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Lithium atom population transfer by population trapping in a chirped microwave pulse*

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Using a time-dependent multilevel approach, we demonstrate that lithium atoms can be transferred to states of lower principle quantum number by exposing them to a frequency chirped microwave pulse. The population transfer from $n = 79$ to $n = 70$ states of lithium atoms with more than 80% efficiency is achieved by means of the sequential two-photon $\Delta n = -1$ transitions. It is shown that the coherent control of the population transfer can be accomplished by the optimization of the chirping parameters and microwave field strength. The calculation results agree well with the experimental ones and novel explanations have been given to understand the experimental results.

Keywords: time-dependent multilevel approach, two-photon transition, frequency chirped microwave pulse

PACC: 3100, 3120, 3280

1. Introduction

Adiabatic rapid passage (ARP) is an approximately 100% efficient way to transfer population from one state to another. Population transfer of atoms from the low-lying initial state to the highly-excited target state becomes practical with a sequence of ARPs.[1,2] The high efficiency of each step allows population to be transferred through many levels by sequential ARPs, which can make population transfer by using a coherent chirped radiation field, unique and quite attractive in atomic physics and chemistry. Two examples are the production of circular Rydberg states and vibrational ladder climbing using chirped laser pulses.[3,4] In addition to the intrinsic interest of this problem it may have practical importance as well. Meerson and Friedlang[5] suggested that using a microwave pulse, initially at the Kepler frequency and chirped to lower frequency, would transfer atoms to a higher n state, leading to ionization at a lower microwave field. Bensky *et al* [6] and Wesdorp *et al* [7] suggested that it might be more interesting to chirp the frequency in the other direction; they demonstrated that it is possible to induce electron–ion recombination into high-lying Rydberg states with half cycle pulses, and the technique could be a way to produce antihydrogen.[8]

Recently the coherent population transfer of Rydberg atoms induced by the frequency- or amplitude-chirped pulses has also been used to carry out quantum control physical and chemical processes by robustly driving transitions between quantum states.[9,10] A resent experiment[11] on highly excited lithium atoms in the microwave field revealed a lot of novel features, in which the valence electron of lithium atom is excited first and then exposed to a microwave pulse, the frequency of the pulse being turned initially to the electron's Kepler frequency, and then chirped to higher frequency. The electron's orbital frequency is increased during the chirp, leading simultaneously to a decreased orbital size and increased binding energy. The experiment concentrated on the transition between $n = 79$ and $n = 70$ states of lithium atoms. In this paper, frequency-chirped microwave pulse is used to realize the two-photon $\Delta n = -1$ transitions of lithium atoms, the calculation results show that the coherent control of the population transfer between $n = 79$ and $n = 70$ states of lithium atoms can be accomplished by the optimization of the chirping parameters with a constant amplitude microwave pulse. The numerical results agree well with Gallagher's experiment.[11] In our calculation, $l = 0$ and $l = 1$ of each n are considered, the calculation results show that two channel calculations ($l = 0, 1$) are the simplest possible way to induce population transfer, and it is the closest one to the one-dimensional calcu-

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lation.

2. Theory and method

By using the TDML method developed by ourselves,[12−14] the driving microwave pulse is designed as

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

to control the population transfer, where E_0 is the amplitude of microwave field, $\omega(t)$ is the time-dependent carrier frequency. So in the presence of a microwave field $E(t)$ (the direction of the field along the z-axis), the Hamiltonian for the outer electron of the lithium atom takes the form

$$H = H_0 + zE(t), \tag{2}$$

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

$$\left[-\frac{1}{2}\nabla^2 + V(r)\right]\phi(r) = E\phi(r), \tag{3}$$

where $V(r)$ is the core potential of an alkali-metal atom, involving the effects of penetration and polarization, and is assumed to be in the form[13]

$$V(r) = -\frac{1}{r}\left\{(Z-1)\exp(c_1 r) + c_2 r\exp(c_3 r) + 1\right\}, \tag{4}$$

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

$$\left[-\frac{\mathrm{d}^2}{2\mathrm{d}r^2} + \frac{l\,(l+1)}{2r^2} + V(r)\right]R_{nl}(r) = E_{nl}R_{nl}(r). \tag{5}$$

