Study on three level system population transfer

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Stimulated-Raman-adiabatic-passage (STIRAP) process provides an effective technique to transfer electron population from an initial state (e.g. ground state) to excited final state for both atoms and molecules. In this paper, we present the results of the study on electron population transfer in three level system. We have analyzed the effects of various conditions on the transfer process, such as the time delay of the two laser beams, two-photon off-resonance, one-photon off-resonance and the change of relative laser intensity. The numerical result is compared with experiment, and the reasons for the effects are also given.

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In order to understand the high temperature combustion media, atmospheric processes under non-equilibrium conditions, one has to investigate the collision dynamics of initial excitation state. Thus finding an efficient way to prepare atoms and molecules in excited state becomes a major topic in atomic and molecular physics. STIRAP process provides an efficient way to transfer the electron population via a decaying state without the detrimental effect of radiative loss, the efficiency can almost be 100%. Using analytic method, it is shown that complete transfer of electron population is possible through adiabatic passage under two-photon resonance[1]. Some numerical results also demonstrated that under two-photon resonance the transfer efficiency is almost 100% and experimental results agree with theoretical analysis[2,3]. Because of the thermally induced Doppler effect, it is almost impossible to achieve exact two-photon resonance for all atoms and molecules exposed to the laser light. Thus it is necessary to analyze the electron population transfer off two-photon resonance. M. P. Fewell *et al.* investigated the population transfer in the condition of off two-photon resonance using an analytic method[4]. They found that the population transfer would not occur through adiabatic passage even the detuning from two-photon resonance is very small. From experiment, we know the transfer efficiency is gradually decreased as the detuning off two-photon resonance is increased. Because there is energy crossing of the two dressed eigenstates, there could be a transition at that point. They found from that point on through adiabatic passage of another dressed state, the population transfer could still occur. Thus the population transfer could still be possible through diabatic process. But the analytic work could not show how much electron will make this transition and the transfer efficiency.

Here, we discuss the population transfer of three level system with numerical simulation. Our preliminary results, without considering the radiative loss of the second state, has been presented elsewhere[5], the theoretical results differs appreciably from the experimental results, especially in the intuitive ordering of the laser beams. In this paper, we include the radiative loss of the intermediate state, the results agree with the experiment much better. We have also examined the adiabatic condition to understand why there is a optimum displacement (or time delay) of the two laser beams.

We mainly discuss the three level system in Lambda configuration, one of the simplest STIRAP schemes. This system involves three nondegenerate states of an atom or molecule: the initial state, the intermediate state, and the final state, shown as $|1\rangle$, $|2\rangle$, and $|3\rangle$, respectively in Fig. 1, with energies of E_1, E_2, and E_3. Two overlapping pulses of coherent laser light induce the desired transition from initial state to final state. One laser, what we call 'pump' pulse couples the initial and intermediate states. The other, the 'Stokes' pulse, couples the intermediate and final states[6]. The Rabi frequency and carrier frequency of the pump pulse are $\Omega_{\rm p}$ and $\omega_{\rm p}$, respectively, while those of the Stokes pulse are $\Omega_{\rm S}$ and $\omega_{\rm S}$. 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.

In order to simplify the calculation, we make $E_1 = 0$. Using rotating wave approximation (RWA)[7] and dipole approximation[8], we can get the Hamiltonian of the system as

$$H = \frac{\hbar}{2}\begin{pmatrix} 0 & \Omega_{\rm p} & 0 \\ \Omega_{\rm p} & 2\Delta_{\rm p} - i\Gamma & \Omega_{\rm S} \\ 0 & \Omega_{\rm S} & 2\Delta_3 \end{pmatrix}, \tag{1}$$

where

$$\Delta_{\rm p} = E_2/\hbar - \omega_{\rm p}, \tag{2}$$

$$\Delta_{\rm S} = (E_2 - E_3)/\hbar - \omega_{\rm S}, \tag{3}$$

$$\Delta_3 = \Delta_{\rm p} - \Delta_{\rm S}, \tag{4}$$

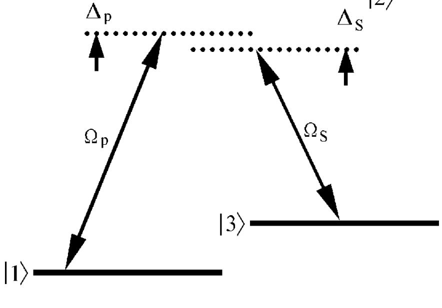

Fig. 1. Three level system in Lambda configuration.

