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Two-photon adiabatic passage in ultracold Rb interacting with a single nanosecond, chirped pulse

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Abstract
A semiclassical, four-level model of a nanosecond, chirped pulse interacting with all optically accessible hyperfine states in an ultracold alkali atom, e.g., the $^8\text{Rb}$, is analyzed aiming at population inversion within $5\text{S}_{1/2}$ electronic state. The nature of two-photon adiabatic passage performed by a single pulse having the bandwidth prior to chirping significantly narrower than the hyperfine splitting of $5\text{S}_{1/2}$ state is investigated in the framework of the dressed state picture. It is shown that two dressed states are involved in the adiabatic dynamics of population inversion. The excited state manifold appeared to play an important mediating role in the mechanism of population transfer.

Keywords: adiabatic passage, Raman transitions, dressed states, avoided crossing, ultracold alkali atoms

(_some figures may appear in colour only in the online journal)
modulation method developed for nanosecond pulses [15] allows to change gradually the instantaneous frequency while preserving the value of the peak intensity, the spectral bandwidth of the pulse is broadened by the chirp, (for the unchirped pulse, the bandwidth would be transform limited).

We consider the energy levels relevant for the $^{85}$Rb and analyze the four-level system which takes into account all optically allowed transitions between hyperfine states belonging to $5^2S_{1/2}$ and $5^3P_{1/2}$ or $5^3P_{3/2}$ states, figure 1. The population inversion is achieved through the Raman transitions that involve hyperfine structure of these states and with the aid of linear chirping of the pulse. The adiabatic solution is found through a systematic numerical analysis of the response of the four-level system to a broad variation of the field parameters. We have made a detailed analysis of the dressed state picture to gain insight into adiabatic mechanism of the two-photon Raman transition by means of a single nanosecond pulse, which reveals the involvement of two dressed states into adiabatic passage producing population inversion. These two dressed states form a subset owing to two energetically close hyperfine states of the transitional $5^3P_{1/2}$ or $5^3P_{3/2}$ state. Because only one chirped pulse is implemented having a bandwidth prior to chirping narrower than the hyperfine splitting of $5^3P_{1/2}$ state, the excited state manifold plays the key role in the passage as a mediator.

The time-dependent Hamiltonian that describes the four-level system interaction with a single nanosecond, chirped pulse reads

$$\hat{H}_{\text{int}} = \hbar \begin{bmatrix} \Delta + \omega_{43} & 0 & -\Omega_R(t) & -\Omega_R(t) \\ 0 & \Delta + \omega_{43} + \omega_{21} & -\Omega_R(t)/2 & -\Omega_R(t)/2 \\ -\Omega_R(t)/2 & -\Omega_R(t)/2 & 0 & 0 \\ -\Omega_R(t)/2 & -\Omega_R(t)/2 & 0 & \omega_{43} \end{bmatrix}$$

Figure 1. Four optically attainable hyperfine states of $5S$ and $5P$ shells with the energy differences that correspond to the D1 line. Initially, the population is in the ground state $|1\rangle$. Note that the hyperfine splitting of the $5S_{1/2}$ orbital is approximately an order of magnitude greater than the splitting of $5P_{1/2}$ orbital, [19].

Here $\Omega_R(t) \equiv -\mu E_0(t)/\hbar$ is the Rabi frequency with the peak value $\Omega_R$, $\alpha$ is the linear chirp parameter, $\alpha/2\pi$ has units Hz s$^{-1}$, and $\Delta$ is the one-photon detuning. Note, that a complete treatment would require inclusion of all the magnetic sublevels of each hyperfine level, but in an experiment, the system could be restricted to four states by preparing a single state via optical pumping and using selection rules based on polarization of the chirped light to restrict the number of states involved. A numerical example of the application of the left or right circularly polarized, chirped pulse to magnetic sublevels in the $^{85}$Rb and taking into account the relevant transition dipole moments is provided in the Appendix.

