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Numerical exploration of population transfer of Rydberg-atom by single frequency-chirped laser pulse*

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This paper has calculated that Rydberg atoms can be transferred to states of lower principal quantum number by exposing them to a frequency chirped microwave pulse. The atoms experience the consequence: 70p-69s-68p-67s-66p by a constant amplitude field in the adopted model. This study shows that the complete population transfer is related to the chirp rate and the carrier frequency.

Keywords: population transfer, frequency sweep, B-splines, multilevel system
PACC: 3100, 3120, 3280

1. Introduction

The coherent transfer of the atomic population into a given quantum state is an important problem to be addressed in a variety of fields of contemporary science and technology.[1−3] It is relevant to optical control of chemical reactions, manipulation of Rydberg electron and spectroscopy. Adiabatic rapid passage (ARP) is an approximately 100% efficient way to transfer population from one state to another, in which the frequency of radiation coupling the two levels is swept through the resonance at a rate small compared to the square of the Rabi frequency. Meerson and Friedland[4] suggested that atoms can be transferred to a higher n state, sequentially can be ionized at a lower microwave field, using a blue-to-red chirped laser pulse initially at the Kepler frequency. Later the works of Bensky *et al*[5] and Wesdorp *et al*[6] suggested that it might be more interesting to chirp the frequency in other directions. Recently the ARP has been extensively discussed using a single pulse to trap population in a multi-level ladder atom. Lambert *et al*[2] demonstrated that Rydberg atoms could be transferred to states of lower principal quantum number by exposing them to a frequency chirped microwave pulse. In their experiment, Li Rydberg atoms had been moved from $n = 75$ to $n = 66$ using chirped 7.8 to 11.8 GHz pulses at the two-photon resonance between states differing in n by 1. Djotyan *et al*[7] proposed a scheme of population transfer between two ground states of the $\wedge$ atom without considerable excitation of the atom using a single frequency-chirped laser pulses. This scheme could be realized when the width of the transform-limited laser pulse envelope frequency spectrum (without chirp) must be smaller and the peak Rabi frequency of the pulse must be larger than that of the frequency interval between the two ground states of the atom. In their analytical studies, two transition dipole elements were assumed to be equal to each other, i.e. $|d_{12}| = |d_{23}|$.[7] But for a real system, this is not always satisfied.

Recently, B-splines have been widely applied to the calculation of atomic and molecular physics, and are useful to solve different problems in this realm.[8] Xi *et al*[9] were the first to use the B-splines to treat the ground and the lower excited states of atoms in external field. They calculated the energy levels of the ground and lower excited states of hydrogen in magnetic field of arbitrary strength. Rao *et al*[10] calculated the ground and the lower excited Stark resonances of hydrogen atom in super-strong electric fields using B-spline method.

In this paper, we numerically investigate the population transfer in the Rydberg hydrogen atom with a single frequency-chirped microwave pulse. We get the energy with high principal number by solving stationary Schrödinger equation using B-spline method. We expect to obtain the condition of complete population

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transfer in the multi-level ladder system.

2. Theory

Figure 1 depicts a simplified energy-level diagram of hydrogen atom, for simple form and rich meaning, we select the sequence: 66p-67s-68p-69s-70p.

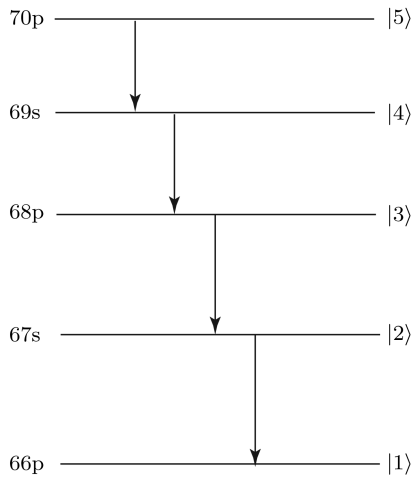


Fig.1. The sketch map of energy levels of ladder system in a hydrogen atom.

In the presence of a linear polarized constant amplitude microwave field $E(t)$, the Hamiltonian of the Rydberg electron has the form (unless units are explicitly given, we use atomic units):

$$H = H_0 + zE(t), \tag{1}$$

where H_0 is the field-free Hamiltonian: $H_0 = -\frac{1}{2}\nabla^2 + V(r)$ and $E(t) = A_0\cos(\omega(t)t)$, $\omega(t) = \omega_0 + \beta t$, ω_0 is the laser initial frequency, β is the chirp rate and A_0 is a constant.

