Scheme for multistep resonance photoionization of atoms

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Traditional schemes for multistep resonance photoionization of atoms let every employed laser beam interact with the atoms simultaneously. In such a situation, analyses via time-dependent *Schrödinger* equation show that high ionization probability requires all the laser beams must be intense enough. In order to decrease laser intensity, we proposed a scheme that the laser beam used to pump the excited atoms (in a higher bound state) into an autoionization state does not interact with the atoms until all the population is transferred by the other lasers from a ground state to the bound state. As an interesting example, we examined three-step photoionization of ^{235}U with our scheme, showing that the intensity of two laser beams can be lowered by two orders of magnitude without losing high ionization probability.

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I. INTRODUCTION

Selectively ionizing atoms (or molecules) can be easily performed by means of a multistep resonance photoionization technique, which has been widely used in the fields of isotope separation, single atom (molecule) detection, and chemical dynamics studies, etc. Generally, one or two laser beams are employed to excite the atoms from a ground state into a higher bound state, and another laser to pump the atoms from the bound state into an ionization (continuum or autoionization) state. In traditional schemes, as well known, all the laser beams are set to interact with the atoms simultaneously and high laser power is required for obtaining high ionization probability. However, the photoionization efficiency is low in most cases, leading to heavy waste of light power. Accordingly, raising the photoionization efficiency and lowering the laser power are highly desirable in the application of the photoionization technique, especially in the engineering of laser isotope separation.

The difficulty of raising multiphoton ionization efficiency can be understood theoretically by examining population dynamics of a multilevel system coupled with laser fields. It was found that the population variation is strongly dependent on the values of Rabi frequencies and detuning [1]. In some cases, when these parameters are badly matched, almost all of the population stays trapped in the ground state and the system behaves as if no laser fields interact with it. In fact, as discussed in Sec. II of the present paper, for a multistep photoionization system, where the highest state is an autoionization state, all the laser beams must be intense enough to ensure high ionization probability even though the parameters (Rabi frequencies and detunings) are best matched. Although the line shape and the lifetime of an autoionization state may be manipulated [2], we must pay a price of employed two additional lasers with high power.

The technique of a π pulse or a Cook-Shore pulse is of interest [3]. For a two-level system coupled resonantly with a single mode laser, the excitation probability is $P(t)=1/2[1-\cos A(t)]$. When the pulse area, $A(t)=\int_{-\infty}^{t}\Omega(\tau)d\tau$, is equal to $(2n+1)\pi$ (multiple π pulse), all the population in the lower level transfers to the upper state. For a multilevel system, one might expect that a series of N pulses, each having the area of π, should completely transfer the population from a ground state to a Nth excited state. However, it is very difficult to determine the Rabi frequency $\Omega(t)$, because transition dipole moments are usually not well known. This problem seriously hinders the operation of π pulse technique in practice.

Recently, a technique of Raman chirped adiabatic passage was developed for dissociation of molecules [4,5]. For a two-level system, the population of a lower state can be completely pumped to the higher level by an intense light pulse with a chirped frequency, which sweeps across the resonance center during the interaction. Although this technique has been well-demonstrated experimentally [6,7], it can hardly be used for isotope separation because the chirped frequency covers a too large range, about $\pm 3\%$ around the resonance frequency.

In the present paper, we analyzed the processes of two- and three-step photoionization via the traditional scheme, i.e., all the laser fields simultaneously interacting with the atoms. It was shown that mainly because the decay rate of the autoionization state is very fast, efficient ionization requires all the laser fields must be intense enough. Based on the technique of population transfer [3,8–11], a photoionization scheme was proposed. The advantage of our new scheme was well demonstrated by numerical calculations of three-step resonance photoionization of ^{235}U and other systems.