Solving numerically the time-dependent Schrödinger equation with the Hamiltonian in equation (1) for various values of the field parameters provides an accurate picture of light-matter interactions and allows for finding the exact values of the field parameters required to obtain a predetermined non-equilibrium superposition state, population invention or population return. It also reveals the adiabatic region of population transfer to the target state $|2\rangle$, which is the upper hyperfine state $F = 3$ of the $5^2S_{1/2}$. Populations of the four states at the end of the pulse as a function of the pulse chirp rate and the full width at the half maximum (FWHM) are presented in figure 2, [16]. FWHM of the Gaussian pulse relates to the pulse duration $\tau_0$ as FWHM $= \tau_0/2\sqrt{\ln 2}$. The adiabatic region of light–matter interaction leading to population inversion is observed for parameters that satisfy the adiabaticity conditions $|\alpha/(2\pi)|\tau_0 > \omega_{21}$ and $|\alpha/(2\pi)| < \Omega_R$. The physical values applicable to $^{85}$Rb are, e.g., the peak Rabi frequency $\Omega_R = \omega_{21}$ (3.035 GHz), the chirp rate $\alpha/2\pi = -0.3[\omega_{43}^2] (-3$ GHz ns$^{-1})$ or faster and the pulse duration $\tau_0 > 5.5[\omega_{31}^2]$ ($>1.8$ ns). The negative value of the chirp rate is well understood since we start from the one-photon blue detuning with the largest transition frequency $\omega_{43}$ and gradually decrease the instantaneous frequency to pass through each one-photon resonance, first with $\omega_{43}$, then with $\omega_{31}$, $\omega_{32}$ and $\omega_{33}$. The exemplified field parameters may be obtained in modern experimental setups such as described in, e.g., [15]. Since the spectral bandwidth of the unchirped nanosecond pulse (0.5 GHz for 1.8 ns pulse) is much narrower than the energy separation between the hyperfine states (3.035 GHz) of the $5^2S_{1/2}$, a question of fundamental interest arises as to what is the mechanism of the adiabatic population transfer performed with the two photons that are never present in the system with the frequency ‘right’ to satisfy the two-photon resonant condition? To answer this question, we performed the dressed state analysis to gain insight into the adiabatic and nonadiabatic nature of quantum control of population dynamics in the four-level system using a single narrowband but chirped laser pulse.

We first outline a basic concept of the dressed state analysis, [17, 18], and its extension to the case when adiabatic passage may occur within a subset of dressed states coupled to each other. A wave function of a quantum system $|\Psi(t)\rangle$ may be written as a linear superposition of the bare states in the field interaction representation $|\ell\rangle$ with the respective time-
dependent probability amplitudes $C_i$

$$|\Psi(t)\rangle = \sum_i C_i |i\rangle.$$ (2)

The time-dependent Schrödinger equation then reads as $i\hbar \dot{C} = \hat{H}_{\mathrm{int}} C$. We apply a unitary time-dependent transformation $T$ to the $\hat{H}_{\mathrm{int}}$ leading to diagonalization of the Hamiltonian. Here, $T$ is an eigenvector matrix of $\hat{H}_{\mathrm{int}}$. The obtained Hamiltonian is the so called dressed state Hamiltonian $\hat{H}_d = T^\dagger \hat{H}_{\mathrm{int}} T$ written in the basis of the dressed states $|i\rangle$ such that $|\Psi(t)\rangle = \sum_i C_i |i\rangle$ and $C_d = TC$. Then, putting the reverse expression $C = T^\dagger C_d$ into the Schrödinger equation and assuming that all quantities are time dependent we arrive at

$$i\hbar \dot{C}_d = \hat{H}_d C_d - i/\hbar T^\dagger \hat{T}_d C_d.$$ (3)

