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Phase shift of the population oscillation and population trapping*

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Using our recently developed time-dependent multilevel approach, we have calculated the phase shift of the population oscillation of the excited potassium atom in frequency-modulated fields when the modulation amplitude is suddenly changed. The numerical results are in good agreement with the measurements. A new way of realizing population trapping is suggested.

Keywords: time-dependent multilevel approach, phase shift
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The phase information about a quantum system is always a hot topic. Nevertheless, in most cases of the studies on the phase in the process of the interaction of the atom with external fields, the emphasis is placed more on the external fields than on the population oscillations. The phase of the external fields is essentially the classical one, while the effects of the quantum phase information disappear in the process of dynamical evolution in most cases. For example, the Berry phase has been neglected for quite a long time because it is hidden in the dynamical phase, since the development of the quantum mechanics. Fortunately, in a recent experiment, Noel *et al* have observed the phase shift of the population oscillations.[1] To our knowledge, the theoretical study of it has not been reported to date. Although little attention has been paid to this effect, we wish to exploit this effect and hopefully develop a new way of achieving the population trapping in a resonant field. The population trapping, especially the coherent population trapping, has received a lot of attention either in theories or in experiments.[2–6] The major reason is that the coherent population trapping is widely used in many aspects of atomic and optical physics. For instance, in the process of the laser cooling of atoms, the fundamental temperature limit associated with the single-photon recoil energy was first overcome by the velocity-selective coherent population trapping.[7] In atom optics and interferometry, the adiabatic transfer of population and momentum based on coherent population trapping has been successfully used for coherent manipulation of an atomic beam.[8]

In this paper, we will report for the first time the calculation of the phase shift of the population oscillations of Rydberg K atoms exposed to a microwave field and a radio-frequency (rf) field using our recently developed time-dependent multilevel approach.[9] We have found some rather surprising results which enable us to suggest a new way of realizing population trapping.

The main content of the time-dependent multilevel approach[9] we presented recently is as follows. In the presence of a static electric field $F_{\rm s}$ (the direction of the field is along the z-axis), a microwave field $F_{\rm mw}\cos\omega_{\rm mw}t$, and an rf field $F_{\rm rf}\cos\omega_{\rm rf}t$, the Hamiltonian for the outer electron of the potassium atom takes the form

$$H = H_0 + H_{\rm s} + zF_{\rm mw}\cos\omega_{\rm mw}t + zF_{\rm rf}\cos\omega_{\rm rf}t, \quad (1)$$

where H_0 is the field-free Hamiltonian and $H_{\rm s} = F_{\rm s}z$. $F_{\rm mw}$ and $F_{\rm rf}$ are the amplitudes of the microwave field and the rf field, respectively.

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The wavefunction $\psi(t)$ of the potassium atom in the three fields obeys the time-dependent Schrödinger equation (in atomic units)

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

The Hamiltonian for the outer electron of an alkali-metal atom in the absence of external fields can be written as

$$H_0 = -\frac{1}{2}\nabla^2 + V(r). \tag{3}$$

The core potential of an alkali-metal atom involving the effects of penetration and polarization is assumed to be in the form[10]

$$V(r) = -\frac{1}{r}\left(1 + \frac{\beta'}{(r+\gamma')^2} + \frac{\kappa'}{(r+\xi')^2}\right). \tag{4}$$

Using this potential model, the eigenfunction Φ_i of H_0 can be obtained in the form of $\Phi_i = R_{nl}(r)Y_{lm}(\theta,\phi)$, where $Y_{lm}(\theta,\phi)$ is a spherical harmonic function and $R_{nl}(r)$ has the form

$$R_{nl}(r) = N\exp(-\rho/2)\rho^s(\rho+\gamma)^t(\rho+\xi)^\mu \sum_{\nu=0}^{n-l-1} a_\nu \rho^\nu, \tag{5}$$

in which $\rho = \alpha r$, $\gamma = \alpha\gamma'$, $\xi = \alpha\xi'$ and $\alpha = 2/(n-\mu_{nl})$, where $s, t, \mu, a_\nu, \beta', \gamma', \xi', \kappa'$, which all depend on n and l, are the parameters to be determined, and μ_{nl} is a quantum defect value to be input. Substituting $R_{nl}(r)$ and $V(r)$ into the radial Schrödinger equation, and considering the standard conditions for a radial wavefunction, we acquire the nonlinear algebraic equations for the above parameters. Solving them, we acquire s, t, μ, a_ν, β', γ', ξ', κ', and then $R_{nl}(r)$ and $V(r)$ in the analytical form. See Ref.[10] for details.