II. ANALYSIS OF THE TRADITIONAL SCHEME

A. Two-step photoionization

Two-step resonance photoionization is diagrammed in Fig. 1. Laser L_1 couples a ground state $|1\rangle$ and a bound state $|2\rangle$ with a detuning of Δ. Simultaneously, laser L_2 couples the state $|2\rangle$ and an autoionization state $|3\rangle$. The ionizing process causes the state $|3\rangle$ to decay with a very fast rate ($\geqslant 10^{12}\,s^{-1}$), denoted by γ, whereas the decay of the state

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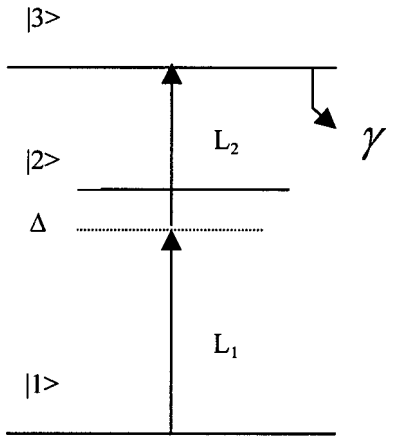

FIG. 1. Energy-level diagram of a three-level system. Laser L_1 couples an initial populated state $|1\rangle$ with a bound state $|2\rangle$, and laser L_2 couples the state $|2\rangle$ with an autoionization state $|3\rangle$. Δ is the frequency detuning, and γ the decay rate of the state $|3\rangle$.

$|2\rangle$, caused mainly by spontaneous emissions, is slower by four or five orders of magnitude, and neglected here.

Within the RWA approximation, the equation about the probability amplitudes C_1, C_2, C_3 of the states $|1\rangle$, $|2\rangle$, and $|3\rangle$ reads

$$\begin{cases} i\dot{C}_1 = \frac{1}{2}\Omega_1 C_2, \\ i\dot{C}_2 = \frac{1}{2}\Omega_1 C_1 + \Delta C_2 + \frac{1}{2}\Omega_2 C_3, \\ i\dot{C}_3 = \frac{1}{2}\Omega_2 C_2 - \frac{1}{2} i\gamma C_3, \end{cases} \tag{1}$$

where Ω_1 is the Rabi frequency of L_1 coupled to the state $|1\rangle$ and the state $|2\rangle$, and Ω_2 is the Rabi frequency of L_2 coupled to the state $|2\rangle$ and the state $|3\rangle$. From the third equation of Eqs. (1) and the initial condition $C_3(0)=0$,

$$C_3 = -\frac{i}{2}\Omega_2 e^{-\gamma t/2}\int_0^t C_2(\tau) e^{\gamma\tau/2}\, d\tau \tag{2}$$

From Eq. (2), we have

$$|C_3(t)|^2 < \left(\frac{\Omega_2}{\gamma}\right)^2 \tag{3}$$

Obviously, if $\Omega_2 \ll \gamma$, then $|C_3(t)|^2 \sim 0$. In this situation, the population oscillates between states $|1\rangle$ and $|2\rangle$, and the probability of photoionization should be very low. Accordingly, one necessary condition for efficient photoionization is that the intensity of L_2 should be strong enough to ensure

$$\Omega_2 \sim \gamma. \tag{4}$$

With this condition, we obtain from Eqs. (1)

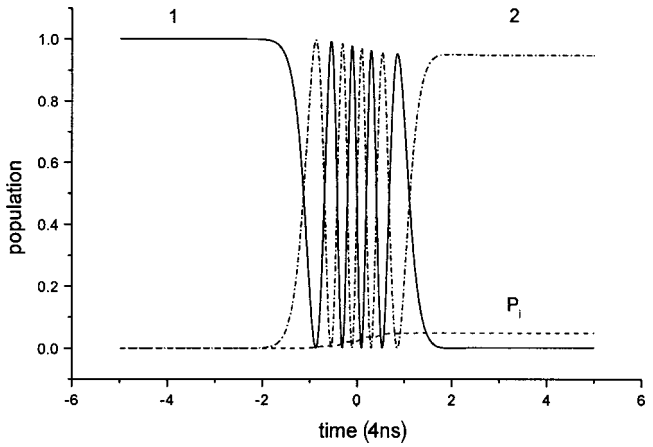

FIG. 2. Population evolution of the three-level system shown in Fig. 1. When the conditions (7) are not satisfied ($\Omega_1^{(0)} \sim \Omega_2^{(0)} \ll \gamma$), most populations oscillates between the states $|1\rangle$ and $|2\rangle$, denoted by curves 1 and 2, respectively, and the final ionization probability (curve P_i) is very low.

