Vol 15 No 7, July 2006
1009-1963/2006/15(07)/1497-05

Chinese Physics

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Coherent population transfer in Rydberg potassium atom by a single frequency-chirped laser pulse*

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(Received 28 August 2005; revised manuscript received 16 October 2005)

By using the time-dependent multilevel approach, we have calculated the coherent population transfer among the quantum states of potassium atom by a single frequency-chirped laser pulse. The results show that the population can be efficiently transferred to a target state and be trapped there by using an 'intuitive' or a 'counter-intuitive' frequency sweep laser pulse in the case of 'narrowband' frequency-chirped laser pulse. It is also found that a pair of sequential 'broadband' frequency-chirped laser pulses can efficiently transfer population from one ground state of the Λ atom to the other one.

Keywords: frequency-chirped laser pulse, coherent population transfer
PACC: 3100, 1240Q, 3280K

1. Introduction

The possibility of coherent transfer of the atomic population into a given quantum state or generation of a given superposition atomic state by laser radiation– that is, coherent laser control of the quantum states– is an important problem to be addressed in a variety of fields of contemporary science and technology such as chemical reaction dynamics,[1] laser-induced cooling[2] and atom optics.[3] An effective method for coherent manipulation of atomic quantum states is demonstrated using two laser pulses in the scheme of stimulated Raman adiabatic passage (STIRAP).[4−10] However, at least two laser pulses are involved in the processes of the coherent population transfer during STIRAP in previous considerations. Recently, using a single laser pulse to generate 'trapped' superposition of the ground states in the Λ atom has been extensively discussed.[11−15] The result showed 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.[15] 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. Otherwise, when they discussed the interaction of the Λ atom with the 'broadband' laser pulse, two transition dipole elements were assumed to be equal to each other: $|d_{12}| = |d_{23}|$.[14,15] For a realistic physical system, other states' effects cannot be omitted, and the transition dipole elements about the two different states cannot be equal. In this paper, using a number of coupled states instead of only three states involved, we will calculate the population transfer among the quantum states of Rydberg potassium atom irradiated 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 hope 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 three-level Λ system of a potassium Rydberg atom has three states in a static electric field. They are adiabatically connected to the zero-field states 21s, 19f and 19g, which are all shown in Fig.1. It is assumed that only level 21s is initially populated.

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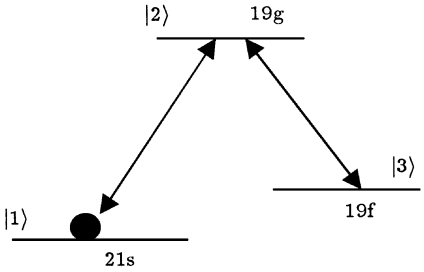


Fig.1. The scheme of the Λ-atom level.

Consider the interaction of the system with the laser pulse

$$E(t) = A(t)\cos[(\omega_0 + \chi\Delta t)\Delta t], \tag{1}$$

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

$$A(t) = A_0\exp(-t^2/2T_c^2). \tag{2}$$

In the above expressions, $\omega = \omega_0 + \chi\Delta t$ is the central frequency, $\chi = \mathrm{d}\omega/\mathrm{d}t$ is the chirp rate and $\Delta t = t - T_c$, where T_c is the time at maximum of the pulse envelope. In the presence of a static electric field F_s and a laser field $E(t)$ in the direction along the z-axis, the Hamiltonian for the 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 the alkali-metal atom, involving the effects of penetration and polarization, is assumed to be in the form[16]

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

Using this potential model, the eigenfunction ϕ_i of H_0 can be obtained. We choose 78 field-free wavefunctions of potassium atom in the vicinity of 21s and 19g as a basis set. The wavefunction of the potassium atom in the presence of a static electric field can be written as

$$\psi_i = \sum_{j=1}^{78} c_{ij}\varphi_j. \tag{6}$$

By using ψ_i(i=1, 2, 3, ...,78) as the new basis set, the wavefunction of potassium atom in the presence of a static electric field and field $E(t)$ can be written as

$$\psi(t) = \sum_{k=1}^{78} a_k(t)\psi_k \mathrm{e}^{-\mathrm{i}E_k t}, \tag{7}$$

where $a_k(t)$ is coefficient of the expansion, which represents the amplitude of the transition probability.

