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Population transfer via adiabatic passage in the Rydberg potassium atom*

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Using the time-dependent multilevel approach, we have calculated the coherent population transfer between the quantum states of potassium atom by a single frequency-chirped laser pulse. The result shows that a pair of sequential 'broadband' frequency-chirped laser pulses can efficiently transfer population from the initial state of the ladder system to the target state. It is also found that the population can be efficiently transferred to a target state and trapped there by using an 'intuitive' or a 'counterintuitive' frequency sweep laser pulse in the case of 'narrowband' frequency-chirped laser pulse. Our research shows that the complete population transfer is related to the pulse duration, chirp rate, and amplitude of the laser pulse.

Keywords: coherent population transfer, adiabatic passage, laser pulse, frequency sweep
PACC: 3100, 1240Q, 3280K

1. Introduction

In recent years, the design of laser pulse sequences to achieve efficient and robust population transfer between quantum states has been the subject of many theoretical and experimental studies.[1−3] This subject is relevant to many application, including spectroscopy, collision dynamics, and optical control of chemical reactions. Adiabatic rapid passage is a robust method of transferring population from one energy level to another.[4,5] If the frequency of radiation coupling two levels is swept through the resonance at a rate small compared with the square of the Rabi frequency, the efficiency of population transfer is very close to 100%. The high efficiency of each step allows population to be transferred through many levels by sequential adiabatic rapid passages.

Meerson and Friedland[6] suggested using a laser pulse, initially at the Kepler frequency which would be chirped to lower frequency later, to transfer atoms to a higher n state, leading to ionization at a lower microwave field. Recent studies of Bensky *et al*[7] and Wesdorp *et al*[8] have suggested that it might be more interesting to chirp the frequency in other direction. They have demonstrated that it is possible to induce electron-ion recombination into high-lying Rydberg states with half-cycle pulses, and the technique could be a way to produce antihydrogen.[9] However, the recombined atoms are left in high-lying states, and it would be useful to move them to lower-lying states. Recently, it has been extensively discussed using a single laser pulse to generate 'trapped' superposition of the ground states in the Λ atom.[10−14] The result shows that a single laser pulse with a chirped frequency can generate 'trapped' superposition of the ground states in the Λ atom when the width of the transform-limited laser pulse envelope frequency spectrum (without chirp) is smaller but the peak Rabi frequency of the pulse is larger than the frequency interval between the two ground states of the Λ atom.[14] However, these analytical studies were performed in rotating-wave approximation in an ideal system which included only three coupled states, while the effects of other states were ignored. Moreover, when they discussed the interaction of the Λ atom with the "broadband" laser pulse, two transition dipole elements were assumed to be identical with each other, i.e. $|d_{12}| = |d_{23}|$.[13,14] For a realistic physical system, other states' effects cannot be omitted, and the transition dipole elements of the two different states are possibly not identical with each other. Zhang *et al*[15]

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have investigated the coherent population transfer in Rydberg potassium atom via stimulated Raman adiabatic passage in the Λ atom, while there has been published no paper so far, to our knowledge, about the ladder system from higher to lower Rydberg states by a frequency-chirped laser pulse.

In this paper, we use a number of coupled states instead of only three states involved to calculate the population transfer in Rydberg potassium atom from higher to lower Rydberg states by a single frequency-chirped laser pulse. The potassium atom is exposed to a static electric field and interacts with a single frequency-chirped laser pulse. We expect to find the condition or probability of complete population transfer between the two Stark states of Rydberg potassium atom.

2. The time-dependent multilevel approach

A simplified energy-level diagram of potassium atom is depicted in Fig.1. In a 22p → 22s → 21p sequence (hereafter also denoted by $|1\rangle \rightarrow |2\rangle \rightarrow |3\rangle$) in the potassium atom, population transfer to the lower state is accomplished most efficiently when the instantaneous frequency of the laser pulse follows the spacing of the ladder system. It is assumed that only level 22p is initially populated.

