

Distinction of different two-photon transition pathways by spatial coherent control

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Abstract: The spatial coherent control of resonant two-photon transitions in atomic rubidium using counter-propagating ultrashort laser pulses is investigated in this work. There are two transition pathways involved in the spatial excitation pattern. A special pulse shaping scheme is designed to distinguish their individual contribution. The scheme consists of a π -step function flipping at the poles and zeros of the transition amplitude function and two segmented linear function with different slopes, and it can make the desired transition pathway dominate at the assigned spatial position. The simulations show that our scheme is quite effective compared to the previous single linear shaping scheme.

Key Words: spatial coherent control, pathway distinction, resonant two-photon transitions, pulse shaping

1 Introduction

Quantum coherent control via light-matter interactions can steer the quantum dynamics in atomic and molecular systems. In early experiments, shaped ultrashort laser pulses [1] were employed to control the quantum processes. Two-photon absorption (TPA) is one fundamental quantum process and numerous control schemes have been proposed to achieve optimal enhancement and selective excitation. The simplest case is one two-photon transition pathway in a three-level quantum system [2–6]. When there are two pathways in a four-level quantum system, the quantum coherent control and pathway manipulation can be further investigated by considering the interference between the two pathways [7–10]. Spatial coherent control can induce different interference patterns in space, and thus has many applications in physics [11–13].

Itan Barmes introduced the spatial degree of freedom into the TPA control by using a pair of counter-propagating ultrashort frequency combs [14, 15], and they demonstrated spatially selective excitation of individual species in a multi-component gas mixture for non-resonant TPA. Subsequently, Dirk Boonzajer [16] and Woojun Lee [17] investigated the spatial coherent control for the resonant TPA of $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D$ in atomic rubidium.

In this paper, we described a spatial coherent control scheme for the distinction of two resonant TPA pathways in atomic rubidium, which are $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D$ and $5S_{1/2} \rightarrow 5P_{1/2} \rightarrow 5D$. In Section 2, the spatial coherent control experiment set-up and TPA theoretical model induced by the counter-propagating ultrashort pulse pairs are introduced. Section 3 gives several phase shaping schemes and the simulations, and the scheme containing double linear

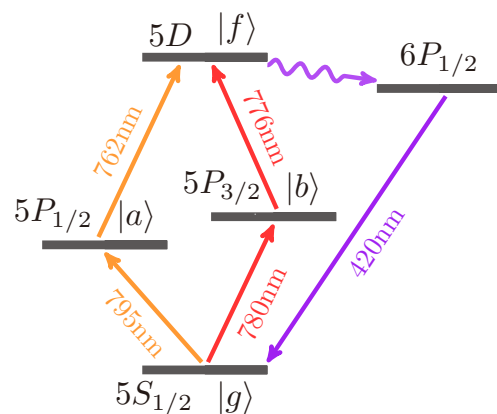


Figure 1: (Color online) The energy-level diagram of atomic rubidium. The atoms are excited from the ground state $5S_{1/2}$ ($|g\rangle$) to the final state $5D$ ($|f\rangle$) through the two intermediate states $5P_{1/2}$ ($|a\rangle$) and $5P_{3/2}$ ($|b\rangle$), respectively. There are two two-photon transition pathways: (i) pathway 1, $|g\rangle \rightarrow |a\rangle \rightarrow |f\rangle$ and (ii) pathway 2, $|g\rangle \rightarrow |b\rangle \rightarrow |f\rangle$. The pulse spectrum is centered on the two-photon transition frequency ($\omega_{fg}/2$). Fluorescence decayed from the $6P$ state at 420 nm is used to detect the excited state population.

functions can distinguish the pathway amplitude quite well. The conclusion is given in Section 4.

2 Theoretical considerations

The spectral spatial coherent control of atomic rubidium (level diagrams are presented in Fig. 1) with counter-propagating ultrashort pulse pairs is considered in this work. The layout of the experimental setup is shown in Fig. 2. Briefly, femtosecond optical pulses are shaped in a 4f-configuration zero-dispersion shaper and the phases are controlled by a spatial light modulator. The shaped pulse is firstly focused in a rubidium vapour cell and then reflected back by a mirror, so each counter-propagating laser pulse collides with the next pulse in the vapor cell. The excited state population is proportional to 420 nm fluorescence de-