In the ideal consideration, the basis set should run from $n = 0$ to $n = \infty$, which, however, can hardly be realized. In order to significantly save the computing with little accuracy lost, we choose 78 field-free wavefunctions of potassium atom in the vicinity of 21s and 19f states as a basis set. See Ref.[9] for details.

Using the above potential model and the 78 field-free wavefunctions $\Phi_i = |nlm\rangle$ as a basis set, the matrix elements of the Hamiltonian $H_1 = H_0 + H_\mathrm{s}$ have the following form

$$H_{1nlm,n'l'm} = -\frac{\delta_{nlm,n'l'm}}{2(n-\mu_{nl})^2} + F_\mathrm{s}\langle nlm|\, z\,|n'l'm\rangle. \tag{6}$$

By diagonalizing the matrix of H_1, we can obtain the eigenvalue E_k and the eigenvector $\psi_k^{(s)}$ of H_1. They obey the following equation:

$$H_1\psi_k^{(s)} = E_k\psi_k^{(s)}, \tag{7}$$

where

$$\psi_k^{(s)} = \sum_i C_{ki}\,\Phi_i. \tag{8}$$

Using $\psi_k^{(s)}$ as the new basis set, the wavefunctions of the potassium atom in the presence of static electric field, together with the microwave field and the rf field, can be written as

$$\psi(t) = \sum_{k=1} a_k(t)\psi_k^{(s)}\mathrm{e}^{-\mathrm{i}E_k t}, \tag{9}$$

where $a_k(t)$ is the coefficient of the expansion, which represents the amplitude of the transition probability. $\mathrm{e}^{-\mathrm{i}E_k t}$ contains the phase information that determines the moment when resonance occurs. Substituting Eq.(9) into Eq.(2), 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 can be written as

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

Using the above formula, we can calculate the state-to-state transition probability, from which the phase shift of the Rabi oscillation of the transition probability and the population trapping can be obtained.

To our knowledge, the phase shift of the population oscillations was first observed by Noel *et al* experimentally.[1] The experiment concentrated on the transition between the two Stark states which were adiabatically connected to the 21s and 19f zero-field states of Rydberg K atom, and we will label the states 21s and 19f. They prepared the potassium atom into the 21s state in the static electric field, and then exposed the Rydberg K atom to the 9GHz microwave field and the rf field simultaneously. The parameters they used were as follows. The microwave amplitude was set to yield a microwave Rabi frequency of Ω_mw1=156MHz, the rf frequency was set to ω_rf=330MHz. Next they measured Rabi oscillations for various rf field amplitudes, and compared the measured rf multiphoton Rabi frequencies with the theoretically predicted results which were given by[1]

$$\Omega_{\mathrm{rf}_0} = \Omega_\mathrm{mw1}J_0(kF_\mathrm{rf}/\omega_\mathrm{rf}), \tag{11}$$

where J_0 is a zero-order Bessel function, F_rf is the rf field amplitude. The Bessel function oscillates in sign, implying that the rf Rabi frequency is likely to

be negative for some rf amplitude. They plotted in Fig.1 some of their observed Rabi frequencies as being negative along with the theoretical results calculated from Bessel function. The negative Rabi frequency was explained as a 180° phase shift of the population oscillations observed by Noel *et al.*[1] However, Eq.(11), which was deduced by the two-state approximation, can only give the correct results of the Rabi frequency but not the population evolution. The two-state approximation (given in Ref.[1] as Eq.(1)) cannot give the correct results of the population evolution.[9] In Figs.2 and 3, the lower curves are the experimental results with the amplitude of rf modulating field changing from the point labelled with a to point b and point c in Fig.1 at the 3/4 Rabi period,

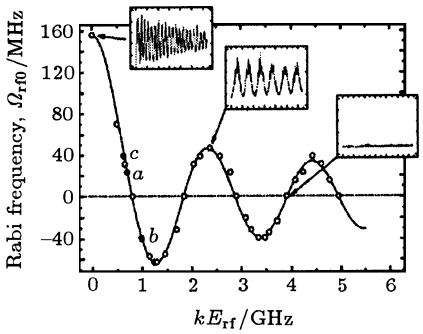


Fig.1. Zero-order Bessel function changes in sign with regard to the rf amplitude imply that for some rf amplitudes the Rabi oscillations will be negative, which leads to the phase shift of the population evolution. The points labelled a, b and c are different modulation amplitudes which are also used in Figs.2 and 3. (This figure is reproduced from Ref.[1]).