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where Γ is the loss rate of level 2, defined as the reciprocal of the lifetime of the state.

Since each laser beam interacts with atom or molecule in time T, the Rabi frequencies are as

$$\Omega_{\mathrm{p}}(t) = \Omega_0 p(t - t_1), \tag{5}$$

$$\Omega_{\mathrm{S}}(t) = \Omega_0 p(t), \tag{6}$$

where

$$p(t) = \begin{cases} \sin^4(\pi t/T), & 0 < t < T \\ 0, & t \le 0, t \ge T \end{cases}, \tag{7}$$

t_1 is the time delay of pump pulse Ω_{p} relative to Stokes pulse Ω_{S}.

We choose the parameters with the same value as those in experiments done by Gaubatz[3], so the comparison can be made. Here, we make $\Omega_0 = 100$ MHz, $T = 170$ ns, $\Gamma = 80$ MHz and use Runge-Kutta method to solve time dependent Schrödinger equation of the Hamiltonian given above.

Figure 2 shows the efficiency of population transfer from level 1 to level 3 for $\Delta_{\mathrm{p}} = \Delta_3 = 0$ with different displacement of the two laser beams in the condition of two-photon resonance. Coordinates D/w_E is the separation of the two laser beams (D) to the waist of the laser beam (w_E)[3], the negative values indicating that the atoms or molecules encounter the Stokes pulse earlier than the pump pulse. Figure 2(a) is our theoretical calculation, and (b) experimental result[3]. From Fig. 2, we can see the transition is more efficient when the displacement is negative, that is the Stokes pulse proceeds before the pump pulse. Specially, we note that the most efficient transfer occurs near 1.0, besides this point, the efficiency decreases little by little. The numerical result agrees with the experimental result pretty well.

The results of the simulation and experiments above are based on the condition of two-photon resonance, which is one of the two basic conditions for complete population transfer as we know. So when the problem comes to why does the extremum exist, that is why does the best efficiency occur at only one point, we put our attention on another basic condition: adiabatic procedure.

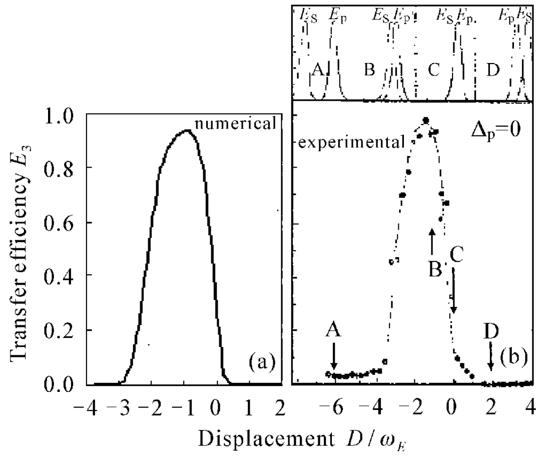

Fig. 2. The transfer efficiency as a function of displacement of the two lasers. (a) Theoretical result, and (b) experimental result.

The adiabatic condition can be expressed as: $\left|\frac{d\Theta}{dt}\right| \ll \left|\omega^0 - \omega^{\pm}\right|$, here we name them Dtheta and Domega respectively, where Θ is the mixing angle of the adiabatic state and ω^0 and $\omega^{\pm}$ are eigenvalues of the three dressed eigenstates, see Ref. [4]. In order to transfer the population completely, this condition should be maintained in the whole transferring process. Here, we calculate the ratio of $\left|\frac{d\Theta}{dt}\right|$ to $\left|\omega^0 - \omega^{\pm}\right|$ when the time delay of the two laser beams is -0.1, -0.2 and $-0.7T$ (T is the width of the laser pulse), respectively, which correspond to laser displacement D/w_E of -0.46, -0.93, and -3.22, respectively.