Since the Hamiltonian $\hat{H}_d$ is diagonal, the dressed states would evolve without mixing with each other if we disregard the second term on the right side. This is the essence of the adiabatic approximation, when the system, once placed in a selected dressed state by the initial conditions, continues evolution within this dressed state only. The second term is responsible for the nonadiabatic coupling between the dressed states, it contains matrix operator $T\hat{T}_d^\dagger$ which is non-diagonal and determines the degree of non-adiabatic mixture between the dressed states. If the matrix elements of $T\hat{T}_d^\dagger$ are much less than the energy splittings between the respective dressed states, the dynamics may be considered as adiabatic. Analyses of the time-dependence of the dressed state energies and the wave functions as well as a comparison of non-adiabatic and adiabatic terms help to estimate the degree of adiabaticity and a possibility for quantum control. From another hand, if we aim to find the field parameters that provide the adiabatic solution, it is useful to move to the dressed state basis and within the adiabatic approximation find the field conditions and parameters for dynamics in a single dressed state. When

Figure 2. The end-of-pulse population distribution in the four-level system, achieved via two-photon transitions using a single, linearly chirped laser pulse. The values of the system parameters are $\omega_1 = 3.035$ GHz, $\omega_3 = 0.362$ GHz, characteristic for $^{85}\text{Rb}$ [19], the peak Rabi frequency is $\Omega_{\text{R}} = 3.035$ GHz, and one-photon detuning $\Delta$ is zero.
implemented in the exact Schrödinger equation, these parameters may yield quasi-adiabatic behavior within the exact Schrödinger picture.

In a multi-level case, the non-adiabatic coupling may be small for some dressed states, but significant for the others. Then coupled dressed states may create a subsystem within which the dynamics occurs adiabatically. For the four-level system described by the Hamiltonian in equation (1), a superposition of two energetic close excited states demonstrates adiabatic passage to the final state |2⟩. The parameters of the Hamiltonian in equation (1) are chosen from the numerical solution of the Schrödinger equation, these parameters may yield quasi-adiabatic behavior within the exact Schrödinger picture.

We demonstrate the concept by analyzing the time dependence of the dressed state energies in the four-level system and the squares of the respective eigenvector elements. The respective dressed state energies for the same parameters are Δ = 0, ΩI = 3.035 GHz, FWHM = 2.995 ns and α/2π = −2.947 GHz ns⁻¹. The inset of the figure contains the enlarged central region of the main figure with a fine time scale.

The exact time-dependent picture of the adiabatic passage to the final hyperfine state obtained by numerically solving the time-dependent Schrödinger equation with the parameter values Δ = 0, ΩI = 3.035 GHz, FWHM = 2.995 ns and α/2π = −2.947 GHz ns⁻¹. The inset of the figure contains the enlarged central region of the main figure with a fine time scale.

The respective dressed state energies for the same field parameters FWHM = 2.995 ns, α/2π = −2.947 GHz ns⁻¹, ΩI = 3.035 GHz and Δ = 0 are depicted in figure 4. It demonstrates that if the chirp rate is large enough, the change of the instantaneous carrier frequency is sufficient to swipe adiabatically through the two-photon resonance. The dynamics in the system begins within the dressed state |I⟩ (blue in figure 4), which coincides with initially populated bare state |1⟩, (shown in blue dashed color). Further on, the dressed state |I⟩ (blue in figure 4) approaches the dressed state |III⟩ (green in figure 4) near the peak value of the field amplitude to form an avoided crossing. In its vicinity, population moves efficiently from the |I⟩ to the |III⟩ dressed state and, thus, resides transitionally on the excited bare state manifold for a restricted period of time before moving to the final bare state |2⟩ as the dressed state |III⟩ evolves to become 100% constitutes of it. Thus, the excited states |3⟩ and |4⟩ keep population for the time needed for the instantaneous frequency to acquire the value needed to accomplish the two-photon resonance. In such a way, the adiabatic passage is performed by a subset of coupled through the avoided crossing dressed states and requires the excited state manifold...
to mediate the dynamics owing to only one chirped pulse used to perform the inversion.

