Coherent control of resonant two-photon transitions by counter-propagating ultrashort pulse pairs

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We describe optimized coherent control methods for two-photon transitions in atoms of a ladder-type three-state energy configuration. Our approach is based on the spatial coherent control scheme, which uses counter-propagating ultrashort laser pulses to produce complex excitation patterns in an extended space. Because coherent control requires constructive interference of constituent transition pathways, applying it to an atomic transition with a specific energy configuration requires specially designed laser pulses. We show in an experimental demonstration that two-photon transition with an intermediate resonant energy state can be coherently controlled and retrieved from the resonance-induced background, when phase-flipping of the laser spectrum near the resonant intermediate transition is used. A simple reason for this behavior is the fact that the transition amplitude function (added to give an overall two-photon transition) changes its sign at the intermediate resonant frequency, and thus, by proper spectral-phase programming, the excitation patterns (or the position-dependent interference of the transition given as a consequence of the spatial coherent control) can be well isolated in space along the focal region of the counter-propagating pulses.

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I. INTRODUCTION

Femto-second laser optics have been widely used over the last two decades as a time-resolving spectroscopic means of studying the ultrafast time-scale dynamics of a variety of quantum systems, including atoms, molecules, and quasi-particles in solids [1–5]. The extreme peak intensity of femto-second laser pulses has also enabled high-order nonlinear optical processes such as multi-photon excitations, high-harmonic generations, and above-threshold ionizations, to list a few [6–9]. Besides these uses, femto-second lasers has gradually become an important tool in the field of coherent control [10–12], where shaped laser pulses steer quantum processes towards certain desirable outcomes. The information of as-obtained laser pulse shapes through coherent control often plays a crucial role in understanding the quantum structure of the materials under consideration [13–16]. In this regard, researchers can furthermore analytically design the optimized laser pulse shapes for more selective and efficient nonlinear optical processes [17–23].

The recent demonstration [24] of a counter-propagating pair of ultrafast laser pulses coherently inducing Doppler-free two-photon transitions of atoms shows the intriguing possibility for its use in ultra-precision spectroscopy [25], especially in conjunction with a femto-second frequency comb [26, 27]. This method, termed spatial coherent control, coherently arranges in time the spectral components of the laser pulse in such a way that all of the counter-propagating photon pairs, energy-resonant to the atomic transition (i.e., \( h\omega_1 + h\omega_2 = E_e - E_g \)), collide only at specific locations along the beam direction. Because the resonance condition varies from one atom species to another, the atom-specific spectroscopic information can be retrieved by imaging the distinct spatial excitation profile, if a proper coherent control scheme is used. So far, this powerful method of spatial coherent control has been demonstrated for non-resonant two-photon transitions (or for cases in which intermediate states are not directly involved with the transition), and not yet demonstrated for resonant two-photon transitions (two-photon transitions with a resonant intermediate state or states).

In this paper, we describe an experimental demonstration of the spatial coherent control of the resonant two-photon transition \( 5S_{1/2} \rightarrow 5P_{2/3} \rightarrow 5D \) of atomic rubidium (\(^{85}\text{Rb}\)). Ultrashort laser pulses are programmed in such a way that not only (1) counter-propagating photon pairs, and no photon pairs in the same propagation direction, induce the given transition, but also (2) the contributions from all possible combinations of photon energies involving such transition are coherently added for an optimal net transition probability. To accomplish this, we first adopt spectral pulse-shaping methods: V-shape spectral phase programming for condition (1) and a spectral step phase for condition (2) in the first and second experiments. On the basis of single-pulse-based pulse-shaping method [17, 23], we resolve the issue of the resonant intermediate transition and extend the method in the context of the spatial excitation pattern formation by counter-propagating pulses. In the subsequent experiment, we use additionally a spectral amplitude shaping method to further enhance the net two-photon transition, in which the resonant intermediate transition is spectrally blocked so that the spatially extended excitation caused by sequential two-photon transitions through the

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intermediate resonant state is completely avoided. 

In the remaining sections, we first theoretically sketch the laser pulse shaping ideas relevant for the resonant and non-resonant two-photon transitions, respectively, in Sec. II, before the experimental procedure is described in Sec. III. We then present the experimental results of the spatial coherent control of the two-photon transition of $^{85}$Rb in Sec. IV and our conclusions in Sec. V.