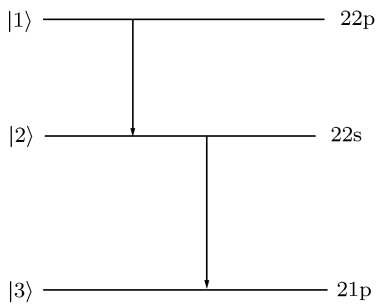


Fig.1. Energy levels of the three-state ladder in a potassium atom.

Consider the interaction of the system with an $E(t)$ laser pulse

$$E(t) = A(t)\cos[\omega_{\rm m}(t)\Delta t], \tag{1}$$

where $A(t)$ is the envelope of the pulse and

$$A(t) = A_0 \sin\Big(\frac{t^2}{2t_{\rm c}^2}\Big), \tag{2}$$

$\omega_{\rm m}(t) = \omega_{\rm m} + \beta\Delta t$, $\omega_{\rm m}$ is the carrier frequency of the laser pulse, β is the chirp rate and $\Delta t = t - t_{\rm c}$, with $t_{\rm c}$ being the time at maximum of the pulse envelope. In the presence of both a static electric field F_s in the direction along the z-axis and an electric field $E(t)$, the Hamiltonian for an outer electron of potassium atom takes the form

$$H = H_0 + zF_s + zE(t), \tag{3}$$

where H_0 is the field-free Hamiltonian and can be written as

$$H_0 = -\frac{1}{2}\nabla^2 + V(r). \tag{4}$$

The core potential of alkali-metal atom involving the effects of penetration and polarization is assumed to take the following form:[16]

$$V(r) = -\frac{1}{r}\Big(1 + \frac{\beta'}{(r+\gamma')^2} + \frac{k'}{(r+\xi')^2}\Big). \tag{5}$$

Using the potential model, the eigenfunction ϕ_i of H_0 can be obtained. We choose 117 field-free wave functions of potassium atom in the vicinity of 23p and 20s as a basis set. The wave function of the potassium atom in the presence of static electric field can be written as

$$\psi_i = \sum_{j=1}^{117} c_{ij}\phi_j\,. \tag{6}$$

Using ψ_i as the new basis set, the wave function of the potassium atom in the presence of static electric field, together with field $E(t)$ can be written as

$$\Psi(t) = \sum_{k=1}^{117} a_k(t)\psi_k {\rm e}^{-{\rm i}E_k t}, \tag{7}$$

where $a_k(t)$ is the coefficient of the expansion, which represents the amplitude of the transition probability. ${\rm e}^{-{\rm i}E_k t}$ contains the phase information that determines the moment when the resonance occurs. The wave function $\Psi(t)$ obeys the time-dependent Schrödinger equation (use atom unity)

$${\rm i}\frac{\partial}{\partial t}\Psi(t) = H\Psi(t). \tag{8}$$

Substituting expression (7) into Eq.(8), and solving the time dependent Schrödinger equation numerically, we can obtain $a_k(t)$. Then the probability of the outer electron inhabiting in state k can be

$$P_k = |a_k|^2. \tag{9}$$

Using the above formula, we can calculate the state-to-state transition probability from which we can observe the Rabi oscillation, the square wave of modulated Rabi oscillation and other novel features directly.

3. Results and discussion

Firstly, we consider the interaction of a 'narrow-band' laser pulse, the envelope spectrum of which (without chirp) is smaller than $|\omega_{12} - \omega_{23}|$, with the ladder system. We set the static electric field F_s = 28.9 V/cm and have ω_{12} = 382.26 GHz and ω_{23} = 468.65 GHz according to the Stark splitting. We first use the 'intuitive' sequence of the pulse ($\beta > 0$). We take the carrier frequency ω_m = 389.276 GHz because of the change of the electric field. Set the electric field A_0 = 15.5 V/cm and then tune the laser to follow the ladder level spacing, take $\omega_m(t)$ as the time-dependent frequency of the laser pulse and assume a linear chirp in time for $\omega_m(t) = 389.276 + \beta t$ with β = 1.0 GHZ/ns and t_c = 0.11 ns, then we will obtain the result of population transfer between the three states as illustrated in Fig.2(a). It is shown that the population is transferred from 22p state to 21p state in the case of the intuitive red–to-blue chirp, and two sequential adiabatic passages driven by a single frequency-swept laser pulse first transfer the population from the upper level to the intermediate level, and subsequently from the intermediate level to the lower level. Because the laser frequency is resonant with the first step of the ladder (22p–22s) in the beginning of the pulse, and with the second step (22s–21p) at the end with the change of the laser frequency, so with such an intuitive frequency sweep, 97% of the population can be transferred to the lower state, while halfway during the pulse most of the population resides in the intermediate 22s state. Our calculation also reveals the fine structures involved in the time-evolution process. From Fig.2(a), we can see that the population in the final state 21p slightly oscillates in time for a long time because of large maximum amplitude A_0 and pulse duration t_c.

