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Coherent Control of Population Transfer in Li Atoms via Chirped Microwave Pulses *

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We demonstrate theoretically that Li atoms can be transferred to states of a lower principal quantum number by exposing them to a frequency chirped microwave pulse. The population transfer of Li atoms from the $n = 75$ state to $n = 70$ with more than 90% efficiency is achieved by means of the sequential single-photon $\Delta n = -1$ transitions. The calculation fully utilizes all of the available orbital angular momentum l states for a given n, and the interference pattern and population evolution dynamics of individual l states are analyzed in detail. Using the time-dependent multilevel approach, we describe the process reasonably well as a sequence of adiabatic rapid passages.

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Coherent population transfer via adiabatic rapid passage (APR) has become an important aspect of many forms of laser manipulation of atoms and molecules. Examples are using a chirped microwave pulse to change the principal quantum number n of atomic Rydberg states,[1,2] using a chirped infrared-laser pulse to make a sequence of vibrational transitions in a slightly anharmonic diatomic molecule,[3] using half cycle pulses to induce electron-ion recombination into high-lying Rydberg states, and the technique could be a way to produce antihydrogen.[4] In the above examples the frequency sweep of the atomic or molecular motion is required to couple the two concerned states sequentially through many levels, if the frequency is swept through the resonance at a rate smaller than the square of the Rabi frequency, the population transfer from one quantum state to others with approximately 100% efficiency.

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.[5−10] Coherent population transfer via ARP in atoms through two-photon[11] or multiphoton[12] transitions using chirped pulses has been recently demonstrated. By exposing the Li atoms to a chirped microwave pulse, the population via two-photon resonance transferred to the lower Rydberg states successfully,[11] and the same population transfer can also be completed via a single multiphoton adiabatic passage, which requires only a small frequency sweep.[12] In Refs. [11,12], the authors assume that there is one level of each n and the Floquet states are superpositions of n states, but in a real atom there are n levels per n state for $m = 0$, and the Floquet states are superpositions of n and l states of $m = 0$, where l and m are the orbital and azimuthal angular-momentum quantum numbers. Thus each n level is coupled to at least two $n - 1$ levels, which ensures that there must be interferences in a sequence of $\Delta n = -1$ transitions. In this Letter, the basis B-spline basis set combined with model potential method is used to obtain the energy values of Li atoms of each $|n, l\rangle$ state for $m = 0$ and the transition matrix, by using time-dependent multilevel approach, the calculation fully utilizes all of the available orbital angular momentum l for a given n, and the interference pattern in population evolution dynamics of individual l states are analyzed in detail. The calculation result show that the interference in population transfer is distinct, consider the interference between different l states of the same n, the population transfer from $n = 75$ to $n = 70$ of Li atoms can be accomplished by choosing appropriate parameters of the microwave field.

By using the time-dependent multilevel approach (TDML) we developed previously,[13,14] The driving microwave pulse is designed as

$$E(t) = E_0 f(t) \cos[\omega(t)t], \tag{1}$$

to control the population transfer. Here E_0 and $f(t)$ are the strength and shape of microwave field respectively, and $\omega(t)$ is the instantaneous frequency of the microwave field, which has the form of

$$\omega(t) = \omega_0 + \beta \tanh\left(\frac{t - t_0}{\tau}\right), \tag{2}$$

where ω_0 is the time dependent carrier frequency of the microwave pulse; β, τ and t_0 can be used for controlling the frequency sweeping range and the steepness of the chirped function respectively. The shape of the microwave field is slowly ramped as

$$f(t) = \begin{cases} \sin^2 \frac{\pi t}{T}, & 0 \le t \le mT, \\ 1, & t \ge mT, \end{cases} \tag{3}$$

where T and m are the period and the number of the period of the microwave pulse respectively. Thus in the presence of a microwave field $E(t)$ (the direction

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of the field along the z-axis), the Hamiltonian for the outer electron of the Li atom takes the form

