Vol 16 No 7, July 2007
1009-1963/2007/16(07)/1947-05

Chinese Physics

© 2007 Chin. Phys. Soc.
and IOP Publishing Ltd

Numerical exploration of coherent excitation in three-level ladder systems*

Zhang Xian-Zhou(张现周)[a], Li Xiao-Hong(李小红)[b)c)†], and Yang Xiang-Dong(杨向东)[c]

[a] *Department of Physics, Henan Normal University, Xinxiang* 453007, *China*
[b] *College of Science, Henan University of Science and Technology, Luoyang* 471003, *China*
[c] *Institute of Atomic and Molecular Physics, Sichuan University, Chengdu* 610065, *China*

(Received 8 July 2006; revised manuscript received 13 October 2006)

The technique of stimulated Raman adiabatic passage (STIRAP) is used to transfer potassium atoms from the 22p state to the 21p Rydberg state through the intermediate state 22s. The results show that complete population transfer is related to pulse duration and overlap, and occurs when the pulse duration and overlap have adequate values. At the same time, population trapping is also formed. Complete population transfer can also occurs when the two-photon resonance condition ($\Delta_s = \Delta_p$) is met.

Keywords: STIRAP, coherent population transfer, pulse duration, overlap
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.[1−24] This problem is relevant to many applications, including spectroscopy, collision dynamics, and optical control of chemical reactions. A considerable number of studies have been devoted to the process of stimulated Raman adiabatic passage (STIRAP) in three-level[1−7] and multilevel systems.[11−19] The STIRAP process provides the possibility of effective population transfer using relatively simple experimental setup. It is achieved using a counterintuitive pulse sequence in which the Stokes pulse precedes the pump pulse and both pulses are tuned to resonance.

STIRAP is a modern technique employed widely to coherently transfer population in a three-level Λ system.[20] Using this model, Kukinski *et al*[1] 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 autoionizing[21] or is described by a continuum of levels.[22,23] However these analytical studies have been performed in the ideal system which includes only three coupled states, while the effects of others states are ignored. STIRAP is equally appropriate for a ladder system. Camp *et al*[24] experimentally investigated the population dynamics of a coherently excited three-level ladder system in which spontaneous emission are included and adiabaticity is not assumed. Cubel *et al*[25] experimentally investigated STIRAP excitation to transfer population from a lower Rydberg level to a higher one with an average, phenomenological efficiency of 50% by using a low-pressure vapour-cell magnetoptical trap. To our knowledge, definite calculations of coherent population from a higher level to a lower one about 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 rigorously calculate the population transfer of a three-level ladder system between quantum states of Rydberg potassium atom via STIRAP. The potassium is exposed to static electric field and interacts with two partially overlapping lasers. Some interesting results are obtained.

2. The time-dependent multilevel approach

The three-level ladder system consists of three states of a potassium Rydberg atom in an electric

*Project supported by the National Natural Science Foundation of China (Grant No 10574096).
†E-mail: lorna639@163.com

field. They are adiabatically connected to the 21p, 22s and 22p zero-field states which have been labelled as shown in Fig.1. It is assumed that only level 22p is initially populated. A pump pulse links the initial state 22p with an intermediate state 22s, which in turn interacts via a Stokes laser with the final state 21p. The frequencies for the Stokes laser and the pump laser are $\omega_{\rm s}$ and $\omega_{\rm p}$, respectively. $\Delta_{\rm s}$ is the detuning of the Stoke laser from the 21p–22s transition and $\hbar\Delta_{\rm s} = E_{22{\rm s}} - E_{21{\rm p}} - \hbar\omega_{\rm s}$. $\Delta_{\rm p}$ is the detuning of pump laser from the 22s–22p transition and $\hbar\Delta_{\rm p} = E_{22s} - E_{22p} + \hbar\omega_{\rm p}$.

