Impact of decoherence on internal state cooling using optical frequency combs

S. A. Malinovskaya* and S. L. Horton

Department of Physics and Engineering Physics, Stevens Institute of Technology, Hoboken, New Jersey 07030, USA

*Corresponding author: smalinov@stevens.edu

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We discuss femtosecond Raman-type techniques to control molecular vibrations, which can be implemented for internal-state cooling from Feshbach states with the use of optical frequency combs (OFCs) with and without modulation. The technique makes use of multiple two-photon resonances induced by optical frequencies present in the comb. It provides us with a useful tool to study the details of molecular dynamics at ultracold temperatures. In our theoretical model we take into account decoherence in the form of spontaneous emission and collisional dephasing in order to ascertain an accurate model of the population transfer in the three-level system. We analyze the effects of odd and even chirps of the OFC in the form of sine and cosine functions on the population transfer. We compare the effects of these chirps to the results attained with the standard OFC to see if they increase the population transfer to the final deeply bound state in the presence of decoherence. We also analyze the inherent phase relation that takes place owing to collisional dephasing between molecules in each of the states. This ability to control the rovibrational states of a molecule with an OFC enables us to create deeply bound ultra-cold polar molecules from the Feshbach state. © 2013 Optical Society of America

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Decoherence at ultracold temperatures is a subject of particular interest and importance in light of the development of methods to manipulate ultracold gases, create and control ultracold molecules [1-3], and study ultracold collisions and chemical reactions [4]. Decoherence is inherently present in ultracold dynamics, and we study it semiclassically within the process of creation of diatomic KRb molecules from Feshbach weakly bound states. Experimentally, ultracold polar KRb molecules were obtained using stimulated Raman adiabatic passage (STIRAP) [5]. As a viable substitute to the STIRAP process, we created a method that makes use of optical frequency combs (OFCs) to perform two-photon resonances and transfer population in a stepwise manner from the Feshbach state to the ground, ultracold state. The implementation of an OFC may be beneficial owing to its intrinsic ability to address the manifold of excited states simultaneously. We make use of a standard OFC and one with sinusoidal phase modulation to induce two-photon Raman transitions. Our theory demonstrates that the impact of decoherence may be minimized by implementing the sinusoidally modulated OFC.

The standard OFC is generated by the phase-locked pulse train of the form

\[ E(t) = \sum_{k=0}^{N-1} E_0 \exp\left[-(t - kT)^2/(2\tau^2)\right]\cos(\omega_{0L}(t - kT + \phi)). \]  

Equation (1) gives a periodic envelope of the field which oscillates with an optical carrier frequency \( \omega_{0L} \), and \( k \) is an integer number. The pulse-train period \( T \) is much greater than \( \tau \), where \( \tau \) is an individual pulse duration. A strictly periodic envelope function can be expressed as a power spectrum by the Fourier series, which is a comb of laser frequencies precisely spaced by the pulse-repetition rate \( \omega_r \) equal to \( 1/T \). Thus, the period of the pulse train determines the spacing between modes in the frequency comb and may be within a 10 ns time scale, for instance as in [6], giving radio frequency \( \omega_r \) of 100 MHz. The Ti:sapphire laser gives rise to about \( 10^6 \) modes in the comb. We investigate a phase modulation across an individual pulse introduced in the form of the sine or cosine function. The temporal field then may be written as

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

(2)

Here, \( \Phi_0 \) is the modulation amplitude and \( \Omega \) is the modulation frequency. The time-dependent phase of the field in the form of a sine/cosine function brings additional peaks to the frequency comb. Laser frequency \( \omega_r \) determines the center of the frequency comb, while \( \Omega \) determines the uniform spacing of the individual sets of modes. However, the envelope of the power spectrum is shaped by the Bessel function and leads to different intensities of modes depending on the value of the modulation amplitude. Within each set of modes, the uniform spacing between the modes is determined by \( T^{-1} \). The Fourier transform of Eq. (2) reads

\[ E(\omega) = ((E_0\pi)/2) \sum_n J_n(\Phi_0) \exp\left((-1/2)(\omega_{0L} + n\Omega - \omega)^2\tau^2\right) \]
\[ \cdot \sum_k \exp(ik\omega T). \]  

