amplitude $\Phi_0 = 4$. The $\lambda$-system transition frequencies are $\omega_{31}=4.9$, and $\omega_{32}=5.9$. Adiabatic population transfer is achieved from the initial $|1\rangle$ to the final $|3\rangle$ state via the transitional state $|2\rangle$, which is insignificantly populated. A negligible population of the intermediate state is favorable for minimizing spontaneous emission losses. Each pulse brings a fraction of population, ($\sim 1\%$), to the final state and contributes to the accumulative effect. Total population transfer occurs after 112 sequential pulses and is accomplished in 2.5 ns, which is within the lifetime of the Feshbach KRb molecules.

When the carrier frequency $\omega_L$ and the modulation frequency $\Omega$ of the optical frequency comb are detuned off resonance with the transitional frequencies $\omega_{31}$ and $\omega_{21}$ respectively, the stepwise adiabatic population transfer takes place in a fashion similar to the resonance case. The time evolution of population is presented in the Fig. for parameters $\Omega_R=1$, the carrier frequency $\omega_L=5.4$, the modulation frequency $\Omega=4.4$, and the modulation amplitude $\Phi_0 = 4$; the $\lambda$-system transition frequencies are $\omega_{21}=4.9$, and $\omega_{32}=5.9$, giving $\delta = \omega_{31}/2$. Figure shows the coherent accumulation of the population in the final state, taking place
FIG. 2: Population transfer in the three-level $\lambda$-system, achieved via the resonant Raman transitions and using an optical frequency comb as in Eq. (1) having the carrier frequency and the modulation frequency in resonance with one-photon transitions in the $\lambda$-system. The values of the parameters are the carrier frequency $\omega_L=5.9$, (410.7 THz), the modulation frequency $\Omega=4.9$, (340.7 THz), the modulation amplitude $\Phi_0=4$, and the peak Rabi frequency $\Omega_R=1$, (70THz); the system one-photon transition frequencies are $\omega_{21} = \Omega$, $\omega_{32} = \omega_L$. Stepwise, adiabatic accumulation of the population is observed in state $|3\rangle$, (green), which is the ultracold KRb state. The population of the Feshbach state $|1\rangle$, (black), reduces gradually to zero, while the excited state manifold $|2\rangle$, (red), is slightly populated during the transitional time. Full population transfer is accomplished in 112 pulses.

within 42 pulses and accomplished in about 1 ns. However, the population of the transitional, electronic excited state is substantial, almost 50%. Compared to the resonance case, the population dynamics is faster with less number of pulses in the pulse train involved into full population transfer.

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 individual pulse in the pulse train. For the resonant excitation, population dynamics was calculated using different values of the parameter $\Phi_0$, the results are presented in Fig. 4 for $\Phi_0 = 3, 5, 8$. The desired adiabatic accumulation of the population in the final state is not achieved for any of these values of $\Phi_0$. To understand the impact of the modulation amplitude $\Phi_0$, we analyzed the
FIG. 3: Population transfer in the three-level $\lambda$-system, achieved using an optical frequency comb as in Eq.(1) with the field parameters detuned off resonance with the frequencies of the $\lambda$-system. The detuning $\delta$ is equal to $\omega_{31}/2$. The carrier frequency is $\omega_L=5.4$, the modulation frequency is $\Omega=4.4$, the Rabi frequency is $\Omega_R=1$, and the modulation amplitude is $\Phi_0=4$. The $\lambda$-system transition frequencies are $\omega_{21}=4.9$, $\omega_{32}=5.9$ (in the unites of frequency $\omega_{31}=70THz$). Full population transfer to the final, cold state, (green), occurs within 42 pulses. During this time, population of the initial Feshbach state, (black), reduces to zero and the excited state, (red), gets substantially populated during the transitional time. The population is reversed by the next 42 pulses.

Fourier transform of the pulse train in Eq.(1). The Fourier transform reads

$$E(\omega) = (E_0\tau)/2\Sigma_n J_n(\Phi_0) \exp(-1/2(\omega_L + n\Omega - \omega)^2\tau^2) \cdot \Sigma_k \exp(i\omega_k T).$$

(2)

Here, $J_n(\Phi_0)$ 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 different number of maxima as it is seen in Fig.5 for $\Phi_0 = 3, 4, 5, 8$. The increase in modulation index brings additional, intense peaks of modes into spectrum and broaden 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 highest one is 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 and rovibrational state and, thus, cooling the internal degrees of freedom.
FIG. 4: Population dynamics in three-level $\lambda$-system, reached using an optical frequency comb as in Eq. (1) with different values of the modulation amplitude equal to $\Phi_0=3.$ (a), $\Phi_0=5.$ (b), $\Phi_0=8.$ (c). Other parameters are the effective Rabi frequency $\Omega_R=1,$ (70THz), the carrier frequency $\omega_L=5.9,$ (410.7 THz), and the modulation frequency $\Omega=4.9,$ (340.7 THz). For all three values of $\Phi_0$, there is only partial population transfer to the final state $|3>$, (green), unlike to the solution for $\Phi_0 = 4.$ in Fig. (2).