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

where H_0 is the field-free Hamiltonian. The stationary Schrödinger equation has the form

$$\Big[-\frac{1}{2}\nabla^2 + V(r)\Big]\phi(r) = E\phi(r), \tag{5}$$

where $V(r)$ is the core potential of an alkali-metal atom, involving the effects of penetration, and polarization is assumed in the form[15]

$$V(r) = -\frac{1}{r}\big\{(Z-1)\exp(c_1 r) + c_2 r\exp(c_3 r) + 1\big\}, \tag{6}$$

for the Li atom, $c_1 = -3.395$, $c_2 = 3.212$, $c_3 = -3.207$, the wave function $\phi(r)$ has the form $\phi_{nlm}(r) = R_{nl}(r)Y_{lm}(\theta,\phi)$, where $Y_{lm}(\theta,\phi)$ is a spherical harmonic function, the radial function $R_{nl}(r)$ is a solution for the reduced Schrödinger equation

$$\Big[-\frac{d^2}{2dr^2} + \frac{l(l+1)}{2r^2} + V(r)\Big]R_{nl}(r) = E_{nl}R_{nl}(r). \tag{7}$$

Using B-spline basis set, $R_{nl}(r)$ can be expanded as

$$R_{nl}(r) = \sum\nolimits_j c_j \frac{r^l B_{j,k}(r)}{r}. \tag{8}$$

where $B_{j,k}(r)$ is the jth B-spline of order k, defined in the region $[0, R_{\max}]$ with $R_{\max} = 40000$ a.u., the convergence behavior can be examined by changing the basis size and $R_{\max}$.

Substituting Eq. (8) into Eq. (7), we obtain the matrix equation

$$HC = ESC, \tag{9}$$

where H is the Hamiltonian matrix and S is the overlap matrix,E and C are eigenvalues and eigenvectors. By solving Eq. (9), we can obtain E and $R_{nl}(r)$. The wavefunction in the presence of microwave field $E(t)$ can be written as

$$\psi(r,t) = \sum_{k=70}^{75}\sum_{l=0}^{5} a_{kl}(t)\phi_{kl}e^{-iE_{kl}t}, \tag{10}$$

where E_{kl} is the field free energy of atomic ground or excited state in field free, the values of E_{kl} are listed in Table 1, the calculated Li energies agree with the quantum defect method.[15] Table 2 presents the population transfer state selected and the values of E_{kl}. $\psi(r,t)$ obeys the time-dependent Schrödinger equation (in atomic mass unit)

$$i\frac{\partial\psi(r,t)}{\partial t} = H\psi(r,t). \tag{11}$$

Substituting Eq. (10) into Eq. (11) and solving the time-dependent Schrödinger equation numerically, we can obtain $a_{kl}(t)$. Then the probability of the outer electron inhabiting the state k can be written as

$$p_k = \sum\nolimits_{l=0}^{l=5} |a_{kl}|^2. \tag{12}$$

Using the above formula, we can calculate the state-to-state transition probability, from which the variation of population of each state can be observed.

Table 1. Comparison of the calculated Li energies with the quantum defect method in units of a.u. Two rows of energies $E(n,l)$ are listed, the upper row refers to our calculated result, and the lower row refers to the result calculated with the quantum defect method.

	$E(n,l)$			
n	$l=0$	$l=1$	$l=2$	$l=3$
2	−0.19732	−0.13007		
	−0.19814	−0.13024		
3	−0.07412	−0.05724	−0.05557	
	−0.07418	−0.05724	−0.05561	
4	−0.03861	−0.03198	−0.03126	−0.03125
	−0.03862	−0.03197	−0.03127	
5	−0.02364	−0.02038	−0.02001	−0.02000
	−0.02364	−0.02037	−0.02001	
6	−0.01595	−0.01411	−0.01389	−0.01388
	−0.01594	−0.01411	−0.01390	

Table 2. Li energy values corresponding to the population transfer states in units of a.u.

	$E(n,l)$		
l/n	70	71	72
0	−0.0001032156578	−0.0001003124285	−0.0000975299906
1	−0.0001021814220	−0.0000993214157	−0.0000965798255
2	−0.0001020437274	−0.0000991894592	−0.0000964532926
3	−0.0001020408492	−0.0000991867008	−0.0000964506475
4	−0.0001020408165	−0.0000991866695	−0.0000964506175
5	−0.0001020408163	−0.0000991866693	−0.0000964506173
l/n	73	74	75
0	−0.0000948617350	−0.0000923014986	−0.0000898435286
1	−0.0000939502032	−0.0000914265337	−0.0000890032005
2	−0.0000938288008	−0.0000913099881	−0.0000888912560
3	−0.0000938262628	−0.0000913075516	−0.0000888889156
4	−0.0000938262340	−0.0000913075239	−0.0000888888891
5	−0.0000938262338	−0.0000913075237	−0.0000888888889