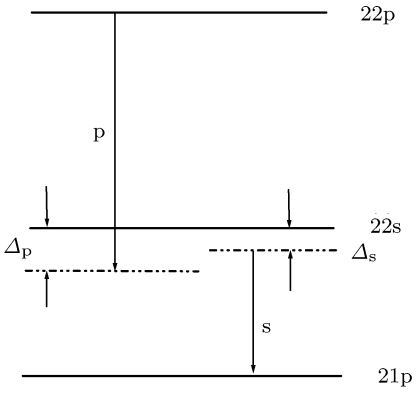


Fig.1. Schematic of the three-level ladder system coupled by two lasers. The initially populated state 22p and the final state 21p are coupled by Stokes laser and the pump laser via an intermediate state 22s. The detuning of the pump and Stokes laser frequencies from the transition frequency to the intermediate state are $\Delta_{\rm p}$ and $\Delta_{\rm s}$, respectively.

Consider the interaction of the system with $E_{\rm p}(t)$ and $E_{\rm s}(t)$ laser pulse:

$$E(t) = E_{\rm p}(t)\cos(\omega_{\rm p}t) + E_{\rm s}(t)\cos(\omega_{\rm s}t), \tag{1}$$

where $E_{\rm p}(t)$ and $E_{\rm s}(t)$[12] are the envelopes of the pulse.

$$E_{\rm s}(t) = \begin{cases} 0 & \text{for } t < 0, \\ A\sin^4(\pi t/T_{\rm s}) & \text{for } 0 < t < T_{\rm s}, \\ 0 & \text{for } t > T_{\rm s}, \end{cases} \tag{2}$$

$$E_{\rm p}(t) = \begin{cases} 0 & \text{for } t < 0, \\ A\sin^4[\pi(t-\tau)/T_{\rm p}] & \text{for } 0 < t < T_{\rm p}, \\ 0 & \text{for } t > T_{\rm p}, \end{cases} \tag{3}$$

where τ is the delay time, $T_{\rm p}$ and $T_{\rm s}$ are the pulse duration of lasers. In the presence of a static electric field $F_{\rm s}$ (the direction of the field being along the z-matrix) and electric field $E(t)$, the Hamiltonian for the outer electron of potassium atom takes the form

$$H = H_0 + zF_{\rm s} + zE(t), \tag{4}$$

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 take the form[26]

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

Using the potential model, the eigenfunction ϕ_i of H_0 can be obtained. We choose 117 field-free wavefunctions of potassium atom in the vicinity of 22p and 21p as a basis set. The wavefunction of the potassium atom in the presence of static electric field can be written as

$$\psi_i = \sum_{j=1}^{117} c_{ij}\phi_j. \tag{7}$$

Using $\psi_i (i = 1, 2, 3, \ldots, 117)$ as the new basis set, the wavefunction of the potassium atom in the presence of static electric field, together with field $E(t)$ can be written as

$$\Psi(t) = \sum_{k=1}^{117} a_k(t)\psi_k {\rm e}^{-{\rm i}E_k t}, \tag{8}$$

where $a_k(t)$ the is 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 wavefunction $\Psi(t)$ obeys the time-dependent Schrödinger equation (using atom units)

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

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 in the state k is

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

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

We set the static electric field $F_s = 28.9$ V/cm and tune the lasers to their respective resonance transition frequencies $\omega_s = 468.67$ GHz and $\omega_p = 382.28$ GHz. Set $A = 40$ V/cm, $T_s = 5.56$ ns, $T_p = 6.67$ ns, $\tau = 3$ ns, we obtain the result of population transfer between the three states as illustrated in Fig.2. It is shown that population is transferred from 22p to 21p and is trapped there. From the graph, we can well understand the STIRAP mechanism. Initially, Stokes laser couples the two empty states, and it does not change the population of the state 22p. However, this laser creates a coherent superposition of the two initially unpopulated states 21p and 22s. When this coherent superposition state is subsequently coupled to the 22p by the pump laser, a 'trapped state' is formed—a state from which the pump laser cannot transfer population to the intermediate state. Rather, the population is directly channelled into state 21p.[20] It is noted that the transfer is incomplete and some population remains in 22s. The incomplete transfer results from the fact that the process is partially adiabatic, in which a part of population is firstly transferred into the intermediate state 22s and trapped there.