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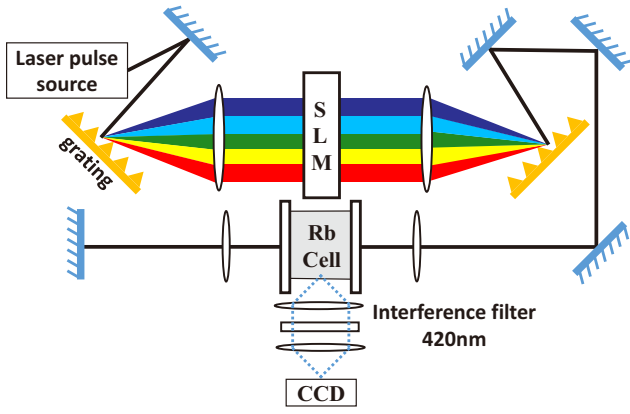


Figure 2: (Color online) Schematic set-up of the spatial coherent control experiment. The pulses are shaped by the spatial light modulator (SLM). Every two counter-propagating pulses collide in the vapour cell and excite the two-photon transitions. The fluorescence pattern is imaged by an charge-coupled device camera.

cayed from the $6P$ state, which is monitored by a charge-coupled device (CCD) camera.

According to the second-order time dependent perturbation theory, the two-photon transitions amplitude $U(t)$ induced by a weak laser field $\varepsilon(t)$ is

$$U(z, t) = -\frac{1}{\hbar^2} \sum_{n=a,b} \mu_{fn} \mu_{ng} \int_{-\infty}^t \int_{-\infty}^{t_1} \varepsilon(z, t_1) \varepsilon(z, t_2) \times e^{i\omega_{fn}t_1} e^{i\omega_{ng}t_2} dt_2 dt_1, \quad (1)$$

where μ_{fn} and μ_{ng} are the transition dipoles, and $\omega_{ij} = (E_i - E_j)/\hbar$ are the frequency difference between the state $|i\rangle$ and $|j\rangle$. The summation is performed over all possible intermediate states. The electric field is $\varepsilon(z, t) = \varepsilon_1(z, t) + \varepsilon_2(z, t)$, in which $\varepsilon_1(z, t)$ and $\varepsilon_2(z, t)$ propagate along the $+z$ and $-z$ directions respectively.

With the Fourier transformation, the final amplitude can be written in the frequency domain when the pulse is over

(i.e., $t \rightarrow \infty$).

$$U(z) = \sum_{n=a,b} i \frac{\mu_{fn} \mu_{ng}}{\hbar^2} \left[i\pi E(\omega_{ng}) E(\omega_{fn}) + \wp \int_{-\infty}^{\infty} \frac{E(\omega) E(\omega_{fg} - \omega)}{\omega_{ng} - \omega} d\omega \right], \quad (2)$$

where $E(\omega)$ is the Fourier transform of $\varepsilon(t)$.

For convenience, we define $\hat{\omega} = \omega_{fg} - \omega$, $v_n = \mu_{fn} \mu_{ng} / \hbar^2$ and $\omega_0 = \omega_{fg}/2$. Then the TPA amplitudes are can be rewritten as

$$U^{(n)}(z) = U_r^{(n)}(z) + U_{nr}^{(n)}(z), \quad (3)$$

$$U(z) = \sum_{n=a,b} \left[U_r^{(n)}(z) + U_{nr}^{(n)}(z) \right], \quad (4)$$

with

$$U_r^{(n)}(z) = -\pi v_n E(\omega_{ng}) E(\omega_{fn}), \quad (5)$$

$$U_{nr}^{(n)}(z) = i v_n \wp \int_{\omega_0}^{\infty} \frac{(\omega_{fn} - \omega_{ng}) E(\omega) E(\hat{\omega})}{(\omega - \omega_{ng})(\omega - \omega_{fn})} d\omega. \quad (6)$$

Here $U_r^{(n)}(z)$ denotes the resonant part of each pathway amplitude, which only depends on the resonance frequency; $U_{nr}^{(n)}(z)$ denotes the non-resonant part, which considers the contributions of the other spectral components.

$$E(\omega) = E_1(\omega) e^{-i\omega z/c} + E_2(\omega) e^{i\omega z/c}, \quad (7)$$

where $E_1(\omega) = f[\varepsilon_1(0, t)]$ and $E_2(\omega) = f[\varepsilon_2(0, t)]$ are, respectively, the Fourier transformation of electric field, and c is the speed of light.