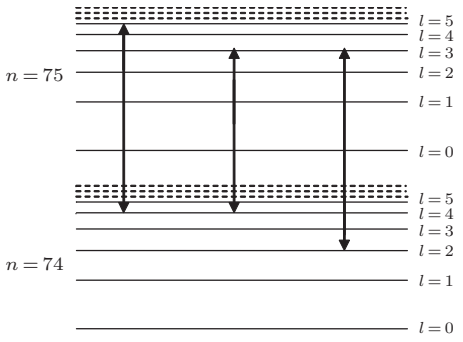

Fig. 1. Scheme of the Li atom energy level.

Firstly we study the population transfer from the $n = 75$ state to $n = 73$, the Li atoms initially populated in the $|75,5\rangle$ state and then placed in a chirped microwave pulse. The scheme of the energy level is shown in Fig. 1. The transition probability of the electron to move from the $|n,l\rangle$ state to $|n-1,l'\rangle$ is related to the strength and the slew rate of the chirped frequency, to coherently control the population transfer,

the field strength as well as the rate of change of the instantaneous frequency must be optimized.

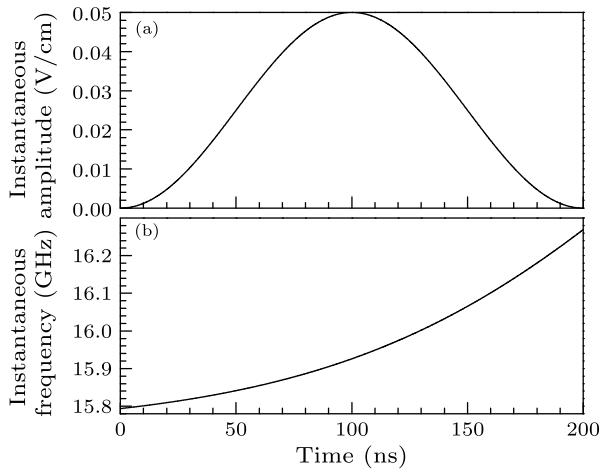

Fig. 2. (a) Instantaneous strength of the microwave pulse. (b) Chirped microwave pulse frequency dependence.

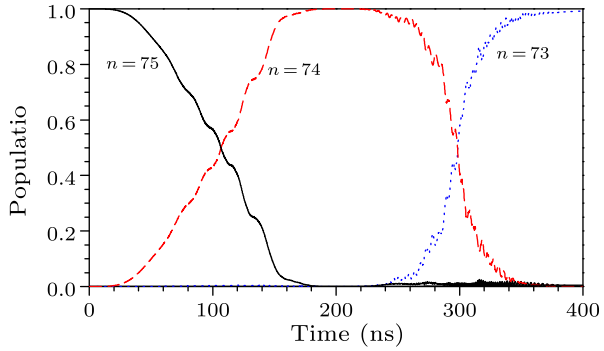

Fig. 3. Population transfer of Li atoms initially located in the principal quantum state $n = 75$ and transferred sequentially to the $n = 73$ state via a chirped microwave pulse. The microwave field strength used is $E_0 = 0.05\,\mathrm{V/cm}$, and the chirped parameters used are $\beta = 7.8\,\mathrm{GHz/ns}$, $t_0 = 250\,\mathrm{ns}$, $\tau = 150\,\mathrm{ns}$.

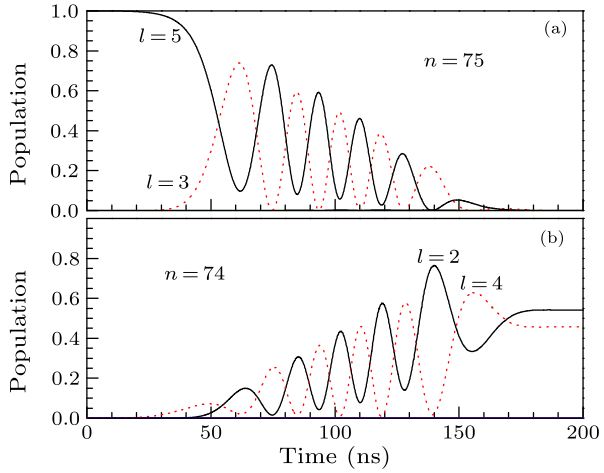

Fig. 4. (a) Depopulation of the initial state $|n = 75, l = 5\rangle$. (b) Initial population transitions to the $|n = 74, l\rangle$. The microwave field strength used is $E_0 = 0.05\,\mathrm{V/cm}$, and $\beta = 7.8\,\mathrm{GHz/ns}$.