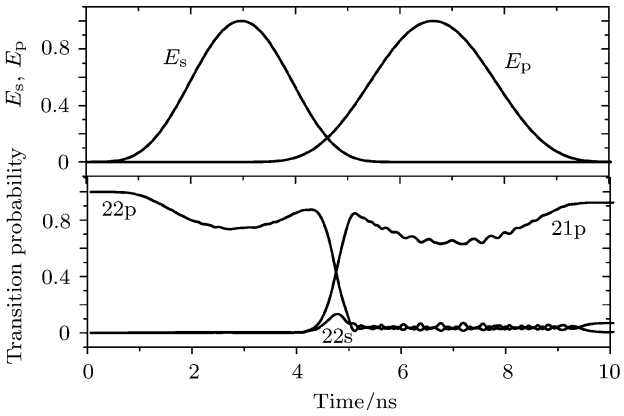


Fig.2. Population transfer in the three-level ladder system using a sequence of two overlapping lasers. Upper frame: intensities of the pulse vs time. Lower frame: evolution of the population in different levels vs time.

If we change the pulse overlap, and set $\tau = 1$ ns, leaving other parameters unchanged, we can obtain the result as shown in Fig.3, where 99.97% of the population of state 22p is transferred to the target state 21p. Our calculation result is the same as that of Ref.[3], which predicted that the adiabatic condition is related to pulse duration, and the efficient population transfer can be obtained by increasing the pulse duration and choosing adequate overlap.

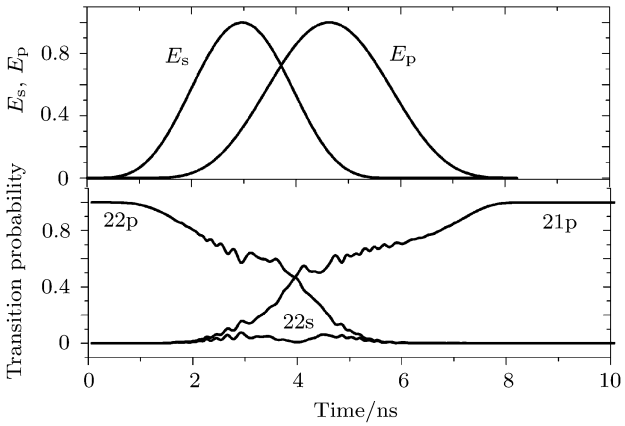


Fig.3. After the change of parameter τ, evolution of population in different levels. 99.97% of the population of state 22p is transferred to the final state. Note that population in the 22s level lags behind that in the 21p level, indicative of adiabatic coherent population transfer.

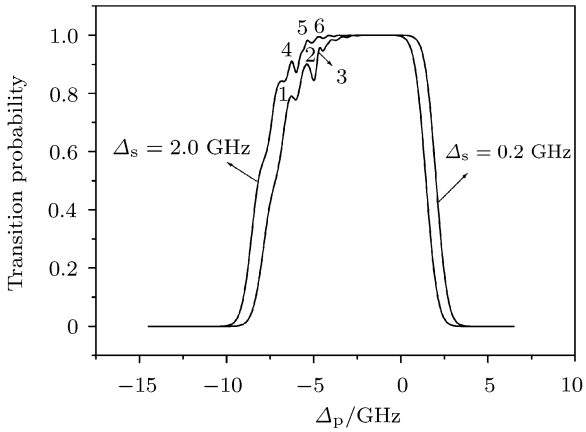


Fig.4. Population transfer from the 22p to the 21p state of potassium for a three-level ladder system (see Fig.1) as a function of pump laser detuning Δ_p. The Stoke laser detuning Δ_s is 0.2 GHz and 2.0 GHz, respectively.

Figure 4 shows the results of the transition probability with the variation of Δ_p when Δ_s is 0.2 GHz and 2.0 GHz. Coherent population transfer occurs when the two-photon resonance condition ($\Delta_s = \Delta_p$) is met. One should notice in particular that every curve has several crests and their heights are not equal. The reason for their appearance is that when Δ_p is close to zero and Δ_s is small, the system is approximately in resonance, which can lead to a part of population transferring to 21p state. When Δ_p gets closer to zero, more and more population transfers to 21p. So the height of crest 1 is lower than that of crests 2 and 3 when Δ_s takes the value 0.2 GHz. A similar result is

shown when Δ_s is 2.0 GHz. That is to say, more and more population transfers to 21p when the frequencies are tuned close to single-photon resonance.

