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Coherent population transfer in Rydberg potassium atom via stimulated Raman adiabatic passage*

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By using the time-dependent multilevel approach, the coherent population transfer among the quantum states of potassium atom driven by partially overlapped pulse is studied. The result shows that the complete population transfer is related to both the pulse duration and the overlap, and takes place when the pulse duration and overlap each reach an adequate value, and at the same time population trapping is also formed.

Keywords: stimulated Raman adiabatic passage, coherent population transfer, delay time
PACC: 3100, 1240Q, 3280K

1. Introduction

Coherent population transfer has already been applied in diverse areas such as chemical reaction dynamics,[1] laser-induced cooling[2] and atom optics.[3] Therefore people are always interested in finding a way the transfer of population to some specific excited state takes place efficiently. For a three-state system, the process in which population is adiabatically transferred from an initial level to a final one without any loss to an intermediate level, is superior to any other scheme. The use of two consecutive π pulses can serve the same purpose, but the result is sensitive to the exact pulse areas and decay of the intermediate level.[4] However, the stimulated Raman adiabatic passage (STIRAP) works perfectly in the adiabatic limit and is insensitive to the pulse shape, intensity and frequency modulation. So the STIRAP has received a lot of attention.[5−10] It demonstrates that a remarkable counter-intuitive mechanism is workable, in which the interaction with Stokes pulse, which couples the intermediate and final level, happens first, followed by the interaction with the pump laser pulse , which couples the initial and intermediate levels.

The simplest model of the STIRAP is a three-level Λ system. Using this model, Kuklinski *et al*[11] investigated the behaviour of population transfer and gave the adiabatic conditions for generating coherent population transfer. Recently, the STIRAP theory has been generalized to the case in which the intermediate state is of auto-ionization[12] or is described by a continuum of levels.[13,14] However, these analytical studies have been performed in an ideal system which includes only three coupled states, while the effects of other states are ignored. To our knowledge, the quantitative calculations about a realistic physical system including other states' effects have not been reported. In this paper, using a number of coupled states instead of only three states involved, we calculate the population transfer among the quantum states of Rydberg potassium atom via the ST1RAP. The potassium is exposed to a static electric field and interacts with two partially overlapped lasers. 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

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A three-level Λ system has three states of a potassium Rydberg atom in an electric field. They are adiabatically connected to the 21s, 19f and 19g zero-field states, which are all labelled as shown in Fig.1. It is assumed that only level 21s is initially populated. A pump pulse p links the initial state 21s with an intermediate state 19g, which in turn interacts via a Stokes s with a final state 19f. The frequencies for the Stokes and pump lasers are ω_s and ω_p, respectively. Δp is the detuning of pump laser from the resonant 21s-19f transition and Δs is the detuning of the Stokes laser from the 19g–19f transition.

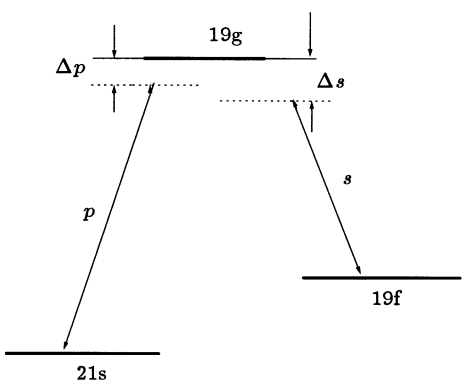


Fig.1. A three-level Λ system coupled by two lasers. The initially populated state 21s and the final state 19f are coupled by Stokes laser s and the pump laser p via an intermediate state 19g. The detuning of the pump and Stokes laser frequencies from the transition frequency to the intermediate state are Δp and Δs, respectively.

Consider the interaction of the system with the laser pulses

$$E(t) = E_p(t)\cos\omega_p t + E_s(t)\cos\omega_s t, \tag{1}$$

where $E_p(t)$ and $E_s(t)$[15] are the envelopes of the pulses:

$$E_s(t) = \begin{cases} 0 & t < 0, \\ A\sin^4(\pi t/T_s) & 0 < t < T_s, \\ 0 & t > T_s, \end{cases} \tag{2}$$

$$E_p(t) = \begin{cases} 0 & t < 0, \\ A\sin^4(\pi(t-\tau)/T_p) & 0 < t < T_p, \\ 0 & t > T_p. \end{cases} \tag{3}$$

In the above expression, τ is the delay time; T_s and T_p are the pulse durations of lasers. 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 the outer electron of potassium atom takes the form

$$H = H_0 + zF_s + zE(t), \tag{4}$$

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

$$H_0 = -\frac{1}{2}\nabla^2 + V(r). \tag{5}$$

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

$$V(r) = -\frac{1}{r}\left(1 + \frac{\beta'}{(r+\gamma')^2} + \frac{k'}{(r+\xi')^2}\right). \tag{6}$$