II. THEORETICAL CONSIDERATIONS

We consider a pair of laser pulses, denoted by $\mathcal{E}_1(z,t)$ and $\mathcal{E}_2(z,t)$, respectively propagating along the $\pm z$ directions interacting with a three-state atom in a ladder-type energy configuration. The dynamics of the three-state system is governed by the Schrödinger equation, which reads

$$i\hbar \frac{dc(t)}{dt} = H(t)c(t),$$

(1)

where $c = [c_g(t), c_i(t), c_r(t)]^T$ is the column vector with the probability amplitudes $c_g(t)$, $c_i(t)$, and $c_r(t)$ of the three-states; $|\psi_g\rangle$ (the ground state), $|\psi_i\rangle$ (the intermediate state), and $|\psi_r\rangle$ (the final state). In the perturbative interaction regime, the two-photon transition amplitude $c_r(t)$ is then obtained from the second-order Dyson series as

$$c_r(z,t) = -i \frac{\mu_{ei} \mu_{ig}}{\hbar^2} \left[ \mathcal{E}_1(\omega_{ig}) \mathcal{E}_1(\omega_{ei}) + \mathcal{E}_2(\omega_{ig}) \mathcal{E}_2(\omega_{ei}) e^{2i\omega_{ig}z/c} + \mathcal{E}_1(\omega_{ig}) \mathcal{E}_2(\omega_{ei}) e^{2i\omega_{ig}z/c} \right],$$

(5)

and

$$c_{nr}(z) = i \frac{\mu_{ei} \mu_{ig}}{\hbar^2} \int_{-\infty}^{\infty} \frac{d\omega}{\omega_{ig} - \omega} \left[ \mathcal{E}_1(\omega) \mathcal{E}_1(\omega_{eg} - \omega) + \mathcal{E}_2(\omega) \mathcal{E}_2(\omega_{eg} - \omega) e^{2i\omega_{ig}z/c} \right] + \mathcal{E}_1(\omega) \mathcal{E}_2(\omega_{eg} - \omega) e^{2i\omega_{ig}z/c} + \mathcal{E}_2(\omega) \mathcal{E}_1(\omega_{eg} - \omega) e^{2i\omega_{ig}z/c},$$

(6)

where $\omega_{eg} = \omega_{ei} + \omega_{ig}$.

Note that the first two terms in each bracket in Eqs. (5) and (6) are the single-pulse contributions from either direction of the pulse propagations while the last two terms are caused by the counter-propagation of the pulses. The phase factors of the last two terms in $c_{nr}(z)$ have both $\omega$- and $z$-dependence and, thus, are involved in the integration over $\omega$, generating a non-trivial macroscopic spatial two-photon excitation profile which can be programmed via spectral phase modulation. The other terms have only rapidly oscillating or constant phase factors, making no contribution to the macroscopic spatial profile. With the last two terms of $c_{nr}(z)$, the spatial excitation probability is given by, proportionally,

$$S(z) \propto \left[ \int_{-\infty}^{\infty} d\omega \frac{A(\omega) A(\omega_{eg} - \omega)}{\omega_{ig} - \omega} e^{i[\Phi(\omega) + \Phi(\omega_{eg} - \omega)]} \left[ e^{2i\omega z/c} + e^{2i(\omega_{eg} - \omega)z/c} \right] \right]^2,$$

(7)

where the spectral amplitude function $A(\omega)$ and the spectral phase function $\Phi(\omega)$ are defined the same for both pulses, when they are split from a single laser pulse, as

$$E_1(\omega) = E_2(\omega) = A(\omega) e^{i\Phi(\omega)}.$$
A. Two-photon transition without a resonant intermediate state

First we consider the non-resonant two-photon transition case, in which the one-photon transition to the intermediate state $|\psi_i\rangle$ is out of the laser spectrum (i.e., $\omega_i < \omega_{\text{min}}$). In this case, we can ignore the sign change of the denominator $\omega_i - \omega$ in Eq. (7) across the laser spectrum $\omega_i$ in the context of spatial coherent control, and the phase function $\Phi(\omega)$ can be programmed in such a way that the two-photon transition components in the integral calculation in Eq. (7) satisfy a constructive interference condition only at specific positions. For example, with a spectral phase such as

$$\Phi_V(\omega) = \alpha |\omega - \omega_o|,$$  \hspace{1cm} (9)