For a comparison, the case when the chirp is not large enough to provide the range of frequencies needed to satisfy the two-photon transition is shown in figure 5. Here the chirp rate is \( \alpha/2\pi = -0.092 \text{ GHz ns}^{-1} \) and the pulse duration is \( \tau_0 = 1.799 \text{ ns} \) (FWHM = 2.995 ns), giving \( \alpha\tau_0 = 0.166 \text{ GHz} \ll \omega_21 = 3.035 \text{ GHz} \). The other parameters are the peak Rabi frequency \( \Omega_R = \omega_21 = 3.035 \text{ GHz} \), and one-photon detuning \( \Delta = 0 \). Here, dressed state \(|1\rangle\) initially coincides with bare state \(|1\rangle\) (blue in figure 5); this picture remains for most of the pulse duration. Then, close to the exponential end of the pulse amplitude, dressed state \(|1\rangle\) approaches the dressed state \(|\text{III}\rangle\) (green in figure 5), which is mainly a superposition of the excited states \(|3\rangle\) and \(|4\rangle\), and nonadiabatically transfers a fraction of population to them before the pulse ceases. As the result, the population remains mostly in the initial, ground state owing to the field parameters not satisfying the adiabaticity condition for population inversion determined by the condition \( \alpha/(2\pi)\tau_0 \gg \omega_21 \), even though the Landau–Zener condition \([20]\), is satisfied, \( \Omega_R^2/|\alpha| \approx 3/2 \times 10^2 \).

We performed a comparative analysis of the results with an effective three-level \( \Lambda \) system, figure 6 that works as a good approximation to the four-level system giving a qualitatively similar quantum yield when the pulse duration and the chirp rate satisfy \( \alpha/(2\pi)\tau_0 \gg \omega_{43} \) and \( \Omega_R \gg \omega_{43} \). A detailed numerical solution for the three-level \( \Lambda \) system is discussed in \([21, 22]\).

From the Hamiltonian in equation (1), we may easily get the following set of equations for the time-dependent probability amplitudes in the field interaction representation assuming the one-photon detuning \( \Delta = 0 \)

\[
\begin{align*}
i\dot{a}_1 &= \alpha(t-T)a_1 - \Omega_R(t)(a_3 + a_4)/2, \\
\dot{a}_2 &= \left(\omega_{21} + \alpha(t-T)\right)a_2 - \Omega_R(t)(a_1 + a_4)/2, \\
\dot{a}_3 &= -\Omega_R(t)(a_1 + a_2)/2 - \omega_{43}a_3, \\
\dot{a}_4 &= -\Omega_R(t)(a_1 + a_2)/2.
\end{align*}
\]

By making a substitution

\[
\begin{align*}
(a_3 + a_4)/\sqrt{2} &= a_+, \\
(a_1 - a_2)/\sqrt{2} &= a_-,
\end{align*}
\]

we arrive at the following relation for \( a_+ \) and \( a_- \).

\[
\begin{align*}
i(\dot{a}_+ &= i(\dot{a}_+ + \dot{a}_-)/\sqrt{2}, \\
&= -\Omega_R(t)(a_1 + a_2)/\sqrt{2} - \omega_{43}/\sqrt{2a_3}, \\
i(\dot{a}_-) &= i(\dot{a}_- - \dot{a}_+)/\sqrt{2} = -\omega_{43}/\sqrt{2a_3}.
\end{align*}
\]

If \( \omega_{43} \sqrt{2} \) is small enough compared to \( \Omega_R \), it may be neglected. Then, \( a_3 - a_4 = \text{const.} \) and \( a_- \) may be omitted from the dynamics calculation. Finally, the set of equations (5) is reduced to a set of three coupled differential equations

\[
\begin{align*}
i\dot{a}_1 &= \alpha(t-T)a_1 - \Omega_R(t)/\sqrt{2}a_+, \\
i\dot{a}_2 &= \left(\omega_{21} + \alpha(t-T)\right)a_2 - \Omega_R(t)/\sqrt{2}a_+, \\
i\dot{a}_+ &= -\Omega_R(t)/\sqrt{2}(a_1 + a_2).
\end{align*}
\]