The wavefunction ψ (t) obeys the time-dependent Schrödinger equation (in atomic mass units)

$$\mathrm{i}\frac{\partial}{\partial t}\psi(t) = H\psi(t). \tag{8}$$

Substituting Eq.(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 the state k is

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

Using the above formula, we can calculate the state-to-state transition probability.

3. Result and discussion

Consider the interaction of a 'narrowband' laser pulse, in which the envelope spectrum (without chirp) is smaller than ω_{13} for the Λ atom. We set the static electric field F_s =289V/cm and have ω_{12} =33.1023GHz, ω_{23} =25.1013GHz according to the Stark splitting. We first use the 'intuitive' sequence of the pulse ($\chi < 0$). Setting A_0 =65V/cm, T_c =15ns, χ=–1GHz/ns, ω_0=33.1023GHz, we can obtain the results of population transfer among the three states as illustrated in Fig.2. From Fig.2(b), we observe that the population can be efficiently transferred to the state 19f, which is the same as the result of dressed-state analysis in Ref.[15]. It follows from the above consideration that the laser pulse frequency has to be swept first through the resonance with the transition from the initially occupied ground state 21s to the excited state 19g, and only afterwards through the transition between the initially empty ground state 19f and the excited state 19g. Our calculation has revealed the fine structures involved in the time-evolution process. From Fig.2(b), we can see that the population in the state 19f slightly oscillates with time for a long time because of large maximum amplitude A_0 and pulse duration T_c.

Now, we consider the interaction of a counterintuitive red-to-blue frequency sweep ($\chi > 0$ laser pulse with the Λ atom, which have not been discussed to our knowledge. When we set A_0 =29V/cm, T_c =2.8ns, χ=0.1GHz/ns, ω_0 =33.1023GHz, we can acquire the result as shown in Fig.3. From Fig.3(b), it can be seen that 99.87% of population in the state 21s is transferred to a target state and trapped there. In

this case, the pulse first couples the two initially unpopulated states 19g and 19f and creates a coherent superposition state; later this coherently superposed state is coupled to the state 21s.

From Fig.2(c) and Fig.3(c) we can see that a majority of the population can be transferred to the 19f state and trapped there when no chirp of the laser frequency is assumed, which is different from the result in Ref.[15]. We think that this is the result of changing the amplitude of the laser pulse. The carrier frequency is taken as the resonant transfer frequency between the state 21s and the state 19g in our calculation. Although the carrier frequency is unchanged with time, the amplitude of the laser pulse changes with time. When the amplitude is changed, the resonant transition frequency between the states also will be changed because of the dynamic Stark effect. In the beginning, the carrier frequency is smaller than both the resonant transfer frequency between the states 19g and 19f and between the states 21s and 19f. The resonant transfer frequency between the states 19g and 19f will decrease with time at reduced amplitude, and first reduce to the carrier frequency at the time when the pulse couples the two states and creates a superposed state. Because the amplitude nears to zero and the dynamic Stark effect is negligible in the final stage, the resonant transfer frequency between the states 21s and 19g approaches to the carrier frequency, thus this superposed state is subsequently coupled to the state 21s.