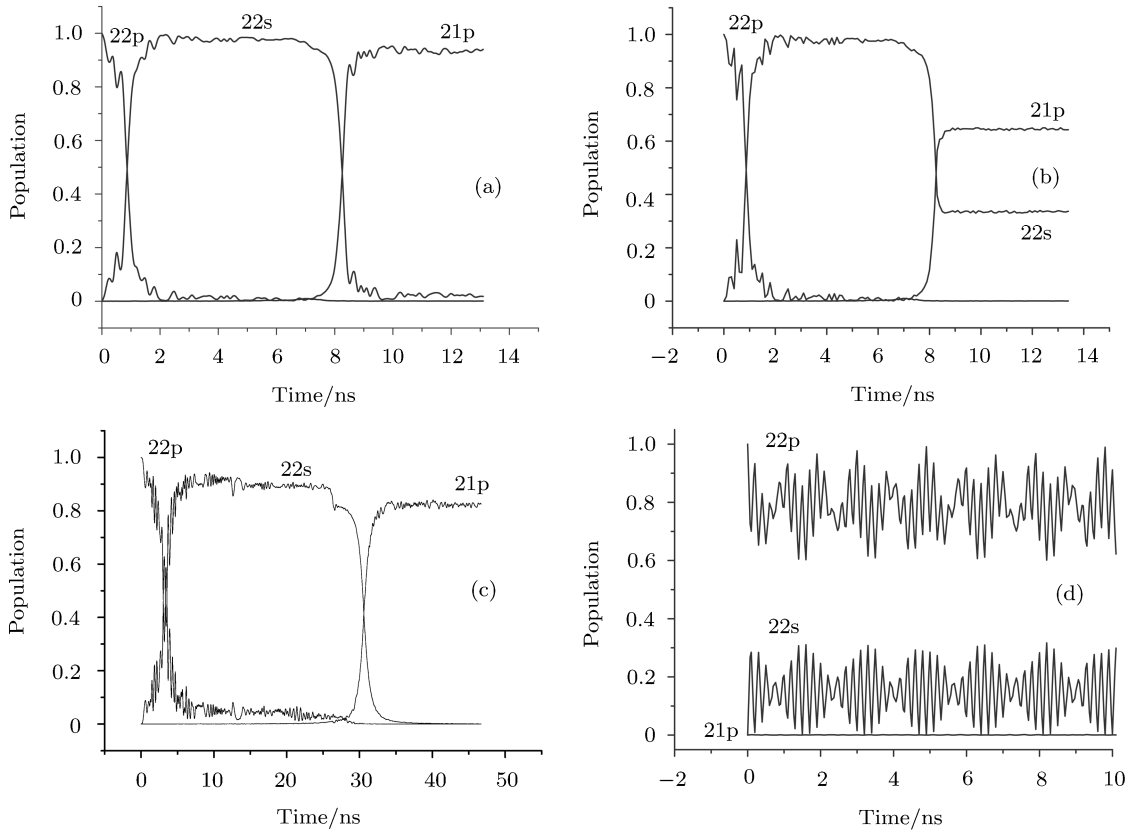


Fig.2. State population dynamics of the ladder system under action of (a) frequency-chirped laser pulse, with a chirp rate β = 1.0 GHZ/ns, (b) laser pulse without frequency chirp, $\beta = 0$. The parameters adopted are a duration of pulse t_c = 0.11 ns, and maximum amplitude A_0 = 15.5 V/cm. (c) State population dynamics of the ladder system under action of laser pulse with a duration time t_c = 0.21 ns. (d) Calculated time evolution of the populations of the levels with a constant $A(t)$ = 15.5V/cm. Other parameters are the same as those in (a).