The Hamiltonian for the three-level approximation in the field interaction representation reads

\[
\hat{H}_{\text{int}} = \hbar \begin{bmatrix}
\alpha(t - T) & 0 & -\Omega_R(t)/\sqrt{2} \\
0 & \omega_{21} + \alpha(t - T) & -\Omega_R(t)/\sqrt{2} \\
-\Omega_R(t)/\sqrt{2} & -\Omega_R(t)/\sqrt{2} & 0
\end{bmatrix}
\]

Analytical diagonalization of the Hamiltonian in equation (13) leading to expressions for the dressed state energies and respective eigenfunctions showed no dark state solution as it was the case, e.g., in conventional STIRAP scheme, \([23]\). The time-dependent wave function describing each dressed state contains nonzero probability amplitudes for all three bare states. Since the expressions for the dressed state
energies and the probability amplitudes look heavy, we do not present them here, but rather show their time dependence obtained numerically. A numerical analysis of the dressed states was performed for the three-level Λ system described by the Hamiltonian in equation (13) within the same range of parameters as for the four-level system. As an example, the results for $\alpha/(2\pi) = -2.947$ GHz ns$^{-1}$, FWHM = 2.995 ns and $\Delta = -2.995$ GHz ns$^{-1}$, FWHM = 3.035 GHz discussed are in more details. The time-dependence of the dressed state energies is depicted in figure 7. Here the dynamics occurs within a single dressed state (I), shown in blue color. Initially, it coincides with bare state $|1\rangle$ (dashed blue), followed by adiabatic transition within the same dressed state in the vicinity of the peak value of the pulse amplitude from the bare state $|1\rangle$ through the excited state $|3\rangle$ (dashed green) to final bare state $|2\rangle$ (dashed black). In this approximate three-level model, the dressed state (I) plays the role of the subset of two dressed states, $|I\rangle$ and $|III\rangle$, in the four-level case. In figure 8, the adiabatic dynamics of population transfer between the bare states within each dressed state is shown by the time dependence of $T^2_f$. It reveals a smooth adiabatic passage from the initial bare state $|1\rangle$ to the final bare state $|2\rangle$ in the dressed state $|I\rangle$, figure 8(a).

However, if we choose a set of parameters such that they do not satisfy the condition $\alpha/(2\pi)\tau_0 > \omega_{21}$, which is, for example, $\alpha/(2\pi) = -0.092$ GHz ns$^{-1}$, FWHM = 2.995 ns, the dynamics still occurs within a single dressed state but does not lead to the population inversion to the final bare state $|2\rangle$. In the beginning, the energy of dressed state $|I\rangle$ coincides with the bare state $|1\rangle$; however, the pulse ceases before the dynamics within dressed state $|I\rangle$ progresses to the final bare state $|2\rangle$, which makes it retaining in the intermediate state $|3\rangle$, figure 9. The chirp rate of the pulse is not fast enough to switch through the two-photon resonance. The bare state population dynamics within each of three dressed states is shown in figure 10, it supports this outcome by manifesting adiabatic passage from the initial bare state $|1\rangle$ to the intermediate bare state $|3\rangle$ within the active dressed state $|I\rangle$, figure 10(a).

In summary, we have analyzed the mechanism of population dynamics in a new method for population inversion within the hyperfine structure in alkali atoms at ultracold temperatures that relies on implementing a single nanosecond chirped pulse of intensity on the order of kW cm$^{-2}$ having the bandwidth prior to chirping significantly narrower than the two-photon transition frequency in the atomic system. The results are based on the developed semiclassical model of the pulse interaction with the four-level system representing all optically accessible hyperfine states of $5^2S_{1/2}$ and $5^2P_{1/2}$ or $5^2P_{3/2}$ states in ultracold $^{85}$Rb. The adiabatic passage leading to population inversion is achieved for parameters that satisfy the condition $|\alpha/(2\pi)\tau_0| > \omega_{21}$ and the Landau–Zener adiabaticity condition $|\alpha/(2\pi)| < \Omega^2_f$. Dressed state analysis was performed to gain understanding about the mechanisms of two-photon Raman transitions performed by a single,
Figure 10. The adiabatic dynamics of population transfer between the bare states within each dressed state which does not lead to population inversion between states $|1\rangle$ and $|2\rangle$. The field parameters are $\Delta = 0$, $\Omega_k = 3.035 \text{ GHz}$, FWHM = 2.995 ns and $\alpha/(2\pi) = -0.092 \text{ GHz ns}^{-1}$.