(3)

Here, \( J_n(\Phi_0) \) is the Bessel function of the order \( n \) and \( \Phi_0 \) is the modulation index. When multiplied by
\[ \exp(-1/2)(\alpha_L + n\Omega - \omega)^2z^2 \], it determines the shape of the power spectrum of the OFC. Depending on the value of \( \Phi_0 \), the power spectrum has different numbers of maxima; for details see [7]. The pulse trains and the power spectra obtained numerically using the fast Fourier transform are shown in Fig. 1. Figure 1(a) shows the temporal field with no modulation (green), sine modulation (black), and cosine modulation (red). Figure 1(b) shows the power spectrum of the OFC with the sine modulation, Fig. 1(c) the same with the cosine modulation, and Fig. 1(d) without modulation. Parameters used in the calculations are the pulse duration \( \tau = 0.25 \) \( [\omega_{31}] \), modulation frequency \( \Omega = 4.9 \) \( [\omega_{31}] \), modulation amplitude of 4, and peak Rabi frequency of 1 \( [\omega_{31}] \). The pulse-train period is taken out of scale for qualitative purposes. The value of \( \omega_{31} \) may be, for example, 70 THz, as in [8] and as used in our analysis. The power spectra presented in Figs. 1(b)–1(d) have visually unresolved fine-mode structure manifesting itself as a solid color under the envelope. Increasing \( \Omega \) broadens the frequency-comb region, while \( \Phi_0 \) determines the amplitude of the comb’s peaks. Frequency-comb peaks are narrower for longer propagation times of the pulse train \( t_{\text{total}} \); that is, \( 1/t_{\text{total}} \) determines the bandwidth of an individual mode.

A sinusoidal modulation at the megahertz frequency of the beam of a cw ring dye laser was implemented in [9] to study absorption resonances in \( I_2 \) with high precision. Here we assume modulation in the terahertz region, which may be accomplished by the approach based on the use of the enhancement cavity as discussed in [10].

We consider a three-level \( \Lambda \) system interacting with the frequency comb to describe semiclassically the dynamics of the Feshbach molecules interacting with radiation, resulting in their stepwise transformation into ultracold molecules. The \( \Lambda \) system is formed by the energy levels that include the Feshbach state, the excited transitional state, and the final ultracold state, as shown in Fig. 2.

For the description of the system dynamics we refer to the Liouville–von Neumann equation, which provides a set of coupled differential equations for density-matrix elements:

\[ \dot{\rho}_{11} = 2i[H_{12}\rho_{21} + H_{13}\rho_{31}], \]
\[ \dot{\rho}_{22} = 2i[H_{21}\rho_{12} + H_{23}\rho_{32}], \]
\[ \dot{\rho}_{33} = 2i[H_{31}\rho_{13} + H_{32}\rho_{23}], \]
\[ \dot{\rho}_{12} = -iH_{12}(\rho_{22} - \rho_{11}) - iH_{13}\rho_{32} + iH_{23}\rho_{13}, \]
\[ \dot{\rho}_{13} = -iH_{13}(\rho_{32} - \rho_{11}) - iH_{12}\rho_{23} + iH_{23}\rho_{12}, \]
\[ \dot{\rho}_{23} = -iH_{23}(\rho_{32} - \rho_{22}) - iH_{21}\rho_{13} + iH_{31}\rho_{13}. \] (4)

The interaction Hamiltonian used in these equations is written beyond the rotating-wave approximation. Nonzero Hamiltonian matrix elements in the interaction representation read \( H_{ij} = \Omega_R(t-T)[\exp(-i((\alpha_L + \omega_{31}))(t-T) + M(t-T))] + \exp(i((\alpha_L - \omega_{31}))(t-T) + M(t-T))] \); \( i, j \) 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 phase modulation in a single pulse, which is zero for the standard OFC; and \( \Omega_R(t-T) = \Omega_R \exp(-((t-T)^2/2\tau^2)) \) is the Rabi frequency, with the peak value \( \Omega_R \). The decoherence effects caused by spontaneous emission and collisions are taken into account through the reduced density-matrix elements