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 the static electric field can be written as

$$\psi_i = \sum_{j=1}^{78} c_{ij}\phi_j. \tag{7}$$

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

$$\Psi(t) = \sum_{k=1}^{78} a_k(t)\psi_k \mathrm{e}^{-\mathrm{i}E_K t}, \tag{8}$$

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

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

$$\mathrm{i}\frac{\partial}{\partial t}\Psi(t) = H\,\Psi(t). \tag{9}$$

Substituting Eq.(8) into Eq.(9), 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 can be

$$P_k = |a_k|^2. \tag{10}$$

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

3. Result and discussion

We set the static electric field F_s=289V/cm and tune the lasers to their respective resonant transition frequencies which are ω_s=25.1013GHz and ω_p=33.1023GHz, respectively. Set A=40V/cm, T_s=59.7ns, T_p=45.3ns, τ=28ns, and we can obtain the result of population transfer among the three states as illustrated in Fig.2. It is shown that the population

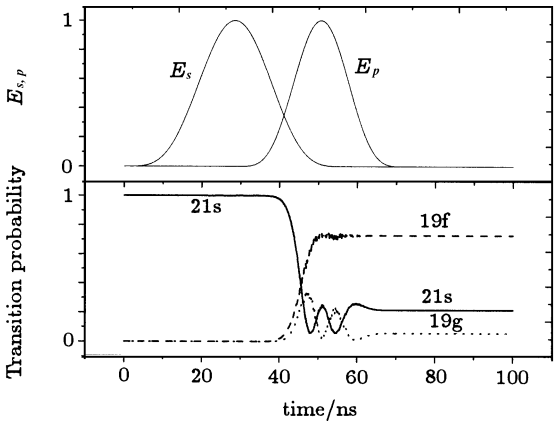


Fig.2. Population transfer in a three-level Λ system using a sequence of two overlapped lasers. Top frame: intensities of the pulse vs time. Bottom frame: evolution of the population in different levels vs time.

is transferred from the 21s state to the 19f state and is trapped there instantaneously. From the figure, we can well understand the STIRAP mechanism as follows. Initially, Stokes laser couples the two empty states, and thus it does not change the population of the state 21s. However, this laser creates the coherent superposition of the two initially unpopulated states 19g and 19f. When this coherently superposed state is subsequently coupled to the state 21s by the pump laser, a "trapped state" is formed—a state from which the pump laser cannot transfer the population to the intermediate state. Rather, the population is directly channelled into the state 19f.[17] It is noted that the transfer is incomplete and some population still remains in the 21s state. The incomplete transfer results from the fact that the process is partially adiabatic, in which a part of population is transferred first into the intermediate state 19g, and then back to the 21s state. If we change the pulse duration and overlap and set T_s=79.7ns, T_p=60.4ns, τ=35ns, and keep other parameters unchanged, then we can acquire the result as shown in Fig.3, from which it can be seen that 99.5% of the population in the state 21s is transferred to a target state. Our calculation is the same as the prediction in Ref.[18], in which it is proved that the adiabatic condition is related to the pulse duration, and the efficient population transfer can be obtained by increasing the pulse duration and choosing the adequate overlap.

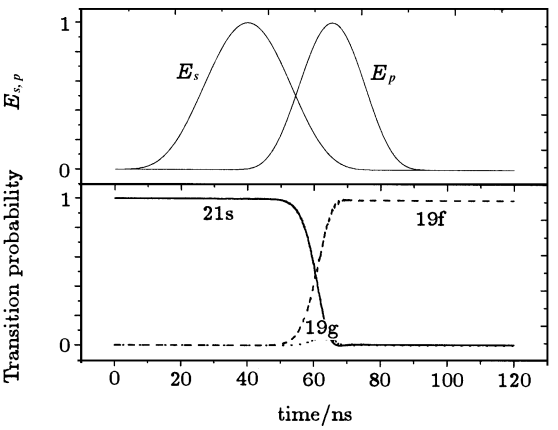


Fig.3. Evolution of population in different levels vs time after changing parameters T and τ. 99.5% of population is transferred to the final state.

Figure 4 shows the variation of the transition probability with Δp when Δs is set at 200MHz and 400MHz, respectively. The coherent population transfer occurs when the two-photo resonance condition ($\Delta p = \Delta s$) holds true. It is worthy of special notice that the two curves each have a second crest at the position of $\Delta p \approx 0$, and their peak values are not equal to each other. The reason for the appearance of the second crest is that when Δp is close to zero and Δs is small, the system is approximately in resonance, which can lead a part of population to transfer to the 19f state. So when the frequencies are tuned close to those in the single-photo resonance, more and more population transfers to the 19f state. With this notion in mind, we can deduce that the value of the second crest and the width of the two-photo resonance curve both increase as the frequencies are tuned close to those in the one-photo resonance. To test this, we plot the population transfer as a function of Stokes and pump laser detuning. The result is shown in Fig.5. From the figure, we can observe that when the frequencies are tuned to those in single-photo resonance, the width of the two-photo resonance curve increases, so does the value of the second crest.