Figure 11. The end-of-pulse population distribution in the four-level system representing magnetic sublevels, optically attainable via two-photon transitions using a single, linearly chirped, circularly polarized laser pulse. The transitions are accomplished by $\sigma^+$ radiation for the initially populated magnetic sublevel $F = 2$, $m_F = 0$. The values of the system parameters are $\omega_{21} = 3.035 \text{ GHz}$, $\omega_{33} = 0.362 \text{ GHz}$, characteristic for $^{85}\text{Rb}$ [19], the peak Rabi frequency is $\Omega_k = 3.035 \text{ GHz}$, and one-photon detuning $\Delta$ is zero.
narrowband, chirped pulse having the transform limited bandwidth, $\Delta \omega < \omega_{21}$. It revealed an existence of a subset of dressed states coupled in the vicinity of avoided crossings that perform the adiabatic passage leading to the population inversion. When considered as an approximation of the four-level system, the three-level $\Lambda$ system demonstrates population inversion within a single dressed state. This dressed state resembles the time-dependence of two dressed states in the active subset in the four-level system. This justifies the validity of several dressed states approach to adiabatic passage.

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Appendix A

To demonstrate the validity of the control method described in this work, we present its application to specific transitions between magnetic sublevels in the $^{85}$Rb atom using circularly polarized light.

Figure 11 shows a numerical solution of the end-of-pulse population distribution between four magnetic sublevels forming the four-level system as a function of the temporal chirp rate and the FWHM of the pulse for the $s^+$ radiation interacting with the $^{85}$Rb atom prepared initially in the magnetic sublevel $m_F = 0$ by optical pumping followed by coherent population transfer. The chosen transition dipole matrix elements correspond to $D_1$ line ($5^2S_{1/2} \rightarrow 5^2P_{1/2}$). For
the initially populated magnetic sublevel \( F = 2, m_F = 0 \), the dipole matrix elements for \( \sigma^+ \) transitions to \( F' = 2 \), \( m_F' = m_F + 1 \) and \( F' = 3 \), \( m_F' = m_F + 1 \) are \(-\sqrt{2}/27\) and \(-\sqrt{10}/27\) respectively. The dipole matrix elements for \( \sigma^- \) transitions between \( F = 3, m_F = 0 \) and \( F' = 2, m_F' = m_F + 1 \) or \( F' = 3, m_F' = m_F + 1 \) are \(\sqrt{1}/27\) and \(\sqrt{5}/27\) respectively. The contour plot for the state \( |2\rangle \) shows a broad region of adiabatic population transfer, (red color in figure 11(b)).

For the linearly chirped, circularly polarized nanosecond pulse interacting with the \(^{85}\text{Rb}\) atom prepared initially in the magnetic sublevel, \( m_F = -1 \), the results are shown in figure (12). The chosen transition dipole matrix elements correspond to \( D_1 \) line (5S\(_{1/2} \) \( \rightarrow \) 5P\(_{1/2}\)). For the initially populated magnetic sublevel \( F = 2, m_F = -1 \), the dipole matrix elements for \( \sigma^+ \) transitions to \( F' = 2, m_F' = m_F + 1 \) and \( F' = 3, m_F' = m_F + 1 \) are \(-\sqrt{1}/9\) and \(-\sqrt{2}/9\) respectively. The dipole matrix elements for \( \sigma^- \) transitions between \( F = 3, m_F = -1 \) and \( F' = 2, m_F' = m_F + 1 \) or \( F' = 3, m_F' = m_F + 1 \) are \(-\sqrt{1}/9\) and \(-\sqrt{1}/27\) respectively. The figure demonstrates a region of robust adiabatic population inversion to the upper hyperfine state of 5S\(_{1/2}\) shown in red in figure 12(b) for a broad range of the values of the chirp rate and the FWHM of the pulse.

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