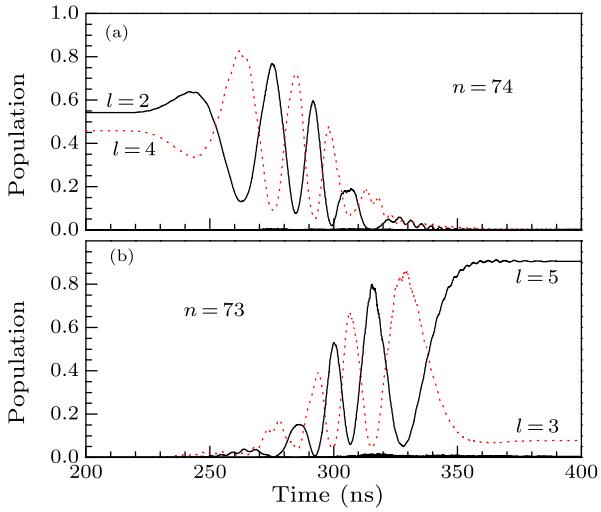

Fig. 5. Dynamical evolution of the most dominant angular momentum l components of the same principal quantum number n, for (a) $n = 74$, the solid line corresponds to $l = 2$, the dotted line corresponds to $l = 4$, for (b) $n = 73$, the solid line corresponds to $l = 5$, the dotted line corresponds to $l = 3$. The microwave field parameters are the same as those in Fig. 4.

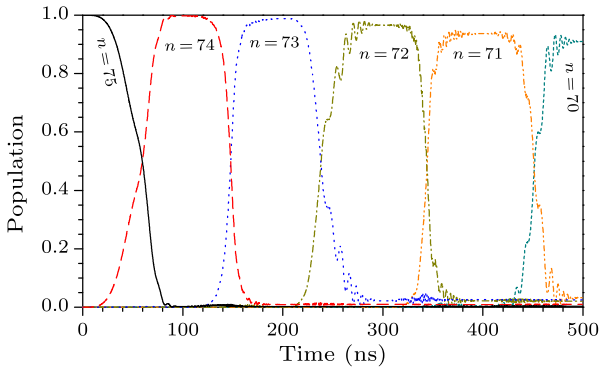

Fig. 6. Population transfer of Li atoms initially located in the principal quantum state $n = 75$ and transferred sequentially to the $n = 70$ state via a chirped microwave pulse.

In our calculation, the parameters of the microwave field are selected to be $E_0 = 0.05\,\mathrm{V/cm}$, $\beta = 7.8\,\mathrm{GHz/ns}$, $\tau = 150\,\mathrm{ns}$, $t_0 = 250\,\mathrm{ns}$, $T = 200\,\mathrm{ns}$, $m = 2$, the time-dependent field strength and frequency are shown in Fig. 2, The instantaneous frequency of the microwave field is positively chirped and tuned to follow the single-photon resonance frequency between the $\Delta n = -1$ transitions. Figure 3 shows the nearly complete population transfer from $n = 75$ to $n = 73$. This calculation result is consistent with the experimental result in Ref. [6], in which the atoms are transferred from $n = 75$ to $n = 66$ with a microwave pulse chirped from 7.8 to 11.8 GHz, the model in Ref. [6] was assumed that there are only one level of each principal quantum number n, so the interference between the $\Delta n = -1$ transitions is missing. In our calculation, the available angle quantum number l of each n state is considered, the interference between l states of the same n state as illustrated in Figs. 4 and 5, due to the electric dipole selection