We can see that a majority of the population can be transferred to the 21p state and trapped there when no chirp of the laser frequency is assumed, which has not been discussed before. We think that this is the consequence of change of the amplitude of the laser pulse. The carrier frequency is taken as the resonant transfer frequency between the 22p state and the 22s state in our calculation. Although the carrier frequency is constant in time, the amplitude of the laser pulse changes in time. When the amplitude is changed, the resonant transition frequency between the states will also be changed because of the dynamic Stark effect. Figure 2(b) shows the evolution of the populations of the levels with no chirped frequency.

If we set $t_c = 0.21$ ns and other parameters to be the same as those Fig.2(a), we can find that the population is not completely transferred from the upper state to the lower state. Only about 82% of the population can be transferred to the lower state. Figure 2(c) shows the evolution of the populations of the levels. Let the envelope of the pulse $A(t)$ be a constant and other parameters adopted be the same as those in Fig.2(a), then we will find that the population is not transferred from the upper state to the lower state. We can see that the populations in states 22p and 22s slightly oscillate in time for a long time because of the change of the chirped laser frequency and pulse duration t_c. Figure 2(d) shows the evolution of the populations of the levels with a constant $A(t)$.

Now, we consider the interaction of a counterintuitive blue-to-red frequency sweep ($\beta < 0$) laser pulse with the ladder system. It is noted that a complete population transfer can also be achieved by applying a reversed, 'counterintuitive' frequency sweep. Set $\omega_m(t) = 468.65 + \beta t$, $\beta = -10$ GHZ/ns, $t_c = 0.096$ ns and $A_0 = 20.5$ V/cm, then we can obtain the result of population transfer between the three states as illustrated in Fig.3(a).