Since the counter-propagating pulses are split from the same single laser pulse, they have the same spectral amplitude function $A(\omega)$ and phase function $\phi(\omega)$,

$$E_1(\omega) = E_2(\omega) = A(\omega) e^{i\phi(\omega)}. \quad (8)$$

Thus the resonant and non-resonant terms can be written as

$$U_r(z) = \sum_{n=a,b} U_r^{(n)}(z) = -4\pi \sum_{n=a,b} v_n A(\omega_{ng}) A(\omega_{fn}) e^{i\phi(\omega_{ng}) + i\phi(\omega_{fn})} \cos\left(\frac{\omega_{ng}z}{c}\right) \cos\left(\frac{\omega_{fn}z}{c}\right), \quad (9)$$

$$U_{nr}(z) = \sum_{n=a,b} U_{nr}^{(n)}(z) = 4i \int_{\omega_0}^{\infty} A(\omega) A(\hat{\omega}) e^{i\phi(\omega) + i\phi(\hat{\omega})} \cos\left(\frac{\hat{\omega}z}{c}\right) \cos\left(\frac{\omega z}{c}\right) f(\omega) d\omega, \quad (10)$$

where

$$f(\omega) = \frac{(\omega - \omega_{fo})(\omega - \omega_{og})(\omega_{fa} - \omega_{ag})(\omega_{fb} - \omega_{bg})}{(\omega - \omega_{fa})(\omega - \omega_{fb})(\omega - \omega_{ag})(\omega - \omega_{bg})},$$

$$\omega_{fo} = \omega_0 + \frac{\gamma}{2} \sqrt{(\omega_{fa} - \omega_{ag})(\omega_{fa} - \omega_{ag})},$$

$$\omega_{og} = \omega_0 - \frac{\gamma}{2} \sqrt{(\omega_{fa} - \omega_{ag})(\omega_{fa} - \omega_{ag})},$$

$$\gamma = \sqrt{\frac{\beta + \alpha}{1 + \alpha\beta}}, \quad \alpha = \frac{v_b}{v_a}, \quad \beta = \frac{\omega_{fb} - \omega_{bg}}{\omega_{fa} - \omega_{ag}}.$$

The $f(\omega)$ changes the sign at the poles (i.e., ω_{fn} and ω_{ng}) and zeros (i.e., ω_{fo} and ω_{og}) [10]. By flipping the phases at these characteristic frequencies, we can avoid the destructive interferences.

At a certain spatial position z , the trigonometric factor (i.e., cosine function) of the non-resonant integral $U_{nr}(z)$ leads to a period of $\pi c/z$ for ω . When the phase item (i.e., $e^{i\phi(\omega) + i\phi(\hat{\omega})}$) is also set as a periodic function with the same period of $\pi c/z$, the maximal transition amplitude will be

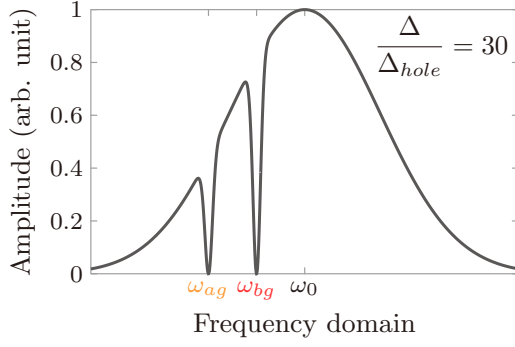


Figure 3: (Color online) The amplitude function $A(\omega)$ vs ω centered at ω_0 . There are two holes located at the resonance frequencies ω_{ag} and ω_{bg} , respectively. Here $\Delta/\Delta_{hole} = 30$ is used.

achieved. As the cosine is an even function, the spatial symmetry relationships are obtained as $U(z) = U(-z)$ and $U^{(n)}(z) = U^{(n)}(-z)$.

3 Results and discussion

To involve the quantum interference between pathways, laser pulses with a wide spectral bandwidth (full width at half maximum) about 45 nm are employed. We use a Gaussian amplitude function $A(\omega)$ with two holes around the two resonant frequencies ω_{ag} and ω_{bg} , shown in Fig. 3, which can effectively eliminate the resonant transition background and thus make the spectral peaks more obvious.

$$A(\omega) = \exp\left\{-\frac{(\omega - \omega_0)^2}{2\Delta^2}\right\} \left[1 - \exp\left\{-\frac{(\omega - \omega_{ag})^2}{2\Delta_{hole}^2}\right\}\right] \times \left[1 - \exp\left\{-\frac{(\omega - \omega_{bg})^2}{2\Delta_{hole}^2}\right\}\right], \quad (11)$$

where $\Delta \gg \Delta_{hole}$.