Figure 5 shows the population transfer when only three states are coupled. The frequency of the Stroke laser is varied in steps while the frequency of the pump laser is tuned across the resonance. The broad feature is related to coherent population transfer, while the narrow feature results from population transfer to the final state by spontaneous emission induced by the pump laser. When the Stokes laser is tuned, say, about 2.0 GHz off resonance, the STIRAP transfer occurs when the pump laser is also tuned off resonance by about 2.0 GHz. Thus the broad features track the two-photon resonance. A high transfer efficiency is observed for detuning as large as 10.0 GHz off resonance with the intermediate level, 92% of the population of state 22p is transferred to the final state, which proved that the high transfer efficiency of STIRAP at two-photon resonance does not change within a certain range of detuning.

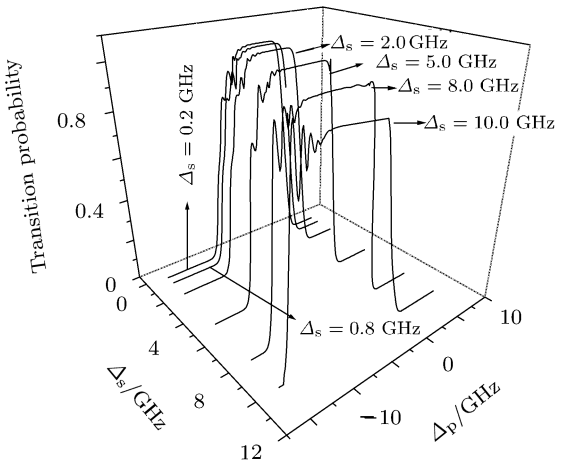


Fig.5. Coherent population transfer from the 22p to the 21p state of potassium for a three-level ladder system (see Fig.1) as a function of the Stokes and pump laser detuning Δ_s and Δ_p. The 'STIRAP ridge' along the line can be visible when the two-photon resonance condition ($\Delta_s = \Delta_p$) is met.

To prove that the population transfer occurs through the STIRAP mechanism, one must measure the transfer efficiency as the overlap between Stoke laser and pump laser is varied while all other parameters, such as laser frequencies and intensities, remain unchanged. Figure 6 shows the variation of 21p state population as a function of the time delay τ between two pulses. When the Stoke laser is shifted far upstream, so that there is no overlap with the pump laser, the Stokes laser does not participate in the transfer process, no photon from both lasers simultaneously interacts with the atom. The pump laser excites atoms from state 22p to the intermediate state 22s, there is small population transferred to the final state 21p because the energy level of the intermediate state is higher than that of the final state and the effect of radiative decay is little. When the delay time τ decreases, the transfer efficiency increases dramatically and reaches a plateau near the optimum delay. This is an indication of the robustness of the transfer process. But population of state 21p begins to decrease as the delay time τ further decreases. When the two lasers coincide, the transfer efficiency is only of the order of 50%. When the delay time is negative, that is to say that the pump laser acts first, followed after some time delay by the Stokes laser, which is the technique of stimulated emission pumping (SEP),[27] then some of the population of state 22p is transferred to the intermediate state 21p at the end of the pump interaction. If the Stokes laser is also strong enough, some of the population of the intermediate state is subsequently dumped into the final state 21p. Little of population transfer occurs when the Strokes laser is moved farther downstream of the pump laser and there is no overlap between them. So it is necessary to have an adequate overlap for the complete population transfer. The result is in agreement with that of Shore.[20]

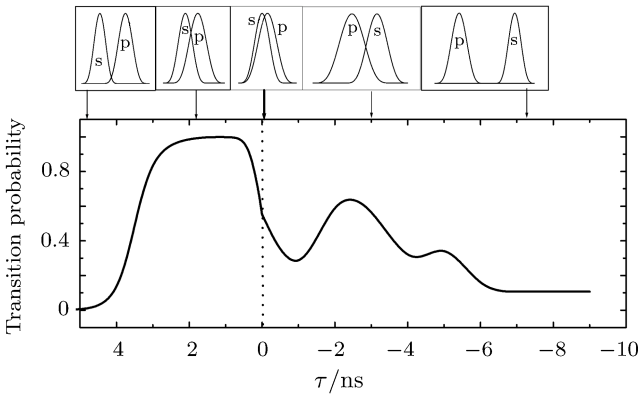


Fig.6. Population transfer from the 22p state to the 21p state, induced by a continuous laser, as a function of the overlap between the Stokes and the pump lasers (shown on top).