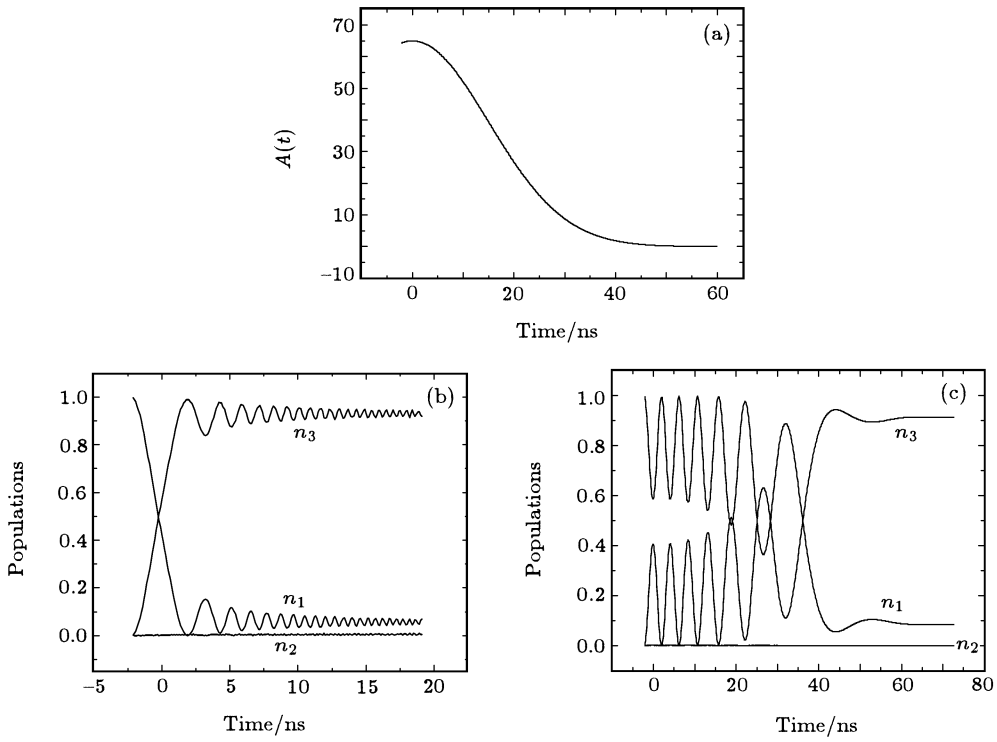


Fig.2. (a) The shape of the laser pulse. State population dynamics of the Λ atom under action of (b) frequency-chirped laser pulse, chirp speed χ=–1GHz/ns, (c) laser pulse without frequency chirp, χ=0. The parameters used are duration of the pulse T_c =15ns, the maximum amplitude A_0 =65V/cm.

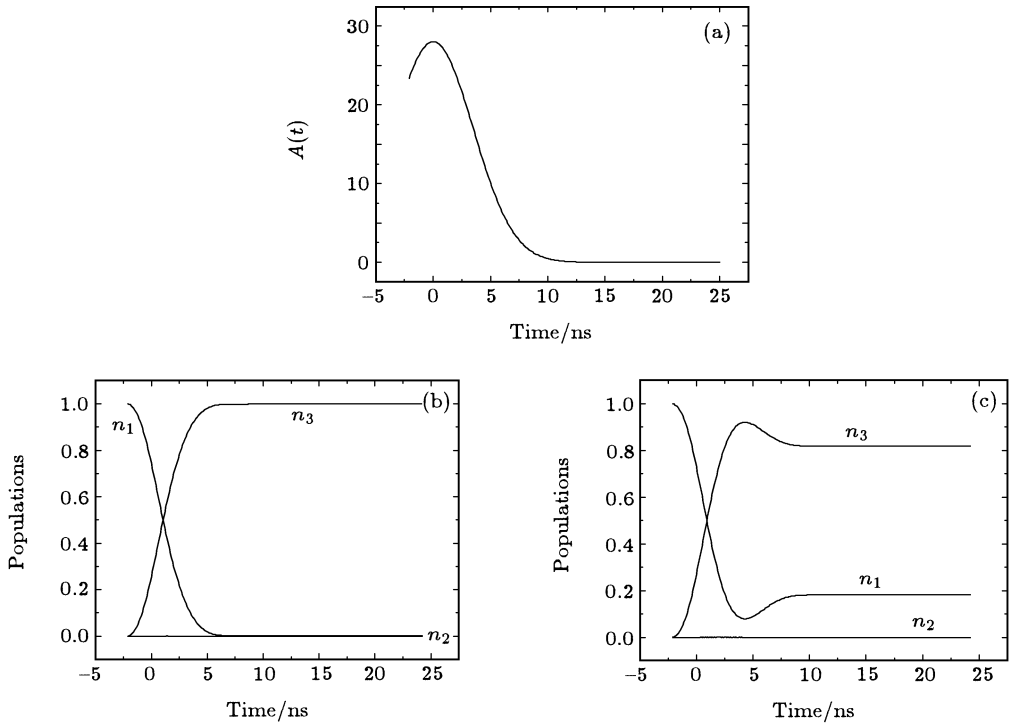


Fig.3. (a) The shape of the laser pulse. State population dynamics of the Λ atom under action of (b) frequency-chirped laser pulse, chirp speed χ=0.1GHz/ns, and (c) laser pulse without frequency chirp, χ=0. The parameters used are duration of the pulse T_c =2.8ns, the maximum amplitude A_0 =29V/cm.