Figure 3 shows that the adiabatic condition was destroyed at the beginning and the end of the transfer process when time delay is greater than $-0.2T$, that is, the Stokes pulse comes before the pump pulse but the time delay of the two laser beams is less than $0.2T$. When the time delay of the two laser beams is $-0.7T$, the adiabatic condition was well at the beginning and the end, but in the middle of the process, $\left|\frac{d\Theta}{dt}\right|$ could not be neglected when compared to $\left|\omega^0 - \omega^{+}\right|$. On the other hand, when the time delay of the two lasers is $-0.2T$, what we call the optimum delay, the ratio is almost always zero in the whole process, shown in solid curve. That is why there is an optimum delay (or optimum displacement) of the two laser beams.

If Δ_3 varies ($\Delta_3 \neq 0$) while Δ_{p} is always 0, we call it two-photon off-resonance. In this case, we will see that the transfer efficiency of population will change with Δ_3.

Figure 4 shows the transfer efficiency in different Δ_3. When system is in two-photon resonance, it shows that the highest efficiency still occurs when $\Delta_3 = 0$. But we should really pay attention to is that, there are still transfer of population when $\Delta_3 \neq 0$. Here, we can draw a conclusion that M. P. Fewell's analysis about the occurring of diabatic transitions is right.

We considered the radiation loss of level 2 in calculating the effect of Δ_3, and we found that the efficiency is really smaller than that when the radiation loss is neglected. According to the analysis of B. W. Shore, there is no population in level 2 during the transfer process in two-photon resonance. The result indicates the incomplete ensurence of adiabatic transition. For the approximation is made without considering the radiation loss of level 2, it is damaged to some degree when the

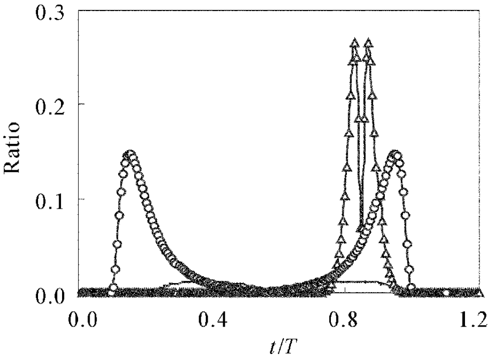

Fig. 3. The ratio of Dtheta to Domega in the transfer process when the time delay of the two lasers is -0.1, -0.2 and $-0.7T$ (T is period of the laser) shown as curve with ○, solid curve and curve with △, respectively.

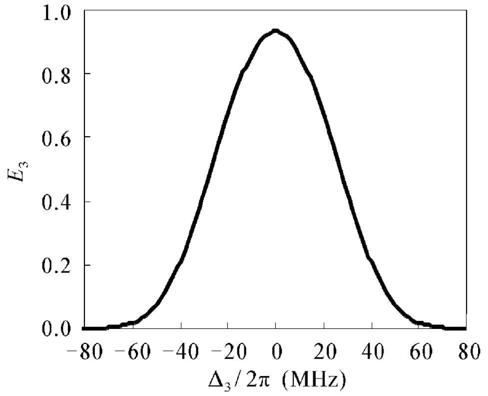

Fig. 4. The efficiency of population transfer of different Δ_3 when $\Delta_p = 0$ and the two laser beams are in optimum time delay.

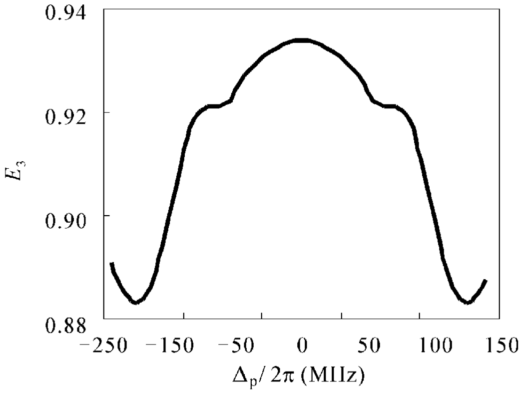

Fig. 5. Transfer efficiency of the populations from level 1 to level 3 for $\Delta_3 = 0$ versus Δ_p.

radiation loss is considered.