respectively. However, their experiment did not give the exact data of the F_{rf}, and they just plotted the approximate position of the modulation amplitudes labelled with a, b and c in Fig.1. In our calculation, in the case of the microwave amplitude $F_{m\omega}$=17.45V/cm and frequency $\omega_{m\omega}$=9GHz, we find that single photon resonance of the microwave takes place at the position of the static field F_s=287.15V/cm and the microwave Rabi frequency $\Omega_{m\omega_1}$=156MHz. Then we add the rf field with the rf frequency ω_{rf}=330MHz and the amplitudes F_{rf} being 1.28, 1.80 and 1.13V/cm at points a, b and c, respectively. The upper curve in Fig.2 shows the present calculations of the transition probability which are obtained by suddenly switching the rf amplitude value 1.28V/cm to the value 1.80V/cm when the evolution time reaches 3/4 Rabi period. In the same way, we have obtained the results as shown by the upper curve in Fig.3 by suddenly switching the rf amplitude value 1.28V/cm to the value 1.13V/cm. It is obvious that the present calculations are in excellent agreement with the experimental results. When we change the modulation amplitude we see not only the Rabi period change, but also a 180° phase shift, which depends on whether the sign changes or not in Fig.1. More specifically, if the Rabi frequency changes sign as shown in Fig.2, the populations of the upper state increase rather than continue to decrease; otherwise, if no sign changes as indicated in Fig.3, the populations continue to decrease.

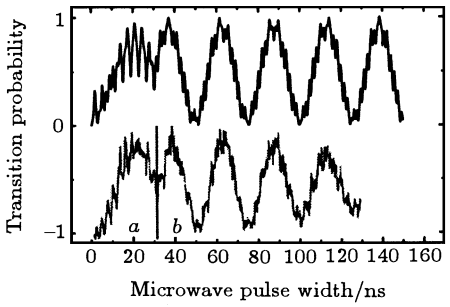


Fig.2. Phase inversion of the population oscillations at the time labelled as a vertical line at which the rf amplitude is suddenly switched from 1.28V/cm to 1.80V/cm. The upper curve is our calculation and the lower is the experimental measurements. The line labelled with ab corresponding to the positions a and b in Fig.1 indicates there are sign changes of Bessel function in Eq.(11).

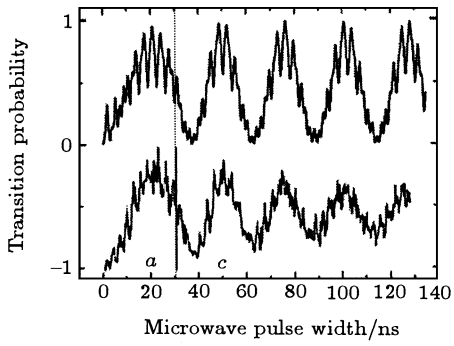


Fig.3. There is no phase inversion of the population evolution with the rf amplitude suddenly switched from 1.28V/cm to 1.13V/cm at the same time as in Fig.2. The line labelled with ac corresponding to positions a and c in Fig.1 indicates there are no sign changes of Bessel function in Eq.(11).

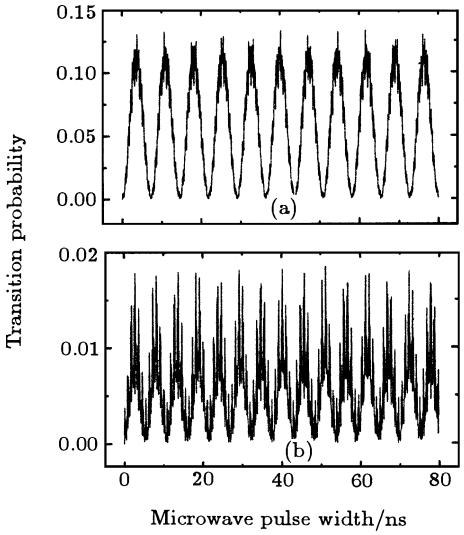


Fig.4. Weak Rabi oscillations derived from the dynamic Stark shift at the position where the total fields greatly exceed the anti-crossing between 21s and 19f states. The upper curve and the lower curve are obtained using microwave with amplitudes of 50V/cm and 60V/cm respectively.