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

$$R_{nl}(r) = \sum_j c_j \frac{r^l B_{j,k}(r)}{r}, \tag{6}$$

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

Substituting Eq.(6) into Eq.(5), the following matrix equation can be obtained

$$HC = ESC, \tag{7}$$

where H is the Hamiltonian matrix and S is the overlap matrix, E and C are eigenvalues and eigenvectors. By solving Eq.(7), we can obtain E and $R_{nl}(r)$. The wavefunction of the lithium atom in the presence of microwave field $E(t)$, can be written as

$$\psi(r,t) = \sum_{k=1}^{21} a_{kl}(t)\phi_k \mathrm{e}^{-\mathrm{i}E_{kl}t}, \tag{8}$$

where E_{kl} is the field free energy of atomic state, the values of E_{kl} are shown in Table 1, which agree well with those obtained by the quantum defect method.[15] Table 2 is the population transfer states we selected and the values of E_{kl}. The $\psi(r,t)$ obeys the time-dependent Schrödinger equation (in atomic mass units)

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

The probability of the outer electron inhabiting the state k can be written as

$$p_k = \sum_{l=0}^{l=1} |a_{kl}|^2. \tag{10}$$

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

Table 1. Comparison of the calculated Li energies with those in the quantum defect method (in a.u.), angular momenta $l = 0$ and $l = 1$.

	$E(n,l)$				
l/n	6	7	8	9	10
0 (present)	–0.015945112	–0.011478495	–0.008656277	–0.00676012	–0.005425072
0 (E^{QD})	–0.015946323	–0.011478979	–0.008656488	–0.006760216	–0.005425116
1 (present)	–0.014110542	–0.010344126	–0.007906499	–0.006238936	–0.00504822
1 (E^{QD})	–0.014106892	–0.010341701	–0.007904824	–0.006237737	–0.005047335

Table 2. Lithium energy values corresponding to the population transfer states (in a.u.).

	$E(n,l)$	
n	l=0	l=1
70	–0.00010322	–0.00010218
71	–0.00010031	–0.00009932
72	0.00009753	–0.00009658
73	–0.00009486	–0.00009395
74	–0.00009230	–0.00009143
75	–0.00008984	–0.00008900
76	–0.00008748	–0.00008667
77	–0.00008521	–0.00008444
78	–0.00008303	–0.00008228
79	–0.00008093	–0.00008021

3. Result and discussion

The study of the use of a chirped microwave pulse to change the principal quantum number n of Rydberg atomic states is a subject of much current experimental interest.[11,14] In this Section, we discuss the chirped microwave pulse how to move population of lithium atoms from higher ($n = 79$) to lower ($n = 70$) Rydberg state. From Table 2, we see that: np states are located nearly halfway between ns and $(n+1)s$ states, the two-photon $\Delta n = -1$ transitions are in close resonance with the one-photon transitions, namely, between the np to ns and ns to $(n-1)p$ transitions; the scheme of the energy level is shown in Fig.1. In Ref.[11], the important simplification is that it assumes there is a single level of each n, and points out that the electrons move in Kepler orbits, with binding energies given by Ry/n^2 (where Ry is the Rydberg constant, n is the principal quantum number), with the energy spacing between adjacent levels being $2\mathrm{Ry}/n^3$; the electrons have a Kepler frequency of $f_k = (2\mathrm{Ry}/h)/n^3$, where $2\mathrm{Ry}/h = 6.6 \times 10^{15}$ GHz. For $n = 70, 71, \ldots, 79$, $f_k = 19.2, 18.4, \ldots, 13.4$ GHz respectively, in the microwave region. In our calculation, the frequency dependence of the microwave has the form: $\omega(t) = \omega_0 \exp(\beta t)$ (as shown in Fig.2), which is positively chirped and tuned to follow the spacing between the $\Delta n = -1$ resonances. The initial frequency is located near Kepler frequency of $n = 79$, the value of which is 13.4 GHz, and the frequency chirp stops right after reaching the $n = 71$ to 70 resonance. The frequency sweeping range is from 13.4 to 16.5 GHz, the population transfer stops at the state $n = 70$. In Fig.3(b), we choose $E_0 = 0.05$ V/cm, $\omega_0 = 13.4$ GHz, $\beta = 0.0002$ GHz/ns, a clear evidence for population transfer is observed, about 60% $n = 79$ atoms are transferred to $n = 70$, in which the $79p$ states were initially populated. To achieve more population transfer from initial state to final state, the ratio of the instantaneous frequency to the time span must be small enough to allow atoms to completely pass over the avoided crossing at the corresponding two-photon resonance frequency, in Fig.3(a), we have changed $\beta = 0.000195$ GHz/ns, more than 80% population transferred to the final state. This result agrees well with the experimental result of Gallagher,[11] in which lithium atoms are excited by three 5-ns pulses of a dye laser via the sequence $2s \to 2p \to 3s \to np$ state of $n = 79$. The atoms are then exposed to a 500-ns microwave pulse chirped from 13 GHz to 19 GHz, the initial frequency of the microwave pulse matches the Kepler frequency of the $n = 79$ state, which equals 13 GHz, and the value of microwave field amplitude E_0 ranges from 0.01 V/cm to 7.1 V/cm, the chirped microwave field phase-locks the electron's motion of an atom in $n = 79$ and increases its orbital frequency from 13 GHz to 19 GHz, decreasing n from 79 to 70. Finally more than 80% of the $n = 79$ atoms are transferred to $n = 70$, the final-state distribution is inferred by the time-resolved field ionization signal.