Figure 5 shows the transfer efficiency of population as a function of Δ_p for $\Delta_3 = 0$, that is one-photon off-resonance. From Fig. 5, we find high efficiency can still be got even Δ_p is very great (compared to Δ_3 in Fig. 4). This indicates that Δ_3 is more important than Δ_p in our calculation. In experiments, we should try our best to make Δ_3 to be 0 and can reduce the requirement to Δ_p.

In order to check how the efficiency depends on the relative laser intensity, we make the intensities of the two laser beams different, as

$$\Omega_p(t) = \Omega_1 p(t-\tau), \qquad (8)$$

We will find that the difference $\Delta\Omega = \Omega_1 - \Omega_0$ will influence the transfer efficiency. Figure 6 shows the result when $\Delta_p = 0$ and $\Delta_3 = 0$.

From Fig. 6, we can see that the influence of the difference is small on the whole and it can also be neglected when compared to the influence of Δ_3. But if we want to improve the efficiency as much as possible, we can make the difference to be larger than 0, that is, make the extremum of Ω_p larger than that of Ω_S slightly.

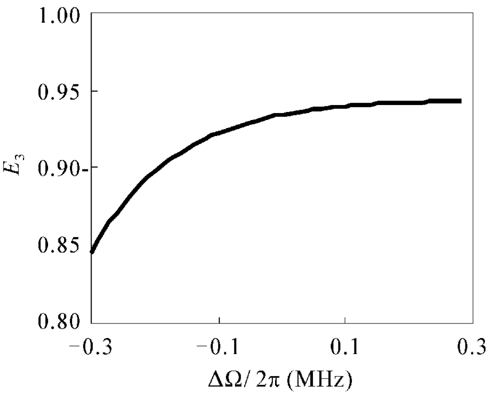

Fig. 6. Transfer efficiency of different differences of Rabi frequecies.

Having completed these calculations, we can find that we could use numerical methods to simulate the population transfer of the three level system, and we have got some satisfactory results. Based on the similar results with theoretical analysis and experiment we have got from the simulation, we also anticipate some possible results that have not been proved by experiments, but we are sure these results can give some direction to the next experiments.

In order to get the highest efficiency, we should ensure two main conditions: the best time delay of the two laser beams and two-photon resonance. The two-photon resonance or Δ_3 is more important than other condition such as Δ_p and $\Delta\Omega$. So we should put our attention to improve the condition of two-photon resonance in experiments. In addition, the main loss of efficiency comes from the radiation loss of the level 2, and it is the right reason why we could not transfer any population when pump pulse precedes Stokes pulse.

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References

1. J. R. Kuklinski, U. Gaubatz, F. T. Hioe, and K. Bergmann, Phys. Rev. A **40,** 6741 (1989).
2. U. Gaubatz, P.Rudecki, M. Becker, S. Schiemann, M. Külz, and K. Bergmann, Chem. Phys. Lett. **149,** 463 (1988).
3. U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann, J. Chem. Phys. **92,** 5363 (1990).
4. M. P. Fewell, B. W. Shore, and K. Bergmann, Aust. J. Phys. **50,** 281 (1997).
5. S. Yang and Y. Kuang, Chin. J. Atomic and Molecular Phys. (in Chinese) **19,** 196 (2002).
6. B. W. Shore, Cont. Phys. **36,** 15 (1995).
7. B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990) pp. 777 – 858.
8. F. H. Wang and S. W. Wang, *The Theory of Atomic and Molecular Excitation* (in Chinese) (Chengdu University of Science and Technology Publishing Co., Chengdu, 1991) pp. 77 – 80.