called a V-shape spectral phase, where $\omega_o$ denotes the two photon center ($\omega_o = \omega_{\text{ig}}/2$), each of the position-dependent oscillating phase terms in the integrand in Eq. (7) is canceled out at the points $z = \pm z_o$, where $z_o = \alpha c$. Because $\omega < \omega_0$ and $\omega > \omega_0$ parts of the modulated phase term have opposite slopes, the V-shape phase in Eq. (9) divides a pulse in the time domain into two sub-pulses, each of which has a group delay of $-\alpha$ for the $\omega > \omega_0$ part and of $\alpha$ for the $\omega < \omega_0$ part, from the group delay relation, $\alpha = -|d\Phi_V/\omega|$. So, a pair of counter-propagating pulses with these sub-pulses makes two distinct local excitations at the points where a red sub-pulse and a blue sub-pulse meet up to fulfill the two-photon transition condition. The spatial excitation profile $S(z)$ in Eq. (7) is, therefore, conceptually given in this case by

$$S(z) \propto \delta(z - z_o) + \delta(z + z_o),$$  \hspace{1cm} (10)

where $\delta(z)$ is a Dirac delta function. Figure 1 compares the numerically calculated spatial excitation patterns for various phase-programming. As shown in Figs. 1 (a,b), the V-shape spectral phase, $\Phi_V(\omega)$, causes local excitations to occur at points $z = \pm z_o$ in non-resonant two-photon transitions [25].

B. Two-photon transitions with a resonant intermediate state

However, in the resonant two-photon transition case, in which the intermediate state is located within the laser spectrum (i.e., $\omega_{\text{min}} < \omega_i < \omega_{\text{max}}$), we need alternative phase function programming to deal with the sign change of the denominator $\omega_i - \omega$ in Eq. (7). When the position dependence is ignored, it is known that, by flipping the phase of either part of $\omega < \omega_{\text{ig}}$ or $\omega > \omega_{\text{ig}}$, the integrand becomes in-phase across $\omega_{\text{ig}}$, resulting in an enhancement of the two-photon transition [17, 23]. This phase flipping can be achieved with step-phase modulation such as $\pi \Theta(\omega - \omega_i)$, where $\Theta(x)$ denotes the Heaviside step function. If we combine this step-phase modulation with the V-shape spectral phase in Eq. (9), the phase modulation is given by

$$\Phi(\omega) = \Phi_V(\omega) + \pi \Theta(\omega - \omega_i),$$  \hspace{1cm} (11)

and with this expression, Eq. (7) becomes

$$S(z) \propto \left| \int_{-\infty}^{\infty} d\omega A(\omega) A(\omega_i - \omega) e^{2i\Phi_V(\omega)} e^{i(2i\omega z/c + 2i(\omega_i - \omega) z/c)} \right|^2,$$  \hspace{1cm} (12)

FIG. 1: (Color online) Spectral phase function $\Phi(\omega)$ vs. the excitation probability $|c_i|^2$ calculated from Eq. (7) for (a,b) the non-resonant two-photon transition case with $\Phi_V(\omega)$, (c-h) the resonant two-photon transition cases with (c,d) $\Phi_V(\omega) = 0$, (e,f) $\Phi_V(\omega)$, and (g,h) $\Phi_V(\omega) + \pi \Theta(\omega - \omega_i)$. The spectral amplitude $A(\omega)$ (orange dotted line) and the dispersion $f(\omega) = 1/(\omega - \omega_i)$ (green dash-dotted line) are plotted in comparison.
when we ignore the small contribution from the spectral tail part ($\omega > \omega_{\text{cs}}$) of the laser spectrum. This expression has both a transition enhancement part, which is achieved by the step phase $\pi \Theta(\omega - \omega_{\text{ig}})$, and a spatial excitation part, which contains $\Phi_\text{TR}(\omega) = \alpha |\omega - \omega_{\text{ig}}|$, in its integrand. Figure 1 shows the numerical calculation of Eq. (7) for three cases; (c,d) $\Phi(\omega) = 0$, (e,f) $\Phi_\text{TR}(\omega)$, and (g,h) $\Phi_\text{TR}(\omega) + \beta \Theta(\omega - \omega_{\text{ig}})$. As clearly shown in Fig. 1(h), the phase modulation scheme of Eq. (11) in a counterpropagating pulse excitation is expected to induce a clear spatial excitation pattern of resonant two-photon transitions.