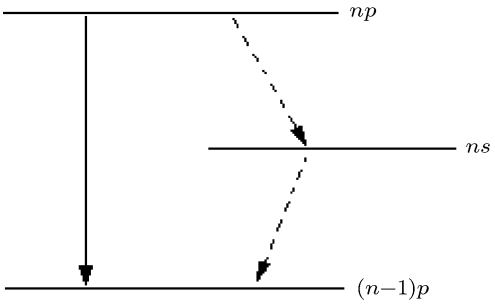


Fig.1. The schematic of the lithium-atom energy level. Solid and dashed arrow lines represent the two-photon and one-photon transition respectively.

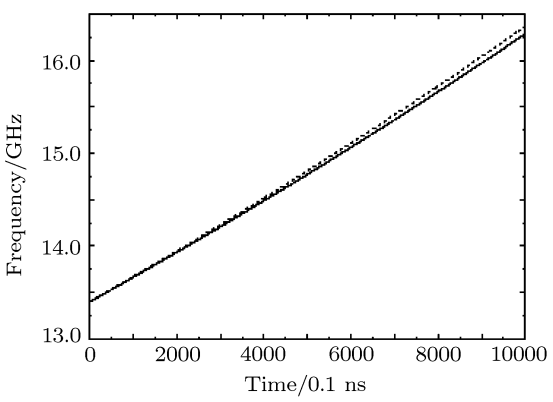


Fig.2. The chirped microwave pulse frequency versus time, the dotted line $\beta = 0.0002$ GHz/ns, the solid line $\beta = 0.000195$ GHz/ns.