We give some pulse shaping methods based on the use of linear and π -step phase-shaping.

$$\phi(\omega) = \phi_L(\omega) + \Theta(\omega), \quad (12)$$

$$\phi_L(\omega) = \begin{cases} k_{d1}(\omega - \omega_0) & \omega \in (\omega_0, \omega_{fo}) \\ k_{d2}(\omega - \omega_{fo}) & \omega \in (\omega_{fo}, +\infty) \end{cases}, \quad (13)$$

$$\Theta(\omega) = \begin{cases} -\frac{\pi}{2} & \omega \in (\omega_0, \omega_{fb}) \cup (\omega_{fo}, \omega_{fa}) \\ +\frac{\pi}{2} & \omega \in (\omega_{fb}, \omega_{fo}) \cup (\omega_{fa}, +\infty) \end{cases}, \quad (14)$$

where $\phi(\omega)$ is a linear function and $\Theta(\omega)$ is a π -step function.

Three phase-shaping schemes are listed for comparison in the following.

- 1) Scheme A uses the transform limited pulse: $\phi(\omega) = 0$, $\omega \in (\omega_0, \infty)$;
- 2) Scheme B is single linear shaping scheme: $k_{d1} = k_{d2}$;
- 3) Scheme C is double linear shaping scheme: $k_{d1} \neq k_{d2}$.

Without loss of generality, we assume $\phi(\omega) = 0$ when $\omega \in (-\infty, \omega_0)$. Here $\Theta(\omega)$ is a π -step function to compensate the sign change of $f(\omega)$, shown in Fig 4(a). And $\phi_L(\omega)$ is linear function with slopes k_{d1} and k_{d2} , shown in Fig

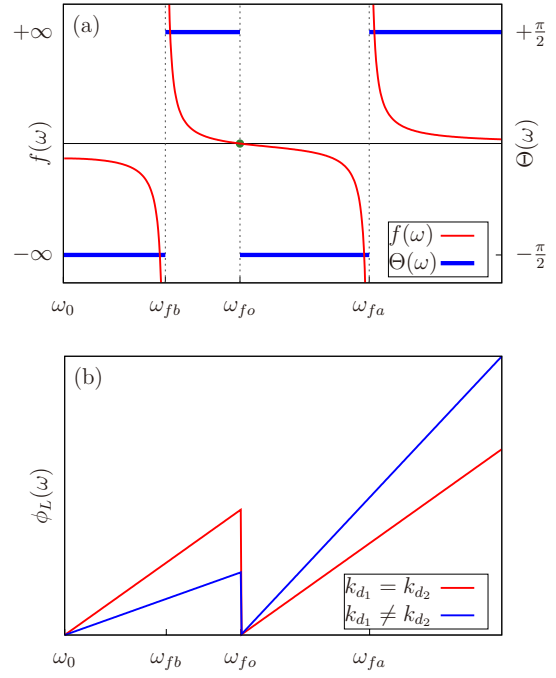


Figure 4: (Color online) The phase shaping schemes. Here $\Theta(\omega)$ and $\phi_L(\omega)$ is 0 when $\omega \in (-\infty, \omega_0)$ (not shown). (a) The step function $\Theta(\omega)$ is around characteristic frequencies where $f(\omega)$ changes its sign. (b) The linear function $\phi_L(\omega)$ with $k_{d1} = k_{d2}$ and $k_{d1} \neq k_{d2}$.

4(b), which generate local extremums at positions $k_{d1}c/2$ and $k_{d2}c/2$ via resonance enhancement. In these schemes, only scheme C can achieve effective pathway distinction.

The numerical simulations are shown in Fig. 5. With the scheme A, there is no significant information, except for the enhancement around $z = 0$, shown in Fig. 5(a). In the scheme B, the single linear function $k_{d1} = k_{d2}$ leads to the enhancement at position $k_{d1}c/2 = 400 \mu\text{m}$, shown in Fig. 5(b). In the inset of Fig. 5(b), the two pathway amplitudes overlap at $400 \mu\text{m}$.