The system wave function $\psi(t)$ obeys the Schrödinger equation:

$$\mathrm{i}\frac{\partial}{\partial t}\psi(t) = H\psi(t), \tag{2}$$

where $\psi(t)$ takes the form:

$$\psi(t) = \sum_{i=1}^{4} a_i(t)\phi \mathrm{e}^{-\mathrm{i}E_i t}, \tag{3}$$

and ϕ is the eigenfunction of H_0. Then the probability of the outer electron inhabiting in the state i can be written as

$$P_i = |\alpha_i|^2. \tag{4}$$

From this formula, we can obtain the state-to-state transition probability from which we can observe the famous Rabi oscillation and some other interesting features directly.

In order to get the eigenfunction and eigenvalue of H_0, we solve the stationary Schrödinger equation using the B-spline basis function. B-spline is a kind of piecewise polynomial, which possesses the characteristics of both analytical and numerical functions, and is a very important tool for numerical calculation. See Refs.[8] and [11] for detailed characteristics of B-spline.

Accounting for the central symmetry of the potential function of hydrogen atom, we find that the eigenfunction of H_0 has the following form:

$$\phi_{nlm}(\boldsymbol{r}) = \frac{U_{nl}(r)}{r}\mathrm{Y}_{lm}(\theta\varphi), \tag{5}$$

where n, l, m are principal, angular momentum and magnetic quantum numbers, respectively, and Y_{lm} is spherical harmonic function. Thus the radial function $U_{nl}(r)$ is a solution for the reduced Schrödinger equation:

$$\left[-\frac{\mathrm{d}^2}{2\mathrm{d}r^2} + \frac{l(l+1)}{2r^2} - \frac{1}{r}\right] U_{nl}(r) = E_{nl}U_{nl}(r), \tag{6}$$

with the boundary condition: $U_{nl}(0) = 0$.

Using B-spline basis set, we cam expand $U_{nl}(r)$ as:[9]

$$R_{nl}(r) = \sum_{i=1}^{N} c_i B_{i,k}(r), \tag{7}$$

where $B_{i,k}(r)$ is the ith B-spline of order k.

For a given fixed angular momentum l, substituting Eq.(7) into Eq.(6), and projecting on $B_{i,k}(r)$, we obtain a matrix equation:

$$H_l C = ESC, \tag{8}$$

where S is the overlap matrix. By means of diagonalizing the equation, we can get the radial function $U_{nl}(r)$ numerically.

In Table 1, we list the calculated eigenvalues of H_0 by B-spline technique and equation: $E = -1/2n^2$. It can be observed that the differences $|\Delta E|$ between our calculated energies and the results of equation $E = -1/2n^2$ are very small and are of the order of $< 10^{-13}$ Hartree.

It is straightforward to understand the process of population transfer using a picture based on the avoided crossings of levels dressed by microwave photons, a picture commonly used for Landau-Zener transition. To plot Fig.2, we have assumed that the energy

of each state, in the absence of any coupling, is given by[2,3,12]

$$E = -\frac{1}{2n^2} - (n - 68)\omega, \tag{9}$$

where ω is the microwave frequency.

Table 1. The values of energy of hydrogen atom with principal number from 66 to 70 are calculated by B-spline technique and equation: $E = -1/2n^2$, respectively. The unit of energy is 10^{-4} Hartree.

n	This work	$E = -1/2n^2$
66	–1.147842053440465	-1.14784205693
67	–1.113833812633164	–1.11383381599
68	–1.081314875919307	–1.08131487889
69	-1.050199534794874	-1.05019953791
70	–1.020408160506715	–1.02040816327

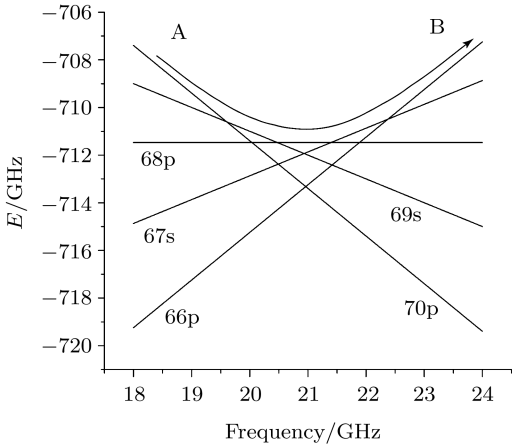


Fig.2. Microwave dressed energy levels of Fig.1 by microwave frequency of constant amplitude microwave field. The states cross at the one-photon resonance because microwave photons are added to or subtracted from the energy of each state. In a microwave pulse chirped from 18 to 24 GHz, there are avoided crossings at these $\triangle n = 1$ resonances, population initially placed in the 70p state, point A, is transferred to the 66p state, point B, via the highest-lying adiabatic curve, as shown by the solid arrow.