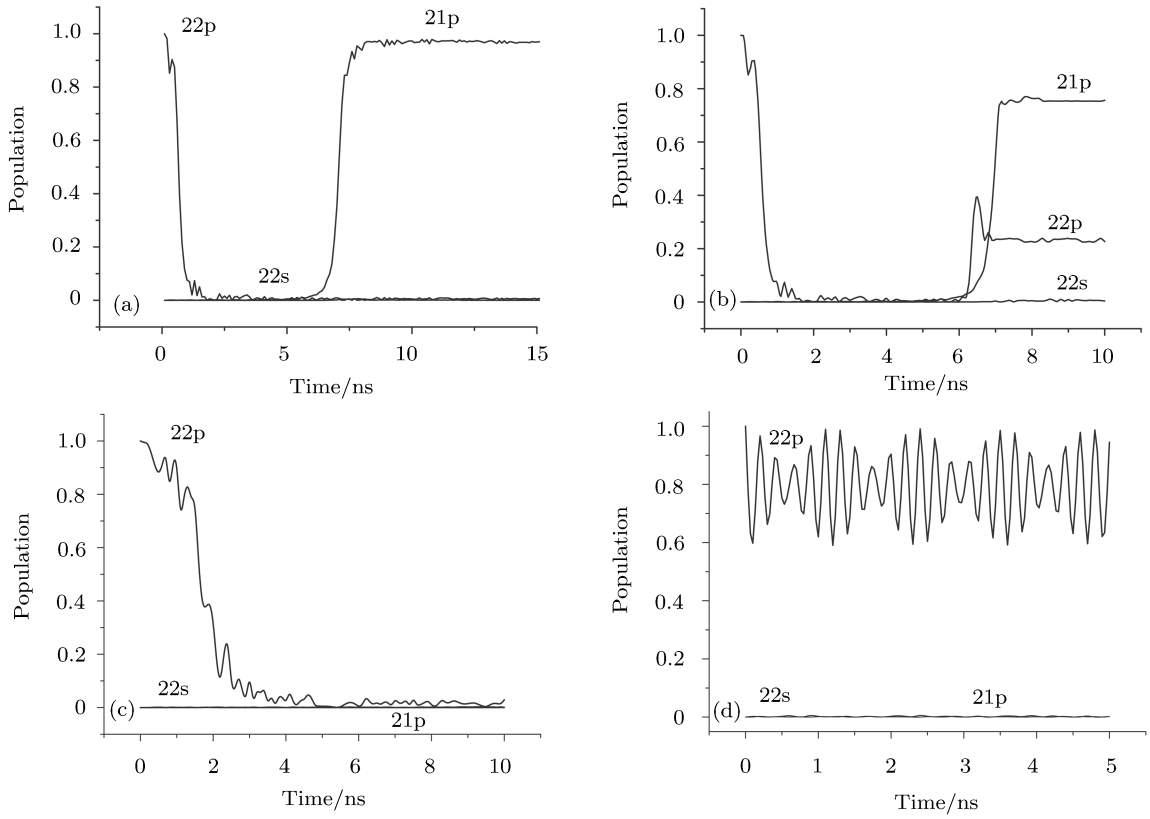


Fig.3. State population dynamics of the ladder system under action of (a) frequency-chirped laser pulse with a chirp speed $\beta = -10$ GHZ/ns, (b) laser pulse without frequency chirp with $\beta = 0$. The parameters applied are a duration of the pulse $t_c = 0.096$ ns, maximum amplitude $A_0 = 20.5$ V/cm. (c) State population dynamics of the ladder system under action of laser pulse with a duration time $t_c = 0.16$ ns. (d) Calculated time evolution of the populations of the levels with a constant $A(t) = 20.5$ V/cm. Other parameters are the same as those in (a).

From Fig.3(a), we can see that the laser pulse is first coupled with the two initially unpopulated states 22s and 21p, later this coherently superposed state is coupled to the state 22p. With this reason, during the pulse only a little of population resides in the intermediate state 22s. Finally, 98% of the population is transferred to the lower state (21p) and trapped there. Moreover, we can observe that there is no population in the three states discussed from 0.7 ns to 6.3 ns. We think this is the effect of other states. There exist a lot of levels between states 22p and 21p because of Stark splitting, when the frequency is chirped, the population in state 22p has been transferred to other states, but the population will be transferred to the state 21p finally with the change of frequency and amplitude of the laser pulse. If we let the laser frequency be a constant and other adopted parameters are the same as those in Fig.3(a), we find that a majority of the population can also be transferred from the upper state to the lower state. Figure 3(b) shows the evolution of the populations of the levels with a constant frequency. From Fig.3(b) we can see that most of the population can be transferred to the 21p state and trapped there when no chirp of the laser frequency is assumed. The reason is similar to that for the case of the "intuitive" sequence of the pulse.

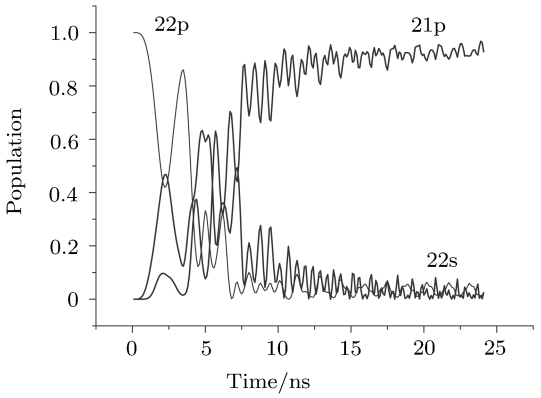


Fig.4. Dynamics of the state populations n_j, $j = 1, 2, 3$, during the action of two sequential frequency-chirped laser pulses in the case of the broadband pumping. The atom is assumed to be in the 22p state initially. The parameters adopted are a duration of pulse $t_c = 0.95$ ns, maximum amplitude $A_0 = 18.59$ V/cm, and chirp rate $\beta = -0.02$ GHz/ns.