To explore this effect further, we study the phase shift of the Rabi population oscillations in the absence of modulation. We keep the same frequency of the microwave field and the static field value as the values used previously but only change the microwave amplitude. The present calculations of the excited-state population for $F_{\rm mw}$ equalling 50V/cm and 60V/cm are shown by upper and lower curves in Fig.4 respectively. In the case of 60V/cm except for the difference in the sign, the numerical values of the oscillation frequency in Fig.4 are in agreement with those obtained from the equation in Ref.[1]

$$\Omega_{\rm mw1} = \Omega_{\rm static} J_1(kF_{\rm mw}/\omega_{\rm mw}), \qquad (12)$$

where $\Omega_{\rm mw_1}$ is the microwave one-photon Rabi frequency; $\Omega_{\rm static}$ is the size of the static field to avoid crossing; J_1 is first-order Bessel function. A special point to notice is that the sign of the first-order Bessel function in Eq.(12) also changes when $F_{\rm mw}$ is switched from 50V/cm to 60V/cm. Can we see a 180° phase shift when suddenly switching the value of the $F_{\rm mw}$ from 50V/cm to 60V/cm at a proper time? The answer is positive and the results are shown in Fig.5. From the figure, we observe not only the Rabi period change, but also a 180° phase shift at the time labelled *ab*, which results in the population of 19f state increasing rather than continuing to decrease as indicated by the upper curve in Fig.4. What is more interesting is that the population of the 19f state increases again when the microwave amplitude $F_{\rm mw}$ changes back from 60V/cm to 50V/cm at the time labelled with *cd*. The population evolution with the microwave amplitude changed twice is shown by the lower curve in Fig.5.

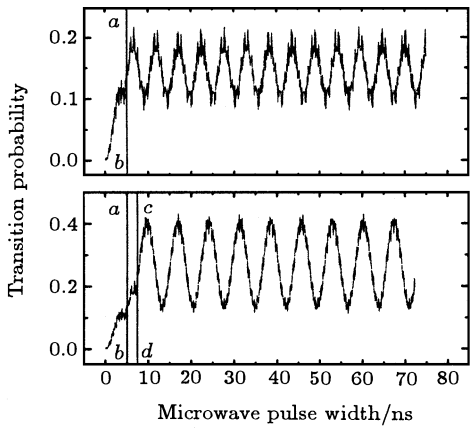


Fig.5. The upper curve shows the change of the microwave amplitude from 50V/cm to 60V/cm corresponding to the sign inversion in Eq.(12), which leads to a phase inversion in the process of the population evolution between 21s and 19f states. We find not only the inversion of the phase but also a magnified amplitude of the transition probability (compare with that in Fig.4). The lower curve shows inversion of the phase happening twice. There are two vertical lines in the lower figure. The left one shows the time at which the rf amplitude is suddenly switched from 50V/cm to 60V/cm and the other one is for the rf amplitude changing from 60V/cm to 50V/cm.

What shall we find if we reiterate this process? The present results of the population evolution of the 19f state with phase inversion repeating seven times are shown in Fig.6. It is surprising that the population is almost all trapped in the 19f state except for a little fluctuation. Similarly, we can also trap the population in the 21s state through different arrangements of the microwave amplitude changed in a certain time sequence. This is very exciting in many aspects, because the population trapping is widely used in various branches of atomic and optical physics. Our results suggest a new way to realize the population trapping. The most important thing in realizing the complete population inversion and the population trapping with this method experimentally is to switch the $F_{\rm mw}$ on and off in a proper time sequence.

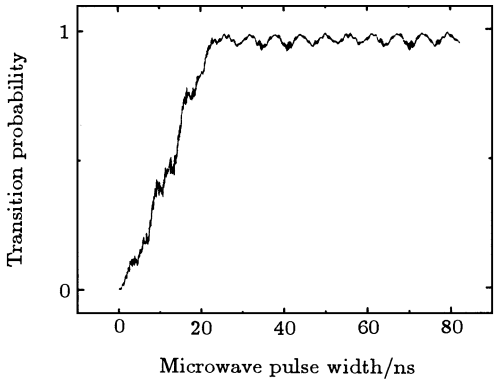


Fig.6. The population oscillations undergo phase inversion seven times and eventually reach the population trapping.

In conclusion, we have used for the first time our recently developed time-dependent multilevel approach to calculate the phase shift of population oscillation of the excited potassium atom in frequency-modulated fields when the modulation amplitude is suddenly changed. The present results are in good agreement with the measurements.

We have also obtained the results of the complete population trapping using the phase-shift method in the absence of the modulation. Our results suggest a new way to realize the population trapping, which is probably useful in the control of a quantum state at our discretion. Further experiments are hoped to be performed to validate the present results.

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