Using Eq.(9), we make the centre of Fig.2 at $n = 68$, then the 68p energy has no photons added, 67s has one photon added, 69s has one photon subtracted, etc. What matters for Landau–Zener transitions is only the difference in slopes of energy levels, not the absolute slopes. Using the dressed energy levels given by Eq.(9), we find that the energy levels of states differing in n by 1 would cross at the one-photon resonance, but when the one-photon $\triangle n = 1$ couplings are included the levels have avoided crossings, as shown in Fig.2. Of course, there are higher-order multiphoton resonances, but they are too weak to produce visible avoided crossings in Fig.2 and are always traversed adiabatically.

3. Results and discussion

Tuning the microwave field ($A_0 = 0.1\,\mathrm{V/cm}$) to follow the atom level spacing, we assume that the population residents in the 70p state. Initially, the frequency is 19 GHz and the chirp rate is 0.012 GHz/ns, i.e. $\omega(t) = 19 + 0.012t$, a linear chirp. The obtained population transfer between atom states is illustrated in Fig.3(a). It exhibits that in the case of the red-to-blue sweep, the population is transferred from 70p state to 66p state. In this process, four sequential adiabatic passages are formed by a single frequency-swept laser, so the population from the 70p state traverses 69s, 68p, 67s states then finally comes to 66p state. Figure 3(a) shows that 96% of the population is transferred to the state 66p. During the pulse, the population of each state reveals the fine structure, slightly oscillation in time, which could be regarded as a small Rabi oscillation.[13]

When the initial frequency equals the resonant transition frequency, some novel features come out, and only 45% of population inhabits 66p state (Fig.3(b)). At the beginning of the process, a more bigger oscillation arises between 70p state and 69s state, which can be seen as more stronger Rabi oscillation. Subsequently, about 50% of the population settle down 70p state, and the other states form three sequential adiabatic passage by the frequency-chirped microwave field. At the end of the process, between 70p state and 69s state, obvious population transfer occurs, followed by population transfer among 70p, 69s and 68p states, though the frequency is far away from the resonance frequency. This is a new phenomenon expecting further discussion.

When the chirp rate is 0.02 GHz/ns, we can see that a majority of the population can be transferred to 66p state and trapped there. Every passage is not completely adiabatic because of the bigger chirp rate and interference of states.[14] Figure 3(c) shows the population dynamics of the system. For making the comparison, we plot the evolution of the population of the levels in Fig.3(d) when the chirp rate is 0.007 GHz/ns and other parameters are the same as those in Fig.3(a). In Fig.3(d), the population transfer is much better than that shown in Fig.3(c), about 90% of the population transfer to the target state. But in the process, there is a little bit high oscillation, which is, we think, a little bit high Rabi oscillation induced by the little chirp rate.