$$\begin{cases} R_1 = -\frac{i}{2}\frac{\Omega_1}{\Omega_2} C_1 - i\frac{\Delta}{\Omega_2} C_2 - \frac{i}{2} C_3, \\ R_2 \approx -\frac{i}{2} C_2 - \frac{1}{2} C_3, \end{cases} \tag{5}$$

where $R_1 = \dot{C}_2/\Omega_2, R_2 = \dot{C}_3/\Omega_2$. Before the interaction of laser pulses $C_2(0) = C_3(0) = 0$. Thus, the contribution to R_1, R_2 results directly from $C_1(0)$. Obviously, if $\Omega_1 \ll \Omega_2$, then R_1, R_2 is kept small all the times, causing the population of the states $|2\rangle$ *and* $|3\rangle$ to close to zero. So, another necessary condition for efficient photoionization is

$$\Omega_1 \sim \Omega_2. \tag{6}$$

From conditions (4) and (6), the necessary conditions for efficient photoionization should be

$$\Omega_1 \sim \Omega_2 \sim \gamma. \tag{7}$$

In order to test these conditions, we carried out the following numerical calculations with $\gamma = 8\times 10^{11}\,\mathrm{s}^{-1}$ and $\Delta = 0$. Laser L_1 and L_2 are chosen to be single mode Gauss pulsed lasers, and the corresponding Rabi frequencies can be expressed as

$$\Omega_i(t) = \Omega_i^{(0)} e^{-t^2/\tau^2}, \quad (i = 1,2), \tag{8}$$

where τ is the half-width of the laser pulse. When $\Omega_1^{(0)}$ and $\Omega_2^{(0)}$ are both equal to 4×10^9 rad/s [the conditions (7) are destroyed], the solution of Eq. (1) shows that the population oscillates between the states $|1\rangle$ and $|2\rangle$, and the ionization probability calculated through $\int_{-\infty}^{\infty} |C_3(t)|^2 \gamma\, dt$, is only about 0.05 (Fig. 2). When $\Omega_1^{(0)}$ is increased to the value of γ with $\Omega_2^{(0)}$ unchanged, the ionization probability is about 0.03 and the population of the states $|1\rangle$ and $|2\rangle$ oscillates much faster (Fig. 3). Full ionization takes place when $\Omega_1^{(0)}$ and $\Omega_2^{(0)}$ are both equal to γ (Fig. 4).

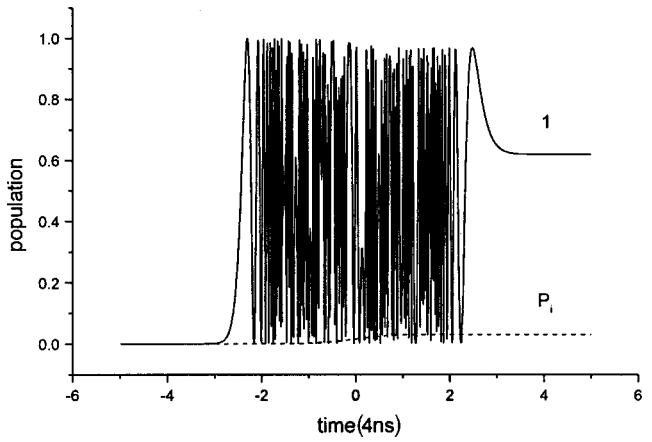

FIG. 3. Population evolution of a three-level system shown in Fig. 1. When $\Omega_1^{(0)} \sim \gamma$ and $\Omega_2^{(0)} \ll \gamma$, the population oscillates more quickly (compared with Fig. 2) between the states $|1\rangle$ and $|2\rangle$, and the final ionization probability is still very low. For clarity, only the population evolution of the state $|2\rangle$ (curve 1) and ionization probability (P_i) are shown here.