\[ \dot{\rho}_{11}^{\text{sp}} = \gamma_1(\rho_{11} - \rho_{22}) + \gamma_2(\rho_{33} - \rho_{11}) + \gamma_3(\rho_{12} + \rho_{21}) + \gamma_4(\rho_{13} + \rho_{31}), \]
\[ \dot{\rho}_{22}^{\text{sp}} = \gamma_1(\rho_{22} - \rho_{11}) + \gamma_2(\rho_{33} - \rho_{22}) + \gamma_3(\rho_{12} + \rho_{21}) + \gamma_4(\rho_{13} + \rho_{31}), \]
\[ \dot{\rho}_{33}^{\text{sp}} = \gamma_1(\rho_{33} - \rho_{11}) + \gamma_2(\rho_{33} - \rho_{22}) + \gamma_3(\rho_{12} + \rho_{21}) + \gamma_4(\rho_{13} + \rho_{31}), \]
\[ \dot{\rho}_{12}^{\text{sp}} = -iH_{12}(\rho_{22} - \rho_{11}) + \gamma_3(\rho_{12} + \rho_{21}), \]
\[ \dot{\rho}_{13}^{\text{sp}} = -iH_{13}(\rho_{32} - \rho_{11}) + \gamma_4(\rho_{13} + \rho_{31}), \]
\[ \dot{\rho}_{23}^{\text{sp}} = -iH_{23}(\rho_{32} - \rho_{22}) + \gamma_3(\rho_{12} + \rho_{21}) + \gamma_4(\rho_{13} + \rho_{31}). \] (5)

where \( \gamma_1, \gamma_3 \) are spontaneous emission rates from the excited state [2] to the initial state [1] and the final state [3], respectively. The \( \gamma_2 \) may also be attributed to inelastic collision rates. The collisional dephasing is taken into account by the rates \( \Gamma_{21}, \Gamma_{31}, \Gamma_{23} \). The reduced density-matrix elements were added to Eqs. (4), which were solved numerically.

To demonstrate the powerful tool of the modulated OFC to control rovibrational degrees of freedom at ultracold temperatures in the presence of decoherence, we analyze the impact of different channels individually within our model. At first, we discuss the case where decoherence is not taken into account and emphasize similarities and differences in the dynamics induced by the standard and the modulated OFCs. Then, we turn on the decoherence in our model experiment and illuminate the advantages the sinusoidally modulated OFC brings to ultrafast dynamics by minimizing the effects caused by fast spontaneous emission and collisions.

The standard OFC and the sinusoidally modulated OFC application to the three-level \( \Lambda \) system leads to a full population transfer to the ultracold state from the Feshbach state [2]. The difference is in the degree of involvement of the intermediate state into dynamics. The standard OFC populates substantially the excited transitional state; its population may rise up to 50% depending on whether the carrier frequency of the pulse train is in resonance with the transition from the excited to the ultracold state or is detuned. The results for the case of the one-photon resonance condition are presented in Fig. 3 using an OFC having \( f = 5 \) GHz, the carrier frequency \( \omega_L = 434.8 \) THz, the pulse duration \( T_0 = 3 \) fs, and the peak
Rabi frequency $\Omega_R = 1.26 \ \text{THz}$; the system parameters are $\omega_{21} = 309.3 \ \text{THz}$, $\omega_{32} = 434.8 \ \text{THz}$, and $\omega_{31} = 125.5 \ \text{THz}$ [5].

In contrast to the effect induced by the standard OFC, the sinusoidally modulated OFC, while performing a stepwise population transfer to the final ultracold state, only negligibly populates the excited state. This dynamic is observed under the condition of the one-photon resonances of the carrier and the modulation frequencies with the Feshbach-to-excited-state and the excited-to-ultracold-state transitional frequencies, respectively. The dynamics are presented in Fig. 4.