If we set $t_c = 0.16$ ns and other parameters to be the same as those in Fig.3(a), we find that the population is not transferred from the upper state to the lower state. Figure 3(c) shows the evolution of the populations of the levels. If we let the envelope of the pulse $A(t)$ be a constant and other parameters adopted be the same as those in Fig.3(a), then the population will be not transferred from the upper state to the lower state. We can see that the population in the state 22p slightly oscillates in time for a long time because of the change of the chirped frequency and pulse duration t_c. Figure 3(d) shows the evolution of the populations of the levels with a constant $A(t)$. From Figs.2 and 3 we can see that the efficient transfer is greatly related to the maximum amplitude A_0, chirp rate β and pulse duration t_c for the "intuitive" sequence of the pulse and the "counterintuitive" sequence of the pulse.

Then, we consider the interaction of a 'broadband' pulse, the envelope spectrum of which (without chirp) is broader than $|\omega_{12} - \omega_{23}|$, with the ladder system, which has not been discussed so far. In this situation, the laser pulse interacts simultaneously with transitions both between the initial state and intermediate state, and between intermediate state and final state. This means that the condition of the two-photon (Raman) resonance is fulfilled during the interaction of the ladder system with the "broadband" frequency-chirped laser pulse. The interaction in this case is similar to the interaction of the atom with a bichromatic frequency-chirped laser pulse consisting of two laser pulses having identical envelopes and identical frequency chirps and the carrier frequencies shifted by an amount of the frequency distance between the initial state and the final state. We take the static electric field $F_s = 305.9$ V/cm and have $\omega_{13} = 0.4452$ GHz, $\omega_{12} = 423.0472$ GHz, and $\omega_{23} = 423.4924$ GHz. Set $A_0 = 18.59$ V/cm, $t_c = 0.95$ ns, $\beta = -0.02$ GHz/ns, and $\omega_0 = 423.4924$ GHz and the atom be in the state 22p initially, then we can obtain the result of the population transfer between the three states as illustrated in Fig.4. As a consequence of the interaction with a single "broadband" frequency-chirped laser pulse in 7.3 ns, about 44% of the population in the state 22p is transferred to the state 22s, 51% of the population is transferred to the state 21p. In the following time, a sequential laser pulse identical with the first one again collects about 94% of the population of the ladder system into the final state 21p that is empty initially. This is a consequence of the simultaneous resonant interaction of the single frequency-chirped laser pulse with transitions both between the initial state and intermediate state, and between intermediate state and final state that takes place only if the Fourier spectrum of the pulse envelop (without chirp) is wider than the fre-

quency interval between the initial state and final state in the ladder system. In the ladder system studied, the adiabatic condition is satisfied and $\Omega t << 1$.[17,18] In Ref.[14], when only three states are considered and the transition dipole elements are assumed to be identical with each other i.e. $|d_{12}| = |d_{23}|$, the population can be completely transferred to the initially empty ground state as a consequence of the interaction of a sequence of two not-overlapping frequency-chirped laser pulses with the Λ atom. In our calculation, because other states' effects are considered and the transition dipole elements are nonidentical with each other, the population cannot be completely transferred to the state 21p finally. We can see that the population in the state 21p slightly oscillates for a long time because of the change of the amplitude and frequency of the laser pulse.

4. Conclusions

We have presented the results of an analysis of the interaction of a single frequency-chirped laser pulse with a Rydberg potassium atom using a number of coupled states instead of only three states involved. Our calculation shows that the population can be completely transferred to a target state and trapped there in the case of 'narrowband' frequency-chirped laser pulse when $\beta > 0$; the population can also be transferred to a target state instantaneously when $\beta < 0$; the population transfer is incomplete or slow when $\beta = 0$. In addition, we have studied the interaction of a 'broadband' frequency-chirped laser pulse with the ladder system. In the situation, two-photon resonance transition occurs; a sequential 'broadband' laser pulse identical with the first one can again collect atomic population into the final state that is empty initially. In the process of our calculation, it is noted that population transfer is related to a pulse duration t_c, maximum amplitude A_0, and chirp rate β. Only when they are tuned to an appropriate value, can an effective population transfer take place.

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