In our distinction scheme C, there are two local extremums at $k_{d1}c/2 = 200 \mu\text{m}$ and $k_{d2}c/2 = 600 \mu\text{m}$, shown in Fig. 5(c). The two peak positions are resulted from the linear functions $k_{d1}(\omega - \omega_0)$ and $k_{d2}(\omega - \omega_{fo})$, respectively. Pathway 2 dominate the two-photon transitions at the position $k_{d1}c/2$, and pathway 1 dominate at $k_{d2}c/2$. The two-photon transitions $|U^{(b)}(z)|^2$ at $200 \mu\text{m}$ and $|U^{(a)}(z)|^2$ at $600 \mu\text{m}$ in scheme C are, respectively, approximately equal to $|U^{(b)}(z)|^2$ and $|U^{(a)}(z)|^2$ at $400 \mu\text{m}$ in scheme B. In other words, scheme C could distinguish the two different transition pathways by making them scatter at different positions with double linear functions.

4 Conclusion

The spatial coherent control of TPA pathways in atomic rubidium is performed using the counter-propagating ultra-short pulse pairs. A special scheme containing a π -step function and double segmented linear functions is designed to distinguish the two pathways in space. The π -step function part flips the sign at the poles and zeros of the transition amplitude function to make constructive interference between

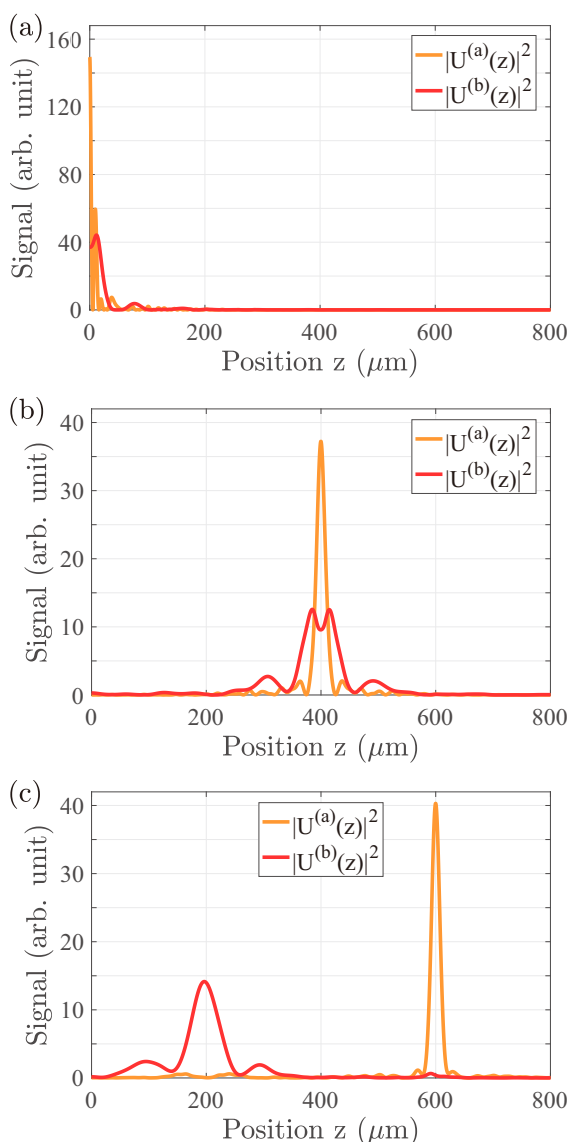


Figure 5: (Color online) The numerical simulations with different phase-shaping schemes. (a) The transform limited pulse (scheme A). (b) Single linear shaping scheme (scheme B) with $k_{d_1} = k_{d_2} = 2.67$ ps. (c) Double linear shaping scheme (scheme C) with $k_{d_1} = 1.33$ ps and $k_{d_2} = 4.00$ ps. Here $|U(z)|^2$ is proportional to the fluorescence imaged by the CCD. And $|U^{(n)}(z)|^2$ ($n = a, b$) is the TPA contribution of each transition pathway. As $U(z)$ and $U^{(n)}(z)$ are symmetric around $z = 0$, only the part of $z > 0$ is shown.

the two pathways. Meanwhile, the slope of each segmented linear function determines the position of the local excited extremums. The numerical simulations also demonstrate the effectiveness of pathway distinction. This scheme also can be extended to distinguish multiple two-photon transition pathways.

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