Before discussing the impact of decoherence, let us point our attention to the dependence of the dynamics on the parity of the applied chirp of the field. The implementation of the odd and even chirps may result in a substantially different response of the system and the quantum yield. We investigated the effect of the sine and cosine modulation within the three-level model and revealed a significant dependence of the population dynamics on the parity of the phase modulation. While the sinusoidal modulation across an individual pulse in a phase-locked pulse train leads to a stepwise adiabatic population transfer as described above, the cosine modulation induces population jumps from the initial to the final state within a much shorter timescale and is accompanied by a significant population of the excited state during the transitional time, as shown in Fig. 5. The system parameters are $\omega_{23} = 410.7 \ \text{THz}$ and $\omega_{31} = 340.7 \ \text{THz}$ [8]. The carrier frequency is $\omega_1 = \omega_{23}$, the modulation frequency is $\Omega = \omega_{32}$, the modulation amplitude is $\Phi_0 = 4$, and the peak Rabi frequency is $\Omega_R = 70 \ \text{THz}$. The results of the implementation of the sinusoidally modulated comb are reproduced from Fig. 4(a) for a comparison, showing a stepwise decrease of population in state [1], an increase of population in state [3], and a negligible population in the transitional state [2]. The cosine-modulated pulse train induces transitions in the three-level system shown by blue color for state [1], by red for state [3], and by orange for state [2]. The induced Rabi oscillations take place an order of magnitude faster, resulting in population transfer from the Feshbach state to the ultracold state by six pulses in the pulse train and occupying the excited state by up to 50%. The phenomenon strongly depends on the magnitude of the Rabi frequency. According to [11], the different results of the sine and cosine chirp are expected. The authors analyzed the Taylor series expansion of the field phase having an arbitrary time dependence and showed that even odd chirps lead to qualitatively different responses in a many-level system.

The mechanism of population transfer induced by the OFC, both modulated or unmodulated, is the Rabi oscillations performed in a stepwise manner with a fraction of the population transferred to the final state by each pulse in the pulse train, depending on the field intensity. For the dynamics induced by the standard OFC, shown in Fig. 3, the pulse area of a single pulse in the pulse train is 0.003$\pi$ and the total population transfer occurs within 242 pulses, giving a total pulse area equal to 0.7$\pi$, indicating a weak field regime achieved by field intensities of about $10^{10} \ \text{W/cm}^2$. In a strong field regime, when the peak Rabi frequency is about $\omega_{31}$, the qualitative picture of Rabi oscillations is preserved; however, the total population transfer is accomplished by just a few pulses in the train (not shown here). When the sinusoidally modulated train is applied, as in Fig. 4, with the effective Rabi frequency equal to the two-photon transitional frequency $\omega_{32}$ that provides a single-pulse area of 0.2$\pi$, the total population transfer is accomplished by 112 pulses. These give the total pulse area of 22.4$\pi$, implying a strong field regime with the field intensity about $10^{12} \ \text{W/cm}^2$. The excited state plays the role of the dark state in the dynamics and gets negligibly populated during transitional times. However, when the Rabi frequency is chosen to be one order of magnitude less, Rabi oscillations are preserved but the excited state is heavily occupied. There are two ways for the formation of two-photon resonances; each is performed in a stepwise manner leading to Rabi oscillations. When the OFC is unmodulated, the multiples of the radio-frequency modes form pairs of optical frequencies.
whose difference is in the two-photon resonance with the three-level system. When the sine modulation is applied, the carrier and modulation frequency difference is chosen to be in resonance with the two-photon transition to efficiently induce the Raman transition. A sequence of pulses carrying resonant frequencies coherently accumulate population in the final state and then move it back, executing the Rabi oscillations. In the case of sinusoidal modulation, the departure from the one-photon resonance condition decreases the impact of the modulation frequency and the carrier frequency difference into the dynamics of population transfer, resulting in the dominant contribution from the channel involving the radio frequencies. This leads to a significant involvement of the transitional state into the dynamics; see Fig. 6. Here the time evolution of population is presented for parameters \( \Omega_R = 1 \), carrier frequency \( \omega_c = 5.4 \), modulation frequency \( \Omega = 4.4 \), and modulation amplitude \( \Phi_0 = 4 \); the \( \Lambda \)-system transition frequencies are \( \omega_{21} = 4.9 \) and \( \omega_{32} = 5.9 \), giving \( \delta = \omega_{31}/2 \). All frequencies are given in the units of \( \omega^{-1} \), where \( \omega = \omega_{31} = 70 \) THz. Figure 6 shows the coherent accumulation of the population in the final state, taking place 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 are faster with fewer pulses in the pulse train involved in full population transfer. Other key control parameters that govern the dynamics of adiabatic population transfer are the pulse-train period, the pulse duration, and the modulation amplitude in the case of sine or cosine phase modulation. Their variation induces changes in the dynamics of population transfer and the quantum yield but preserves the adiabatic nature of the transfer in the form of Rabi oscillations.