B. Three-step photoionization

Figure 5 shows the processes of three-step photoionization. In the traditional scheme, three lasers are set to act with the atomic system simultaneously. Within RWA approximation, the probability amplitudes $C1$, $C2$, $C3$, and $C4$ of the states $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$ satisfy the time-dependent *Schrödinger* equation:

$$\begin{cases} i\dot{C}_1 = \frac{1}{2}\Omega_1 C_2, \\ i\dot{C}_2 = \frac{1}{2}\Omega_1 C_1 + \Delta_1 C_2 + \frac{1}{2}\Omega_2 C_3, \\ i\dot{C}_3 = \frac{1}{2}\Omega_2 C_2 + \Delta_2 C_3 + \frac{1}{2}\Omega_3 C_4, \\ i\dot{C}_4 = \frac{1}{2}\Omega_3 C_3 - \frac{1}{2} i\gamma C_4, \end{cases} \tag{9}$$

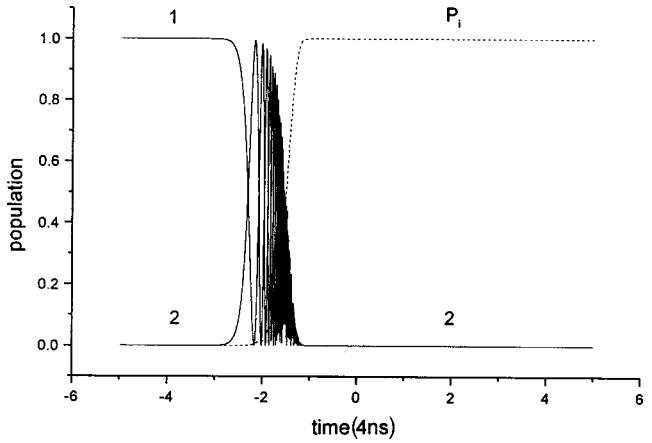

FIG. 4. Similar to Fig. 2, when the conditions (7) are satisfied, i.e., $\Omega_1^{(0)} \sim \Omega_2^{(0)} \sim \gamma$. Curves 1 and 2 represent the population of the states $|1\rangle$ and $|2\rangle$, and the curve P_i the ionization probability.

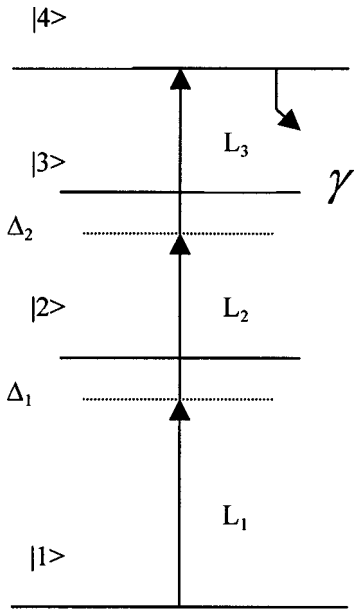

FIG. 5. Energy-level diagram of a four-level system. Laser L_1 couples the initially populated state $|1\rangle$ with an intermediate state $|2\rangle$, laser L_2 couples the two intermediate states $|2\rangle$ and $|3\rangle$, and laser L_3 couples the higher intermediate state $|3\rangle$ with an autoionization state $|4\rangle$. Δ_1 and Δ_2 are frequency detunings, and γ is the decay rate of the state $|4\rangle$.

where γ is the decay rate of the autoionization state $|4\rangle$, and Δ_1, Δ_2 are the detunings of the laser frequencies off the resonance centers of $|2\rangle - |1\rangle$ and $|3\rangle - |2\rangle$ transitions, respectively. Compared with Eqs. (1), Eqs. (9) are essentially the same. Hence, from the fourth equation of Eqs. (9) and the initial condition $C_4(0) = 0$, it can be shown that if $\Omega_3 \ll \gamma$, then

$$|C_4(t)|^2 \sim 0. \tag{10}$$

In this situation, the population is oscillated among mainly the three states $|1\rangle$, $|2\rangle$, and $|3\rangle$, leading to low ionization efficiency. Obviously, one of the necessary conditions for efficient photoionization is

$$\Omega_3 \sim \gamma. \tag{11}$$

Analyzing in the same way as in the case of two-step photoionization, necessary conditions for efficient ionization can be obtained as

$$\Omega_1 \sim \Omega_2 \sim \Omega_3 \sim \gamma. \tag{12}$$

Because γ may be as large as $10^{12}\,\mathrm{s}^{-1}$ or much larger, conditions (7) and (12) require all the laser fields must be intense enough, otherwise, efficient photoionization is impossible. This is the critical disadvantage of the traditional scheme.