FIG. 5: The envelope of the power spectrum of the optical frequency comb as in Eq. (1), (no dense radio frequency comb lines are included). The pulse train parameters are the effective Rabi frequency $\Omega_R=1,$ (70THz), the carrier frequency $\omega_L=5.9,$ (410.7 THz), the modulation frequency $\Omega=4.9,$ (340.7 THz), and the modulation amplitude $\Phi_0=3.$ (red), $\Phi_0=4.$ (green), $\Phi_0=5.$ (blue), $\Phi_0=8.$ (black).
in the KRb Feshbach molecule. Thus, the parameters of the phase stabilized pulse train, needed to accomplish rovibrational cooling from Feshbach states, have to be chosen based on the analysis of the power spectrum of the sin-phase modulated optical frequency comb and the energy levels involved into dynamics of the molecular system.

Lowering the value of the Rabi frequency, which corresponds to decrease in the field intensity, gives qualitatively similar results of coherent, adiabatic accumulation of population in the final, ultracold state leading to full population transfer. The difference is in the time scale of the dynamics which becomes an order of magnitude longer, as calculations show, however, is well within the lifetime of the Feshbach molecules. Elongation of duration of population dynamics is observed also with the decrease in the pulse repetition rate. Due to computational limitations on the propagation time, we use the values of the parameters of the Rabi frequency and pulse train repetition rate that demonstrate the phenomenon on a shorter time scale. Close to experimental values parameters of the field intensity and the repetition rate are implemented to the standard optical frequency comb interaction with the three-level $\lambda$-system, described next.

We investigated the interaction of a single, standard optical frequency comb with the three-level $\lambda$-system. The standard femtosecond optical frequency comb is formed by a phase-stabilized pulse train without phase or amplitude modulation across an individual pulse. The carrier-envelope phase is made to be zero. The pulse train reads

$$E(t) = \Sigma_{k=0}^{N-1} E_0 \exp(- (t - kT)^2 / (2 \tau^2)) \cos(\omega_L(t - kT)).$$  \hspace{1cm} (3)

Here, $T$ is the pulse train period, $\tau$ is the pulse duration, $\omega_L$ is the carrier frequency. 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 $\lambda$-system. The two-photon resonances in the $\lambda$-system are provided by the pairs of optical frequencies present within the frequency comb that are multiples of the radio frequency and satisfy the two-photon resonance condition $mf_r - nf_r = \omega_L - n'f_r = \omega_{31}$, here $m, n, n'$ are integer numbers. Calculations were performed for two values of the peak Rabi frequency: $\Omega_R = 0.1\omega_{31}$ and $\Omega_R = 0.01\omega_{31}$, they determine the electric field intensity in the range from $10^{14}$ to $10^{12}$ W/cm$^2$. A single pulse area is estimated to be $0.03\pi$ to $0.003\pi$ respectively. Two values of the pulse train period were considered: $T = 6.4 \cdot 10^4\tau=0.2$ ns, (5 GHz), and $T = 6.4 \cdot 10^5\tau=2$ ns, (500 MHz), here the pulse duration $\tau$ is 3 fs.
FIG. 6: Population transfer in the three-level $\lambda$-system using a standard optical frequency comb characterized by the radio-frequency equal to 5 GHz and zero offset frequency. Black curve shows population dynamics of the Feshbach state, red - same for the excited electronic state, green - same for the final, ultracold state. Parameters of the pulse train are the carrier frequency $\omega_L = \omega_{32}=434.8$ THz, and the pulse duration $\tau_0=3$ fs.

For the $\lambda$-system, we apply a set of parameters that correspond to experimental data on molecular rovibrational cooling of loosely bound KRb molecules from the Feshbach states presented in [9]. In the described experiments, the STIRAP scheme was implemented to coherently transfer population from Feshbach states to the ground electronic triplet or the ground electronic singlet state with zero rovibrational quantum number. These deeply bound states were achieved with two $\mu$s pulses that were in resonance with the one-photon transitions between the molecular states involved into the process, which are the Feshbach state, the $2^3\Sigma$ electronically excited state, and the triplet or singlet ground electronic states. In our model, we address the fundamentally cold state of the KRb molecule by using parameters that correspond to the experiment involving the singlet ground electronic state. These parameters are the $\omega_{21}=309.3$ THz, and the $\omega_{32}=434.8$ THz, giving the frequency of the two-photon transition $\omega_{31}=125.5$ THz. Fig.6 shows the population dynamics in the $\lambda$-system for $\Omega_R = 0.1\omega_{31}=12.56$ THz and $T=0.2$ ns, which results in full population transfer to the ultracold state within 238 pulses during 47.6 ns. Next 238 pulses restore full population in the initial state $|1\rangle$ - the Feshbach state - and the dynamics repeats. Within the lifetime of the Feshbach state, it is possible to transform the medium from highly vibrationally excited...
molecules to the ultracold configuration and back using a single, phase-locked pulse train. Note, that qualitatively same result was obtained for the pulse train repetition rate 500 MTz, \((T = 2\text{ns})\), with the only difference in the dynamics duration, which was extended by an order of magnitude to 440 ns.