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**Fig. 4.** (Color online) Population transfer in the three-level \( \Lambda \) system achieved via the resonant Raman transitions using a sinusoidally modulated OFC described by Eq. (2) (\( \Phi = 0 \)). The values of the system parameters are \( \omega_{32} = 410.7 \) THz and \( \omega_{21} = 340.7 \) THz [8]. The carrier frequency is \( \omega_c = \omega_{32} \), the modulation frequency is \( \Omega = \omega_{21} \), the modulation amplitude is \( \Phi_0 = 4 \), and the peak Rabi frequency is \( \Omega_R = 70 \) THz. The pulse duration is \( \tau = 3 \) fs and the pulse-train period is (a) 6400 ps (20 ps), giving \( \omega_p = 50 \) GHz, and (b) 640 ps (2 ps), giving \( \omega_p = 500 \) GHz. 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] is in black, and the excited-state manifold [2] is in red. Time is given in the units of \( \omega^{-1} \), where \( \omega = \omega_{31} = 70 \) THz.

**Fig. 5.** (Color online) Population transfer in the three-level \( \Lambda \) system achieved via the resonant Raman transitions using a phase-modulated OFC described by Eq. (2) with cosine modulation; blue: state [1], orange: state [2], indigo: state [3]. For comparison, the results from the previous figure for the sinusoidally modulated comb are presented. The values of the system parameters are \( \omega_{32} = 410.7 \) THz and \( \omega_{21} = 340.7 \) THz [8]. The carrier frequency is \( \omega_c = \omega_{32} \), the modulation frequency is \( \Omega = \omega_{21} \), the modulation amplitude is \( \Phi_0 = 4 \), and the peak Rabi frequency is \( \Omega_R = 70 \) THz. The pulse duration is \( \tau = 3 \) fs, and the pulse-train period is 2 ps. The cosine modulation induces much faster Rabi oscillations and populates the excited state up to 50%.

**Fig. 6.** (Color online) Population transfer in the three-level \( \Lambda \) system achieved using an OFC as in Eq. (2) with the field parameters detuned off one-photon resonance with the frequencies of the \( \Lambda \) system. The detuning \( \delta \) is equal to \( \omega_{31}/2 \). The carrier frequency is \( \omega_c = 5.4 \), the modulation frequency is \( \Omega = 4.4 \), the Rabi frequency is \( \Omega_R = 1 \), and the modulation amplitude is \( \Phi_0 = 4 \). The pulse duration is 3 fs, and the pulse-train period is 20 ps. The \( \Lambda \)-system transition frequencies are \( \omega_{21} = 4.9 \) and \( \omega_{32} = 5.9 \) (in units of frequency \( \omega_{31} = 70 \) THz). 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. Time is given in the units of \( \omega^{-1} \), where \( \omega = \omega_{31} = 70 \) THz.
oscillations. The pulse-train period is one of the parameters of major importance as it determines the spacing of two adjacent comb curves. The decrease of the pulse-train period results in the decrease of efficiency of population transfer owing to larger one-photon detunings of the radio-frequency multiples with the transitional frequencies in the $\Lambda$ system; see Fig. 4(b). Pulse duration determines the overall width of the comb on the one hand; on the other hand, it may be used as a control parameter through which the pulse area is carefully manipulated. The variation of the pulse duration does show the pulse-area type dependence of the population dynamics in the three-level system. More details on the robustness of adiabatic population transfer using OFCs may be found in [7]. It should be noted here that the implemented Rabi frequencies in the terahertz region correlate with the electric fields of $10^6$ V/cm. If the excited state is significantly populated, they may expedite the heating of the ultracold cloud. However, because the sinusoidal modulation induces the dynamics that are essentially owing to the two-photon transitions with a negligible population of the excited state, it allows using this field strength. When the unmodulated comb is implemented, causing population of the excited states, much lower values of the Rabi frequencies, within the gigahertz region, may be successfully applied.