III. A SCHEME

Although conditions (4) or (11) must be satisfied for efficient photoionization, conditions (7) or (12) can be destroyed if the ionization laser pulse (L_2 pulse for two-step photoion-

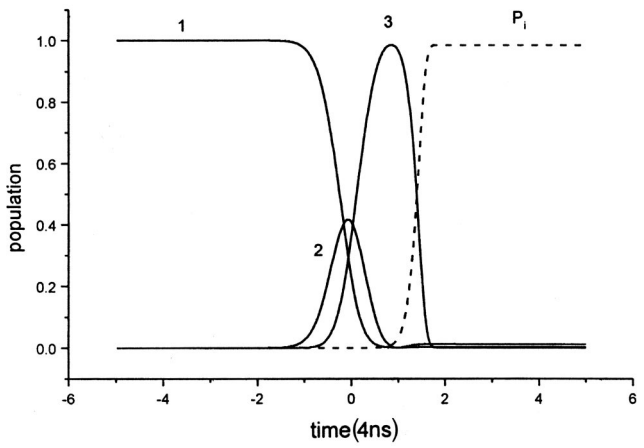

FIG. 6. Population evolution of the four-level system shown in Fig. 5 via the new scheme with $\Omega_1^{(0)}=\Omega_2^{(0)}=8\times10^8$ rad/s. Curves 1, 2, and 3 present the population of the states $|1\rangle$, $|2\rangle$, and $|3\rangle$, respectively, and curve P_i presents the probability of ionization.

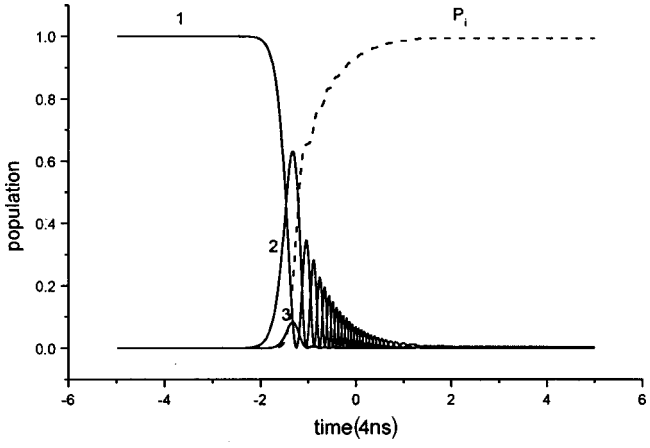

FIG. 7. Similar to Fig. 6, except that the traditional scheme was used with $\Omega_1^{(0)}=\Omega_2^{(0)}=1.4\times10^{10}$ rad/s.

ization, L_3 pulse for three-step photoionization) begins action after the total population is transferred into the state $|2\rangle$ (for two-step photoionization) or the state $|3\rangle$ (for three-step photoionization) via the other laser pulses. In this way, the intensity of the laser pulses, apart from the ionization pulses, may be lowered considerably without losing efficient ionization because the laser power required by total population transfer is not high [3,8–11]

For three-step photoionization (Fig. 5), a scheme is proposed as following: the STIRAP technique [10] is used for population transfer from the ground state $|1\rangle$ to the state $|3\rangle$, i.e., the L_2 pulse acts first, then the L_1 pulse; after all the population is in the state $|3\rangle$, the L_3 pulse is switched on to pump the population of the state $|3\rangle$ into the autoionization state $|4\rangle$. For comparing this new scheme with the traditional one, we performed the following numerical calculations under conditions that $\gamma=8\times10^{11}\,\mathrm{s}^{-1}$, *and* $\Delta_1=\Delta_2=0$. The sequence of the laser pulses can be expressed exactly in the Rabi frequencies:

$$\begin{cases} \Omega_1(t)=\Omega_1^{(0)}\exp[-(t-0.15\tau)^2/\tau^2], \\ \Omega_2(t)=\Omega_2^{(0)}\exp[-(t+0.15\tau)^2/\tau^2], \\ \Omega_3(t)=\Omega_3^{(0)}\exp[(t-2.5\tau)^2/\tau^2], \end{cases} \tag{13}$$

where τ is taken as 4×10^{-9} s, the width of the laser pulses. When $\Omega_1^{(0)}=\Omega_2^{(0)}=8\times10^8$ rad/s, and $\Omega_3^{(0)}=4\times10^{11}$ rad/s, numerical results of Eqs. (9) show that the probability of photoionization is nearly unit without population oscillating, as can be seen in Fig. 6. In contrast with the new scheme, if the three laser pulses are set to act simultaneously (the traditional scheme) with the other conditions unchanged, the photoionization probability was calculated to be 0.015. Keeping the value of $\Omega_3^{(0)}$ unchanged, we increase the values of $\Omega_1^{(0)}$ and $\Omega_2^{(0)}$, step-by-step. When the values of $\Omega_1^{(0)}$ and $\Omega_2^{(0)}$ increase up to 1.4×10^{10} rad/s, about 17 times larger than the original ones, the photoionization probability was found to be unit. Under these conditions, the population oscillates many times among states $|1\rangle$, $|2\rangle$, and $|3\rangle$, as can be seen from Fig. 7. From the above calculations, a conclusion can be drawn that when the new scheme is used, Rabi frequency can be decreased by at least one order of magnitude (i.e., the intensity of L_1 *and* L_2 can be decreased at least two orders of magnitude) without losing high ionization efficiency.

For two-step photoionization (Fig. 1), the new scheme can be realized as follows: After the population in the ground state $|1\rangle$ is transferred into the state $|2\rangle$ by the L_1 pulse with its frequency chirping [4,5] or oscillating [9], the L_2 pulse is switched on to pump the population of the state $|2\rangle$ into an autoionization state $|3\rangle$. Numerical calculations show that the intensity of the L_1 pulse can be lowered considerably. However, this technique can only be used for the areas where high selectivity of photoionization is not required.

IV. APPLICATION FOR ^{235}U

In the natural uranium mine, the abundance of ^{235}U is only 0.72%. In order to ensure the selectivity of laser isotope separation of ^{235}U, three-step resonance photoionization is usually put into use. A possible route of photoionization is diagrammed in Fig. 8. Because of the nuclei spin momentum $I=7/2$, an electronic level splits into 8 sublevels (HFS). The relevant parameters of every energy level are listed in Table I. Although there is no report on autoionization states of ^{235}U up to now, autoionization states of rare-earth elements have been observed in plenty of experiments. So, we suppose the existence of an autoionization state of ^{235}U (Fig. 8) with a lifetime of 8×10^{-11} s, corresponding to a linewidth of 4 cm^{-1}. According to the Fano theory, the reason for an autoionization level broadening is that a bound state above the ionization threshold couples with the continuum states. Thus the autoionization state of ^{235}U should correspond to a bound state above the ionization threshold, and we assume that the HFS constants of this bound state are the same as that of $^7M_6^0$.

We consider three linear polarization lasers interact with the multilevel system of ^{235}U, as shown in Fig. 8. From the selection rule of dipole transition, $\Delta F=0,\pm1$, the allowed

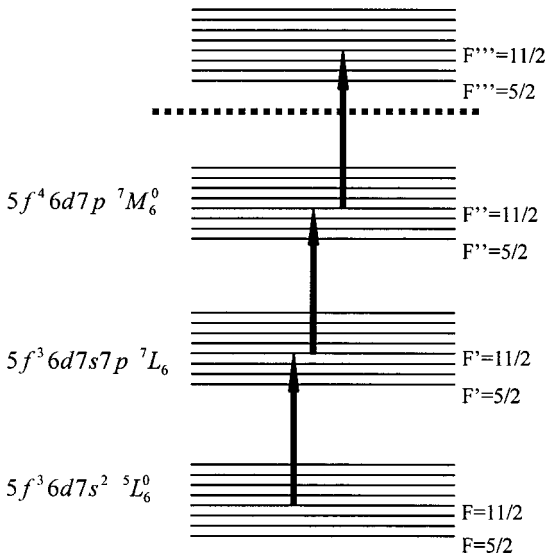

FIG. 8. The diagram of three-step phtonionization of ^{235}U. For clarity, only shows the resonance of center frequencies of three lasers with the sublevels with equal F number (11/2). For convenience, the lowest sublevel is chosen as $F=5/2$. In fact, the sublevels with larger F number maybe have lower energy than the one with smaller F number.