Off-resonant Raman transitions are readily induced by a single, standard optical frequency comb, provided there are optical frequencies in the comb that satisfy the two-photon resonance condition and differ by exactly the Feshbach-to-ultracold state transitional frequency. In Fig.7 the case for the carrier frequency detuned off resonance with the \(\omega_{32}\) transitional frequency is presented for detuning \(\delta = 3/2\omega_{31}\) and two values of the Rabi frequency, the \(\Omega_R = 0.1\omega_{31} = 12.55\text{THz}\) and \(\Omega_R = 0.01\omega_{31} = 1.26\text{THz}\) related to the field intensity on the order of \(10^{14}\) and \(10^{12}\ W/cm^2\) respectively. The chosen value of detuning \(\delta\) is about \(\sim 10^{14}\text{Hz}\). Bold black, bold red, and bold green lines show population dynamics in the Feshbach state, the excited electronic state and the final, ultracold state respectively, for the Rabi frequency \(\Omega_R = 1.26\text{THz}\), thin lines - same for the Rabi frequency \(\Omega_R = 12.55\text{THz}\). Since here we demonstrate the effects of the detuning, and the impact of different field intensity on the population dynamics to the deeply bound rovibrational state in the KRb molecule, the pulse train period is chosen to be short to accommodate the computational limitations, \(T = 6400\tau = 0.02\text{ns}\). (Same as for the resonance case, pulse train period in 2 ns elongates the duration of population accumulation.) Figure Fig.7 shows that almost complete population transfer, \((\sim 98\%)\), takes place when the carrier frequency is off-resonance with the one-photon transitions in the molecular system. It also demonstrates the field intensity dependence of the duration needed to accumulate population in the ultracold state. For the \(\Omega_R=12.55\ \text{THz}\), \((10^{14} W/cm^2)\), the population transfer dynamics is accomplished five times faster than for the \(\Omega_R=1.26\ \text{THz}\), \((10^{12} W/cm^2)\).

In conclusion, we have demonstrated a possibility of molecular rovibrational cooling from the Freshbach states using a single femtosecond, optical frequency comb. We focused on the KRb molecule and modeled it by a three-level \(\lambda\)-system with the energy levels taken from [8] and [9]. Coherent accumulation of the population in the cold state is achieved by applying a standard optical frequency comb with zero offset frequency, and by an optical frequency comb generated by a pulse train with a phase modulation in the form of the sinusoidal function across an individual pulse. An optimal dynamics takes place when the carrier-envelope phase is equal to zero, \((f_0 = 0)\). The mechanism of the accumulative effect leading
FIG. 7: Population transfer in the three-level λ-system, achieved using an optical frequency comb as in Eq. (3) with the carrier frequency of the electric field detuned off resonance with the $\omega_{32}$ in the λ-system. The detuning $\delta$ is $3/2\omega_{31}$. Bold black, bold red, and bold green lines show population of the Feshbach state, the excited electronic state and the final, ultracold state for the $\Omega_R=1.26$ THz, ($10^{12} \text{ W/cm}^2$), thin lines show respective populations for $\Omega_R=12.55$ THz, ($10^{14} \text{ W/cm}^2$). Pulse train period is $T = 6400\tau = 0.02\text{ns}$.

To full population transfer is based on the excitation of the two-photon Raman resonances. For both, the resonant and detuned Raman transition to happen, the radio frequency $f_r$ characterizing the optical frequency comb has to be such that, multiplied by integers $n$ and $m$, it forms the modes at the pump and Stokes frequencies, whose difference, $nf_r - mf_r$, is in resonance with the two-photon Raman transition frequency. In the case of sinusoidally modulated optical frequency comb, the Raman transitions are stimulated by the carrier and the modulation frequencies. A special attention has to be paid to adjusting the modulation amplitude (the index of the Bessel function, describing the optical frequency comb), based on the comparison of the power spectrum with the energy levels in a molecular system to be cooled. Compared to [7, 8], implementation of a single optical frequency comb rather than two combs is more robust for experimental realization. A single femtosecond optical frequency comb, with no modulation or sinusoidally phase modulated, induces coherent adiabatic accumulation of population in the ground state. Similar to the mechanism behind the two frequency combs approach, a small fraction of population is transferred by each pulse in the pulse train due to its very small pulse area. When sinusoidal phase modulation
is applied across individual pulse, it results in negligible population of the intermediate, electronically excited state which is a valuable result in light of preserving population and minimizing the spontaneous emission losses.

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