Then, we consider the interaction of a 'broadband' pulse, in which the envelope spectrum (without chirp) is broader than ω_{13}, with the Λ atom. In this situation, the laser pulse interacts simultaneously with both transitions between the two ground states and the excited one. This means that the condition of the two-photon (Raman) resonance is fulfilled during the interaction of the Λ atom with the 'broadband' frequency-chirped laser pulse, which 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 will be shifted by an amount of the frequency distance between the two ground states.[14] We take the static electric field F_s=305.5V/cm and ω_{13}=0.9767GHz, ω_{12} = 26.5744GHz, ω_{23} = 25.5977GHz. Setting A_0 = 85V/cm, T_c = 0.1ns, χ = −25GHz/ns, ω_0 = 26.5744GHz, and assuming the atom is in one of the ground states (n_1=1, n_3=0) initially, we can obtain the result of the population transfer among the three states as illustrated in Fig.4. As a result of the interaction with a single 'broadband' frequency-chirped laser pulse, 40.5% of the population in the state 21s is transferred to the state 19f, and 46.6% of which is still in the state 21s. A sequential laser pulse identical with the first one again collects 81.6% of the population of the Λ atom into the ground state 19f which is empty initially (see Fig.4(b)). So a pair of sequential "broadband" frequency-chirped laser pulses can efficiently transfer population from one ground state of the Λ atom to the other one. In the Ref.[15], when only three states was considered and the transition dipole elements were assumed to be equal to each other: $|d_{12}| = |d_{23}|$, the population had been completely transferred to the initially empty ground state, as a result 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 is considered and the transition dipole elements is unequal, the population cannot be completely transferred to the state 19f finally.

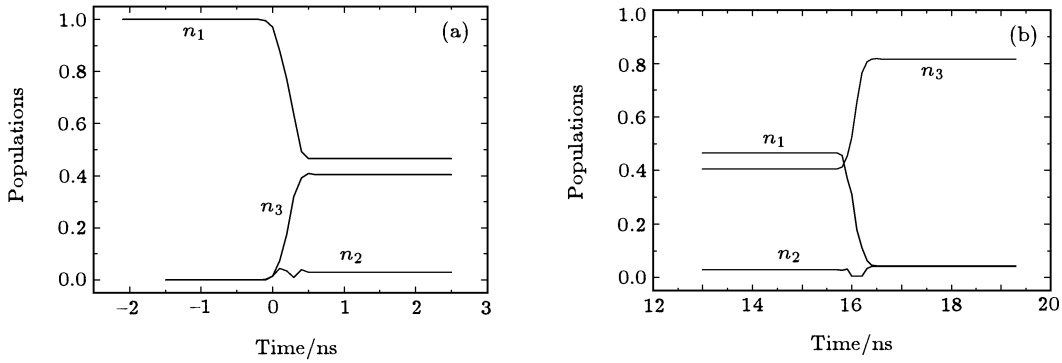


Fig.4. Dynamics of the state populations n_j,j=1, 2, 3, during the action of two sequential frequency-chirped laser pulses in the case of the 'broadband' pumping: (a) during the first pulse and (b) during the second pulse. The atom is assumed to be in the state 21s initially. The parameters used are duration of the pulse T_c =0.1ns, the maximum amplitude A_0 =85V/cm, the chirp speed χ=–25GHz/ns.

4. Conclusion

In conclusion, we have presented the calculated results 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 calculations show that in the case of 'narrowband' frequency-chirped laser pulse the population can be completely transferred to a target state and be trapped there when $\chi > 0$; the population can be transferred to a target state instantaneously when $\chi < 0$; the population transfer is incomplete or slow when $\chi = 0$. In addition, we have also calculated the interaction of a 'broadband' frequency-chirped laser pulse with the Λ atom. In this situation, two-photon resonance transition occurs; a sequential 'broadband' laser pulse identically with the first one can again collect atomic population into the ground state that is empty initially. In the process of our calculation, we found that population transfer was related to the pulse duration T_c, the maximum amplitude A_0 and the chirp rate χ. Only when they are tuned to an appropriate value, an effective population transfer could take place.

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