transitions are shown in Fig. 9. (For clarity, Fig. 8 only shows the resonance transition between the sublevels with the same F number, 11/2.) In fact, because the magnetic quantum number $M=-F,\ -F+1,\ldots,\ F-1,\ F$, there still are $(2F+1)$ fold degeneracies in every HFS level. The result is far more than 32 states coupled with the three lasers. However, because the lasers are linear polarized, the transition rule of magnetic sublevels should be $\Delta M=0$. Therefore, only 32 sublevels shown in Fig. 8 need to be considered theoretically. In the energy representation, the *Schrödinger* equation can be written as

$$i\hbar\frac{\partial C_n}{\partial t}=\sum_m H_{nm}C_m \quad (m,n=1,2,\ldots,32) \tag{14}$$

where C_n is the probability amplitude of every state, and H_{mn} is the Hamiltonian matrix

$$H_{mn}=\begin{pmatrix} H_{11} & H_{12} & 0 & 0\cdots 0 \\ H_{21} & H_{22} & H_{23} & 0\cdots 0 \\ \vdots & \ddots & & \\ \vdots & & H_{31,32} & H_{31,32} \\ 0 & 0 & H_{32,31} & H_{32,32} \end{pmatrix},$$

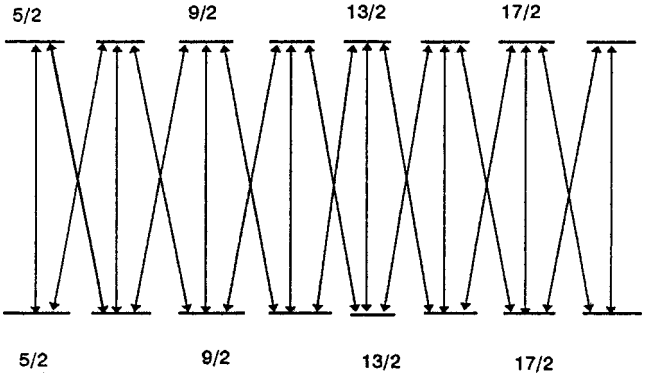

FIG. 9. The diagram of allowed dipole transition among sublevels according to $\Delta F=0,\pm 1$.

where H_{mm} is the detuning of the laser frequencies from the respective levels. A decay term $-i\gamma/2$ is added to the diagonal elements of the Hamiltonian of the autoionization levels, where $\gamma=8\times10^{11}\ \mathrm{s}^{-1}$. The off-diagonal elements are calculated according to Ref. [12]:

$$\langle \gamma FM\|\gamma' F' M'\rangle=[FF']^{1/2}\begin{pmatrix} J & 1 & F \\ F' & 1 & J' \end{pmatrix}\langle \gamma J|P|\gamma' J'\rangle. \tag{15}$$

In most experiments, pulsed lasers are usually used for the photoionization of ^{235}U. The laser pulses in our consideration are of Gaussian envelope with a pulse width of 4×10^{-9} s. Although the frequencies of the three lasers are fixed at the corresponding transitions ($F=11/2$, c.f., Fig. 8), other sublevels may also be coupled by the lasers because the Fourier broadening of the pulses is of GHz. Accordingly, population evolution of the multilevel system, shown in Fig. 8, should be different from that of the four-level system shown in Fig. 5. In the following numerical calculations, all the 32 levels are included according to Eqs. (14).