4. Summary

In conclusion, we have studied the behaviour of the coherent population transfer dynamics from the higher level 22p to the lower level 21p of potassium

atom by using a number of coupled states instead of only three states being involved. It was found that population transfer is related to pulse duration and overlap. When the pulse duration and overlap are tuned to a suitable value, population can be directly transferred to the final state 21p leaving no population in the intermediate state 22s. Individual laser frequencies can be tuned off their respective resonance frequency, but coherent population transfer occurs when the two-photon resonance condition ($\Delta_{\rm s} = \Delta_{\rm p}$) is met.

References

[1] Kuklinski J R, Gaubatz U, Hioe F T and Bergmann K 1989 *Phys. Rev.* A **40** 6741

[2] Carroll C E and Hioe F T 1990 *Phys. Rev.* A **42** 1522

[3] Gaubatz U, Rudecki P, Schiemann S and Bergmann K 1990 *J. Chem. Phys.* **92** 5363

[4] G1ushko B and Kryzhanovsky B 1992 *Phys. Rev.* A **46** 2823

[5] Kuhn A, Coulston G W, He G Z, Schiemann S, Bergmann K and Warren W S 1992 *J. Chem. Phys.* **45** 5297

[6] Laine T A and Stenholm S 1996 *Phys. Rev.* A **53** 2501

[7] Vitanov N V and Stenholm S 1996 *Opt. Commun.* **127** 215

[8] Fleischhauer M and Manka A S 1996 *Phys. Rev.* A **54** 794

[9] Vitanov N V and Stenholm S 1997 *Phys. Rev.* A **55** 648

[10] Vitanov N V and Stenholm S 1997 *Opt. Commun.* **135** 394

[11] Oreg J, Hazak G and Eberly J H 1985 *Phys. Rev.* A 32 2776

[12] Shore B W, Bergmann K, Oreg J and Rosenwaks S 1997 *Phys. Rev.* A **44** 7442

[13] Coulston G W and Bergmann K 1992 *J. Chem. Phys.* **96** 3467

[14] Oreg J, Bergmann K, Shore B W and Rosenwaks S 1992 *Phys. Rev.* A **45** 4888

[15] Shore B W, Bergmann K, Kuhn A, Schiemann S, Oreg J and Eberly J H 1992 *Phys. Rev.* A **45** 5297

[16] Smith A V 1992 *J. Opt. Soc. Am.* B **9** 1543

[17] Shore B W, Martin J, Fewell M P and Bergmann K 1995 *Phys. Rev.* A **52** 566

[18] Martin J, Shore B W and Bergmann K 1995 *Phys. Rev.* A **52** 583

[19] Band Y B and Julienne P S 1991 *J. Chem. Phys.* **95** 5681

[20] Bergmann K, Theuer H and Shore B W 1998 *Rev. Mod. Phys.* **70** 1003

[21] Nakajima T and Lambropoulos P 1996 *Z. Phys.* D **36** 17

[22] Carroll C E and Hioe F T 1992 *Phys. Rev. Lett.* **68** 3123

[23] Yatsenko L P, Unanyan R C, Bergmann K, Halfmann T and Shore B W 1997 *Opt. Commun.* **135** 406

[24] Camp H A, Shah M H, Trachy M L, Weaver O L and De Paola B D 2005 *Phys. Rev.* A **71** 053401

[25] Cubel T, Teo B K, Malinovsky V S, Guest J R, Reinhard A W, Knuffman B, Berman P R and Raithel G 2005 *Phys. Rev.* A **72** 023405

[26] He X H, Li B W, Chen A Q and Zhang C X 1990 *J. Phys.* B **23** 661

[27] Hamilton C H, Kinsey J L and Field R W 1986 *Annu. Rev. Phys. Chem.* **37** 493