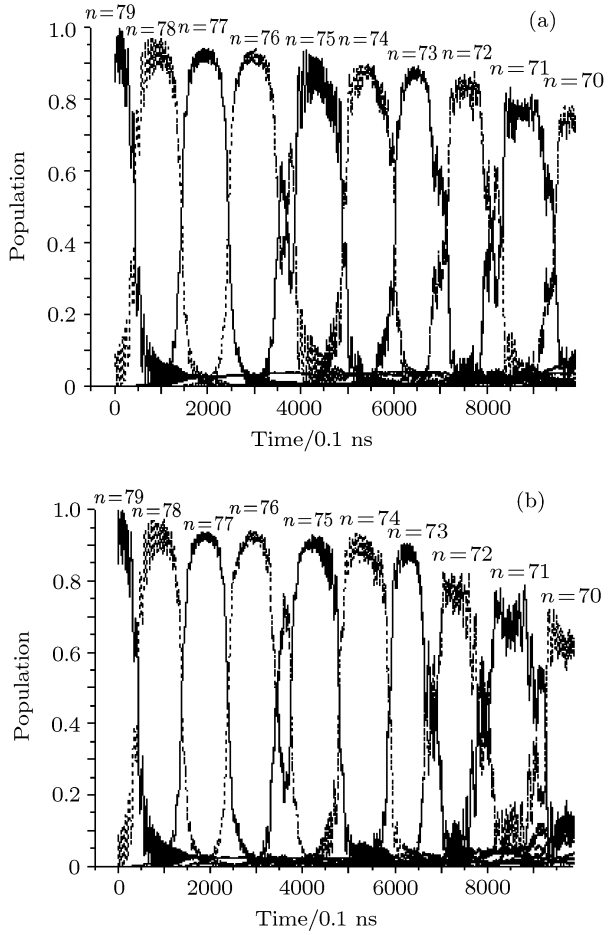


Fig.3. Population transition probability of lithium atoms initially located in the principal quantum state $n = 79$ and driven sequentially to the $n = 70$ state via a chirped microwave pulse. The microwave field strength used is $E_0 = 0.05$ V/cm, and the chirped parameters used are (a): $\beta = 0.000195$ GHz/ns, for (b): $\beta = 0.0002$ GHz/ns.

From Fig.3, we know that the transition probability of the electron to move from the initial state to the final state is related to the slew rate of the chirped frequency, and the ratio of the instantaneous frequency affects the population of the final state efficiently. We define the slew rate of our pulses by $\beta = \Delta\omega/\Delta t$ (GHz/ns). The microwave instantaneous frequency is chirped over a frequency interval from 13.4 GHz to 16.5 GHz. Such a frequency sweeping corresponds to the two-photon resonance transition between $n = 79$ and 70. In Fig.4, we show the effects of frequency-chirped rate on the population transfer from $n = 79$ to 70 state, most of the population take place in the case of $\beta = 0.000195$ GHz/ns. With other values of β, no more population transfer can take place even in the case of large slew rates. Hence, to coherently control the population transfer, the rate of the change of the instantaneous frequency of a constant amplitude pulse must be optimized. In our calculation, complete population did not take place, we attribute this to our ignoring the interference occurring in successive $\Delta n = -1$ transitions, while we assume that there are two levels of each n, whereas in reality there are n levels of $m = 0$, so extending our approach to take all the interferences into account is an interesting and practical work.

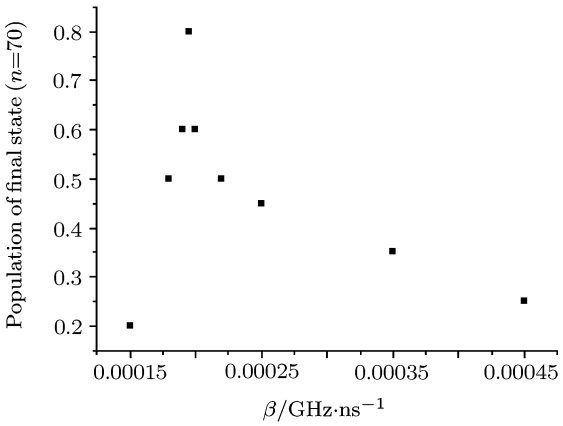


Fig.4. Population of the final state ($n = 70$) of Li atom as a function of the slew rate β for the constant amplitude microwave pulse ($E_0 = 0.05$ V/cm).

4. Conclusion

In conclusion, by exposing lithium atoms to a chirped microwave pulse, the population is transferred to a lower state efficiently with the two-photon resonance. We found that more than 80% of population can be transferred from $n = 79$ to $n = 70$ state, the population transfer can be optimized by tailing the variation of frequency with a constant amplitude microwave pulse. The calculation results agree well with experimental ones. This method appears robust enough to be in practice. Chirping the frequency up can move unstable population[16] to lower, more stable states, which could be quite useful for transporting recombined antihydrogen to lower lying states, and this approach would constitute a useful method of labeling Rydberg states for molecular spectroscopy.

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