rules, the single-photon transitions only link states with $\Delta l = \pm 1$, the initial frequency turns nearly to be the resonant frequency between the $|75, 5\rangle$ and $|74, 4\rangle$ states, so the population starts to transfer from $|75, 5\rangle$ to $|74, 4\rangle$, due to the interference between the $|75, 5\rangle$ and $|75, 3\rangle$ states, the population transfers from $|74, 4\rangle$ to $|75, 3\rangle$ synchronously, and then the interference between $|74, 4\rangle$ and $|74, 2\rangle$ states starts to arise and becomes more important. After the microwave pulse, the population is completely transferred to $|74, 4\rangle$ and $|74, 2\rangle$ states as shown in Fig. 4(b). This population transfer course is similar to the two-photon resonance between the $|75, 5\rangle$ and $|75, 3\rangle$ states. As the frequency of the microwave increases, the population transfers from $n = 74$ to $n = 73$ sequentially, the population evolution of each l state is shown in Fig. 5, the atoms finally populate in the $|73, 5\rangle$ and $|73, 3\rangle$ states with probabilities about 90% and 10% respectively. Thus, to realize the complete transfer between sequent n states with single-photon resonance, the interference between different l states of the same n must be considered.

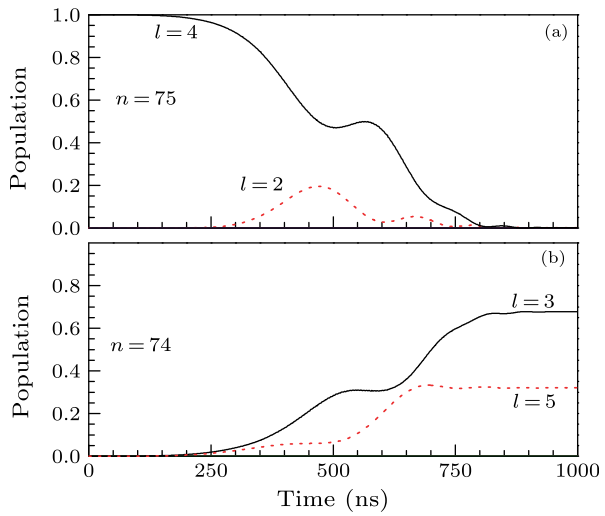

Fig. 7. Population evolution of each l states of $n = 75$ and $n = 74$ states.

Second, we study the population transfer of Li atoms from $n = 75$ to $n = 70$, the atoms initially populated in the $|75, 4\rangle$ states, the parameters of the microwave pulse are $T = 100\,\text{ns}$, $m = 5$ and $\beta = 5.4\,\text{GHz/ns}$, the other parameters are the same as those in Fig. 3. The microwave pulse lasts 500ns, the frequency is chirped from 15.75 GHz to 18.42 GHz, which corresponds to single-photon resonance from $n = 75$ to $n = 70$ sequentially. The population transfer between the selected states is shown in Fig. 6, it exhibits that more than 90% population is transferred to the aim state $n = 70$ by the sequential adiabatic rapid passage, the population transfer between adjacent states last 100 ns, which is due to the periodical change of the microwave field strength and the smooth frequency chirp of the microwave pulse. During the continuous population transfer, there must be interference in the sequential $\Delta n = -1$ transitions. Figure 7 shows the population evolution of each l state for $n = 75$ and $n = 74$ states, oscillations in the population of each l state reflect the interference of the transition probability, while the interference between the l states with the same n is considered, the transition probability from $n = 75$ to $n = 74$ is nearly 100%, otherwise, only about 70%. Thus to coherently control the population transfer of successive states, the microwave pulse should be appropriately designed and the interference in transitions should be considered. The interferences between l states in Figs. 4 and 7 are really different, which is due to different pulse duration periods and frequency-chirped speeds of the microwave pulse, decreasing the period and increasing the frequency-chirped speed can reduce the interference of different l states.

In conclusion, by using the TDML approach, the interaction of frequency-chirped microwave pulse with Li atoms has been studied. The calculation results show that to coherently control the population transfer of successive n states of Rydberg atoms, the parameters of microwave pulse should be appropriately selected, and the interference between l states during the sequent $\Delta n = -1$ transitions must be considered. From our calculation, we find that the interference between different l states for the same n is related to the microwave pulse duration and the frequency-chirped speed, which indicate that the interference can be controlled by choosing different parameters of microwave pulse to enhance the population transition probability. This method appears robust enough to be practical, which would be used to guide the experiment to choose the appropriate parameters of the microwave field to control the population transfer. For example, it is possible to induce the electron-ion recombination into high-lying Rydberg states with chirped microwave pulse to produce antihydrogen if the frequency-chirped in the opposite direction would transfer atoms to a higher n states, leading to ionization at a lower microwave field.

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