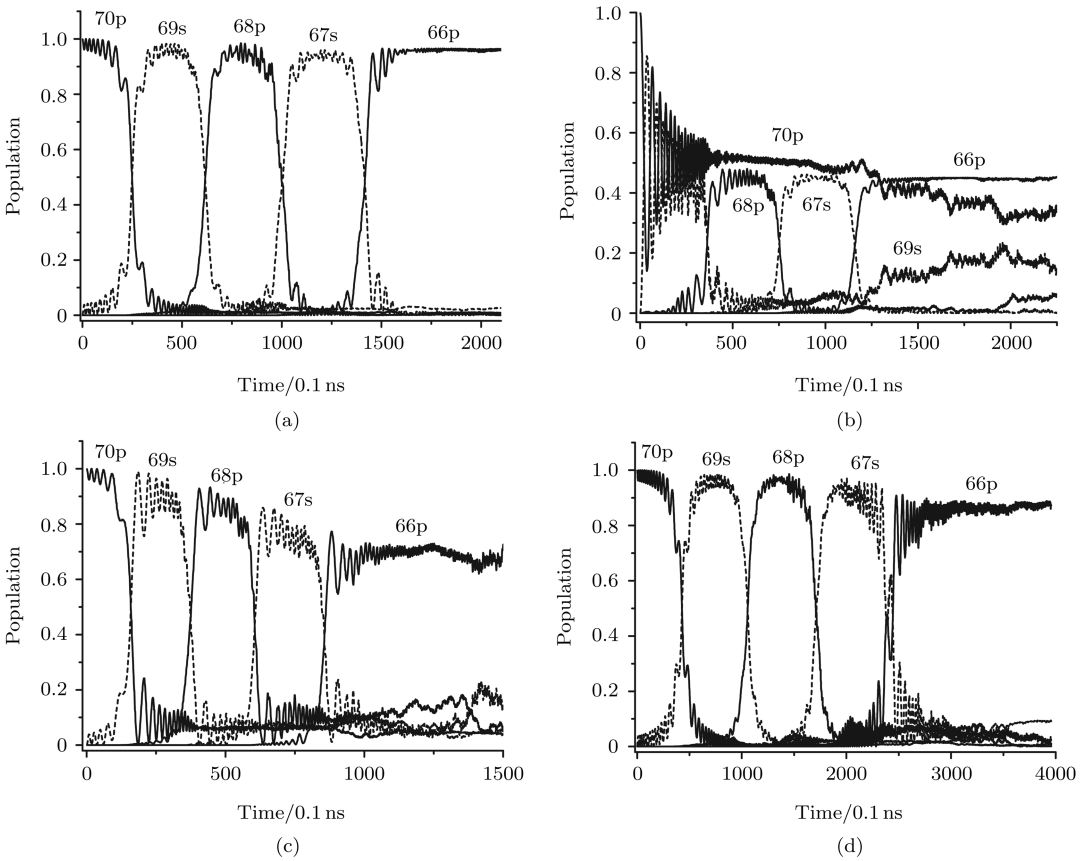


Fig.3. State population dynamics of the system (Fig.1) during the action of frequency-chirped microwave field (0.1 V/cm for constant amplitude) (a) with carrier frequency $\omega_0 = 19\,\mathrm{GHz}$ and chirp rate $\beta = 0.012\,\mathrm{GHz/ns}$, (b) $\omega_0 = 19.6\,\mathrm{GHz}$, (c) $\beta = 0.02\,\mathrm{GHz/ns}$, (d) $\beta = 0.007\mathrm{GHz/ns}$. Other parameters in (b), (c), (d) are the same as those in (a).

In this work, we also consider a situation of counterintuitive frequency swept laser pulse (blue-to-red chirp) with the atom. It is noted that applying a reversed frequency chirped pulse can also achieve a complete population transfer.[3] In our calculation, we adopt $\omega(t) = 23.5 + \beta t$, $\beta = -0.012\,\mathrm{GHz/ns}$ and $A_0 = 0.1\,\mathrm{V/cm}$, and the result of the population transfer is illustrated in Fig.4. 94% of the population, a little lower than that shown in Fig.3(a), arrive at 70p state. There also are small oscillations in the process. We would like to discuss the counterintuitive situation in future work.

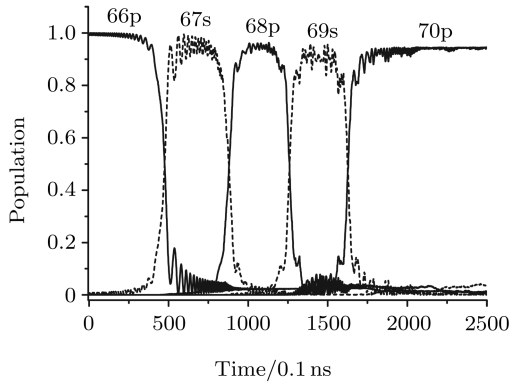


Fig.4. Population transfer in the microwave field of frequency counterintuitive sweep. The atom initially is assumed to be in the 66p state. The parameters are carrier frequency $\omega_0 = 23.5\,\mathrm{GHz}$, chirp rate $\beta = -0.012\,\mathrm{GHz/ns}$ and pulse amplitude 0.1 V/cm.

4. Conclusions

We have proposed a scheme to analyse the population transfer in the Rydberg hydrogen atom with a single frequency-chirped microwave pulse. In our calculation, we consider a nearly realistic system, which has the different transition dipole elements, corresponding to the real atom. The result accords with the experiment very well.[2] The calculation reveals that the complete population transfer in a constant amplitude microwave field is related to the chirp rate β, initial frequency ω_0-the carrier frequency. Recently, Li *et al*[15] studied the population transfer via adiabatic passage between the Stark states of potassium atom by a single frequency-chirped laser pulse. In their analytical study, they used a number of coupled states instead of only three states involved. Because

of the effect of other states, the population transfer exhibited some novel features especially in the counterintuitive sweep process. For the perfectness of our model, we will study the population transfer in real atom with the effect of other states in future work.

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