When our new scheme of photoionization is performed for ^{235}U, i.e., the Rabi frequencies being of the form of Eqs. (13), the atoms with initial population in the 11/2 sublevel of the ground state $5f^3 6d7s^2\ ^5L_6^0$ can be completely ionized with $\Omega_1^{(0)}=\Omega_2^{(0)}=3\times10^9$ rad/s and $\Omega_3^{(0)}=4\times10^{11}$ rad/s. The population evolution is shown in Fig. 10. Under the same conditions, except for simultaneous interaction of the three pulsed lasers, however, the photoionization probability decreases to 0.11. Keeping $\Omega_3^{(0)}$ unchanged and increasing the value of $\Omega_1^{(0)}$ and $\Omega_2^{(0)}$ step-by-step, the photoionization probability reaches to unit when $\Omega_1^{(0)}=\Omega_2^{(0)}=2\times10^{10}$ rad/s. The population evolution under these con-

TABLE I. The constants of ^{235}U level shown in Fig. 8, E is energy, LT is the lifetime, A, B is the constant of HFS.

	$5f^3 6d7s^2\ ^5L_6^0$	$5f^3 6d7s7p\ ^7L_6$	$5f^4 6d7p\ ^7M_6^0$
E (cm^{-1})	0	17 362[12]	34 161[12]
LT (ns)	∞	410[14]	47[14]
A (MHz)	−60.56[12]	−95.45[13]	86.9[15]
B (MHz)	4104.15[12]	2843.0[13]	1313.1[15]

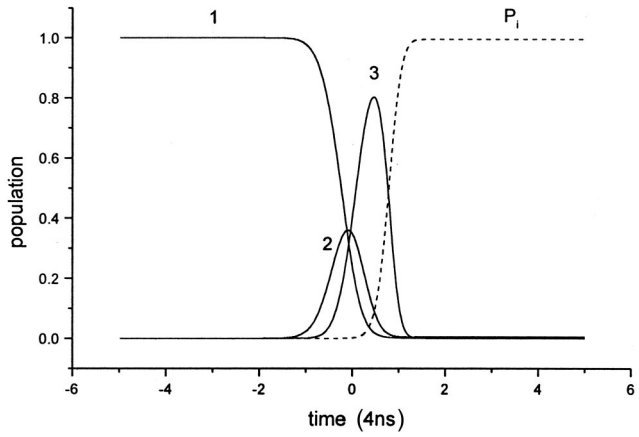

FIG. 10. Population evolution of ^{235}U photoionized by the scheme with $\Omega_1^{(0)}=\Omega_2^{(0)}=3\times10^9$ rad/s. Curves 1, 2, and 3 present the population of sublevels with $F=11/2$, $F'=11/2$, $F''=11/2$, respectively, and curve P_i presents the photoionization probability.

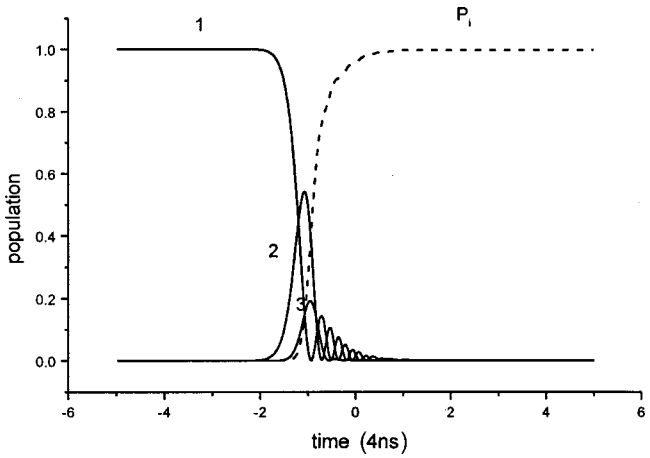

FIG. 11. Similar to Fig. 10 except that the traditional scheme was used with $\Omega_1^{(0)}=\Omega_2^{(0)}=2\times10^{10}$ rad/s.

ditions is shown in Fig. 11. Compared with the new scheme, the intensity of two laser pulses must be increased by about 50 times with the traditional technique for complete ionization.

V. CONCLUSION

A scheme for selective photoionization of atoms (molecules) is proposed, to our knowledge, for the first time. Several numerical calculations of realistic systems show that the intensity of the laser beams coupled with the bound states can be lowered by about two orders of magnitude via this new scheme. Applying the new method to the laser isotope separation of ^{235}U may bring enormous economic benefit.

ACKNOWLEDGMENT

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