In the presence of fast decoherence, the different degrees of the involvement of the excited state into population dynamics induced by the standard and sinusoidally modulated OFCs lead to remarkably different responses of the three-level system to the applied field. We decompose the contributions from spontaneous decay and collisional dephasing in our analysis. The time-dependent dynamics induced by the standard OFC in the presence of only collisional dephasing are presented by solid lines in Fig. 7. Here, the quantum yield to the ultracold state is only 38% in the presence of fast collisions. When we add the spontaneous decay from the excited state, we observe that it reduces the collisional impact and yields a higher steady-state population of the final ultracold state equal to 45%; see Fig. 7 (dashed curves). The data are presented for the following decoherence parameters: solid curves for $\gamma_{21} = 0$, $\Gamma_{12} = \Gamma_{23} = 0.001$; and dashed curves for $\gamma_{21} = \gamma_{32} = 0.001$ and $\Gamma_{12} = \Gamma_{23} = 0.001$, in the units of $\omega_{31} = 125$ THz [5]. The Rabi frequency $\Omega_R$ is equal to 0.1. The interplay of the spontaneous decay rate and the collision dephasing rate may result in an increase of the quantum yield of ultracold molecules.

Next, we demonstrate the effect caused by sinusoidal modulation across an individual pulse in the phase-locked pulse train. It results in a remarkably high quantum yield to the final ultracold state under the same decoherence conditions. A comparison is shown in Fig. 8(a), where the dashed curves correspond to the standard OFC implementation as in the previous figure and the solid curves show the system response to the sine-modulated OFC. The sinusoidally modulated comb negligibly involves the excited state into transitional dynamics (Fig. 4), minimizing the effects caused by decoherence. Two horizontal solid lines coinciding at the population equal to 0.5 show the steady-state solution when the cosine modulation across an individual pulse in the pulse train was implemented, justifying the importance of the parity of the chirp in achieving the desired quantum yield. In Fig. 8(b), the same results are presented for a different set of parameters chosen from [8], which are the system parameters $\omega_{23} = 410.7$ THz and $\omega_{21} = 340.7$ THz; the carrier frequency is $\omega_c = 434.8$ THz, and $\omega_{21} = 125.5$ THz. Solid curves represent the case of pure collisional dephasing, $\Gamma_{12} = \Gamma_{23} = 0.001$; dashed curves correspond to the same values of $\Gamma$, and $\gamma_{21} = \gamma_{32} = 0.001$; and $\Gamma_{12}$ and $\gamma_{12}$ are given in units of $\omega_{31}$. Note that the dashed red curve is visible in the vicinity of the coordinate origin, showing that the population of the excited state decays almost instantaneously within the presented time scale.

In this work, we have uncovered a rigorous expression for the relation between collisional dephasing rates in the three-level $\Lambda$ system. Previously it was observed that for certain values of collisional dephasing rates, the received populations at times incorrectly exceeded the initial total population. However, if looking from the physical point of view at several special cases, one would see that the values of collisional dephasing rates are mutually dependent. The instantaneous phases of three states are $\phi_1$, $\phi_2$, and $\phi_3$. When, for example,
molecules in the ultracold configuration, state |3⟩, collide elastically with the atomic Rb and K at the same rate as the Feshbach molecules, state |1⟩, while state |2⟩ is still, then one would see that the state |3⟩ and |1⟩ amplitudes loose their phase synchronously, which means Δφ_{31} = 0 and thus Γ_{31} must be zero, while Δφ_{32} and Δφ_{33} are equal to each other, thus requiring that Γ_{31} = Γ_{32}. Two other nontrivial cases are when states |3⟩ and |1⟩ dephase synchronously but out of phase with state |2⟩; then Γ_{31} must be zero because Δφ_{31} = 0, while |Δφ_{31}| and |Δφ_{32}| are equal because state |2⟩ looses phase correlation with two other states at the same rate, and thus Γ_{21} = Γ_{32}. Finally, if all three states dephase differently, then the instantaneous phase difference between states |1⟩ and |3⟩ is equal to Δφ_{31} and that between states |1⟩ and |2⟩ is equal to Δφ_{21}, which apparently leads the instantaneous phase difference between states |2⟩ and |3⟩ to be the sum of phase differences |Δφ_{21}| and |Δφ_{31}|, giving Γ_{32} = Γ_{31} + Γ_{21}. The seemingly unique case when states |3⟩ and |1⟩ dephase differently and state |2⟩ stays still casts into the previous case. These observations fit to the phasor diagram presented in Fig. 10, where the instantaneous values of the phase differences among three states are shown by the phasors.