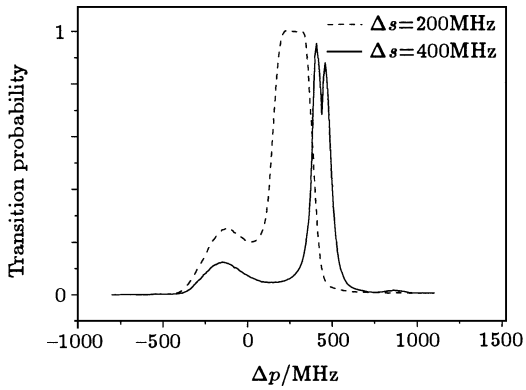


Fig.4. Population transfer from the 21s to the 19f states as a function of pump laser detuning Δp. The stokes laser detuning Δs is 200MHz (dotted line) and 400MHz (solid line), respectively.

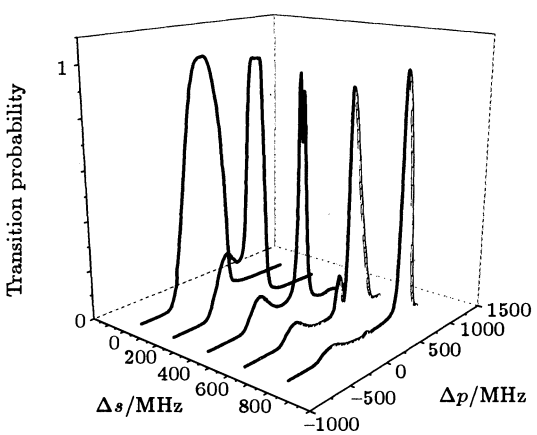


Fig.5. Coherent population transfer from the 21s to the 19f states as a function of Stokes and pump laser detuning Δs and Δp. The "STIRAP ridge" along the line can be seen when the two-photo resonance condition ($\Delta s = \Delta p$) holds true.

Changing the delay time τ between the two lasers and letting other parameters be the same as those in Fig.3, we obtain the result as shown in Fig.6 which shows the variation of the 19f state population as a function of τ. As we can see in Fig.6, when the delay time τ increases, the population of the 19f state increases dramatically and reaches a plateau near the optimum delay. This is an indication of the robustness of the transfer process. But the population of the 19f state begins to decrease as the delay time τ further increases. No population transfer occurs when no overlap exists between the two lasers. This can be understood as follows. Since a Stokes laser is not involved in the transfer process, no photon from both lasers simultaneously interacts with the atom. So the population cannot be transferred from the intermediate state to the final state. The pump laser excites atoms from the state 21s to the intermediate state from which they radiatively decay.[17] A very small fraction of it reaches the state 19f. Therefore nearly no transfer is observed when no overlap occurs between the two lasers. From the figure, we can see that the adequate overlap is necessary to the complete population transfer.

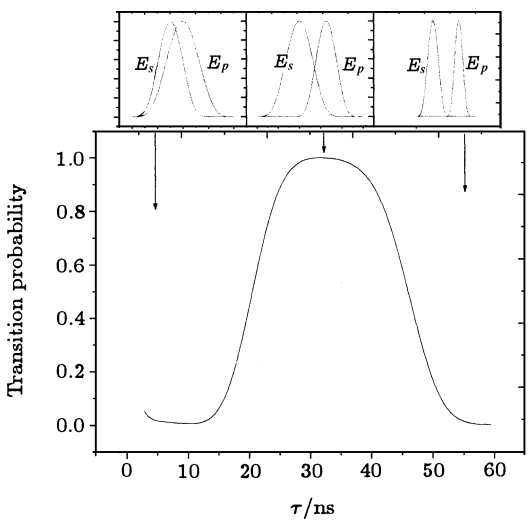


Fig.6. Population transfer from the 21s state to the 19f state,using a sequence of two overlapped laser as a function of the delay between the Stokes and the pump lasers (shown on top).

4. Summary

In summary, we have studied the coherent population transfer of potassium atom using a number of coupled states instead of only three states involved. Our calculations indicate that population transfer is related to the pulse duration and overlap. The population can be directly channelled to the final state, leaving no population in the intermediate state when the pulse duration T and delay τ each are tuned to an appropriate value. The laser frequencies each can be tuned off their respective resonant frequency, but the combination of pump and Stokes frequencies must be resonant with the two-photo Raman transition. The parameters that we obtained can be realized in the experiment, so it is possible to achieve the complete population transfer via the STIRAP.

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