With the help of the phase diagram, we obtained a general relation between dephasing rates in the three-level Λ system, which reads Γ_{33} = Γ_{12} + Γ_{31}. With the values of collisional dephasing rates limited by this relation, we observed that our total population always remained constant. Our results complement the work in [13,14].

In summary, we have analyzed the interaction of an OFC with a three-level Λ system that leads to cooling of internal degrees of freedom in the KRb molecule from the Feshbach state into an ultracold, deeply bound state. Using the reduced density matrix to introduce decoherence in the forms of spontaneous emission and collisional dephasing, we studied the effectiveness of the population transfer into the ultracold state performed by the standard OFC and a sine- or cosine-modulated OFC and unmodulated OFC lead to stationary solutions with the population distribution between the Feshbach and ultracold state close to 50%.

![Fig. 8. (Color online) Population dynamics induced by the sinuso- dially modulated OFC (solid curves) versus the standard one (dashed curves) in the presence of decoherence. The system parameters are (a) as in [5], ω_{31} = 300.3 THz, ω_{32} = 434.8 THz, and Ω = 12.5 THz; (b) as in [8], ω_{31} = 340.7 THz, ω_{32} = 410.7 THz, and Ω = 70 THz. Decoherence parameters are γ_{21} = γ_{32} = 0.001 and Γ_{31} = Γ_{32} = 0.001, in ω_{02} units. Sinusoidal modulation provides almost full population transfer to the ultracold state, while the cosine modulation leads to a stationary solution with equal population distribution between the Feshbach and ultracold state, thus creating the maximum coherence.](image1)

![Fig. 9. (Color online) Population dynamics induced by (a) the sine-modulated OFC, (b) the cosine-modulated OFC, and (c) the standard OFC, in the presence of decoherence. The system parameters are (a) as in [5], ω_{31} = 300.3 THz, ω_{32} = 434.8 THz, and Ω = 12.5 THz [5]. Decoherence parameters are γ_{21} = γ_{32} = 10^{−6} and Γ_{31} = Γ_{32} = 10^{−10} in ω_{02} units. The carrier frequency of the field is ω_{01} = ω_{02}, the modulation frequency is Ω = ω_{21}, the modulation amplitude is Φ_{m} = 4 for the sine-/cosine-modulated combs, and the peak Rabi frequency in Ω_{01} = 1 [ω_{02}], a weak field regime. The sine-modulated OFC provides almost full population transfer to the ultracold state; the cosine-modulated OFC and unmodulated OFC lead to stationary solutions with the population distribution between the Feshbach and ultracold state close to 50%.](image2)

![Fig. 10. (Color online) Phasor diagram analyzing collisional dephasing of the system.](image3)
cosine-modulated OFC and uncovered a remarkable difference in the system’s response. We have shown that in the presence of fast decoherence an efficient cooling of internal degrees of freedom from the Feshbach state may be accomplished by a sinusoidally modulated OFC, while a standard OFC implementation leads to a substantial loss of efficiency in the presence of fast dephasing. The sine-modulated OFC induces the response in the three-level system that resembles the dark state formed in STIRAP. It negligibly populates the excited-state manifold during a stepwise population transfer from the initial to the final ultracold state and, thus, minimizes the effect of decay and optimally resolves the collisional impact.

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