### III. EXPERIMENTAL PROCEDURES

The experimental setup is schematically shown in Fig. 2(a). Femtosecond optical pulses were generated from a conventional Ti:sapphire laser oscillator (with a cavity length $L = 1.75$ m) mode-locked around the center frequency set to the two-photon transition frequency, $\omega_{\text{ig}} = \omega_{\text{cs}}/2$. The laser bandwidth (FWHM) was $25$ nm in wavelength scale. The pulses were then spectrally resolved into a $4f$-geometry Fourier plane and phase-modulated by a $128$-pixel spatial-light modulator (SLM) [28] to program $\Phi(\omega)$. The focal length of the $4f$-geometry was $200$ mm and the groove density of both gratings was $1200$ mm$^{-1}$ [29]. The width of each pixel in the SLM was $100$ µm and the pulse bandwidth incident into each pixel was $0.37$ nm in wavelength scale. After pulse-shaping, the pulses were focused in a rubidium vapor cell ($^{85}$Rb) located at $z = 0$, de-focused, collimated, and then reflected back by a mirror located at $z = L$, so that each counter-propagating laser pulse collided with the next pulse in the vapor cell. The focal length of both focusing lenses was $200$ mm. The beam diameter and the Rayleigh range of the focus were approximately $50$ µm and $2.5$ mm, respectively.

The $^{85}$Rb atoms were two-photon excited from the ground state $|g\rangle = 5S_{1/2}$ to the final state $|e\rangle = 5P_{3/2}$ [see Fig. 2(b)]. The corresponding frequencies (wavelengths) were $\omega_{\text{ig}}/2\pi = 384.6$ THz ($\lambda_{\text{ig}} = 780$ nm), $\omega_{\text{cs}}/2\pi = 386.6$ THz ($\lambda_{\text{cs}} = 776$ nm), and $\omega_{\text{ig}}/2\pi = 385.6$ THz ($\lambda_{\text{ig}} = 778$ nm), respectively [30]. Note that the D1 transition to the $5P_{1/2}$ state was out of the laser spectral range. The excited atoms in the $5D$ first decayed to $6P$, and then the spatial profile of the fluorescence at $420$ nm ($6P \rightarrow 5S_{1/2}$) was imaged by a CCD camera through one-to-one telescope imaging by a pair of $f = 25$ mm lenses. The image resolution of the camera was $4.54$ µm.

### IV. RESULTS AND DISCUSSION

The fluorescence images of position-independent background excitation in the resonant two-photon transition case described in Sec. II, with a dark or bright region in the center, were captured by a CCD camera and are shown in Figs. 3(a) and 3(b). In the resonant two-photon transition case, there occurs a position-independent background excitation in most of the region, that is caused by the excitation from the population remaining in the intermediate state $|i\rangle$ and which does not require the two different colors of sub-pulses simultaneously. Note that a dark region appears in $z < |z_o|$ for $\alpha < 0$ in 3(a) because the $|g\rangle \rightarrow |i\rangle$ excitation occurs later in time than $|i\rangle \rightarrow |e\rangle$. When $\alpha > 0$ in 3(b), however, a brighter region appears in $z < |z_o|$ because of the same reason but with opposite causality. In spite of this aspect, we show that spatial excitation appears in the resonant two-photon transition case, with the phase-flipping step function modulation described earlier. The retrieval of spatial excitation achieved by phase-flipping modulation in Eq. (11) is shown in Fig. 3(c).

Figure 4 shows the result of the experiment performed with the spectral phase function described in Eq. (11), with varying step height, as

$$\Phi(\omega) = \alpha |\omega - \omega_{\text{ig}}| + \beta \Theta(\omega - \omega_{\text{ig}}),$$

in comparison with a corresponding numerical calculation of the spatial excitation probability, from Eqs. (5) and (6). By maintaining the phase slope value fixed at $\alpha = -1.0$ ps, the phase step value was changed from $\beta = 0$ to $2\pi$, and the fluorescence signals (measured on the line profile along the $z$ direction where the position is shown in the horizontal axis) were plotted as a function of $\beta$ (along the vertical axis) in Fig. 4(a). As expected, sharp fluorescence peaks appear at $z = \pm z_o$, when $\beta$ is around $\pi$. The spatial excitation profiles measured at

![FIG. 2: (Color online) (a) Schematic diagram of the experimental setup. Ultrafast laser pulses were programmed by a spatial light modulator to interact with rubidium atoms in a colliding pulse geometry. The cavity length $L = 1.75$ m of the laser was matched with the extra travel of the reflected pulses. (b) The energy level diagram of atomic rubidium. Atoms are excited from $5S_{1/2}$ to $5D$ and decayed to $6P$. The fluorescence from $6P$ was monitored.](image-url)
\[\begin{align*}
\beta &= 0 \quad \text{(purple dash-dot line)} \quad \text{and} \quad 1.2\pi \quad \text{(red solid line)} \quad \text{are} \quad \text{respectively} \quad \text{shown} \quad \text{in} \quad \text{Fig.} \quad 4(c). \quad \text{This} \quad \text{experimental} \quad \text{result} \quad \text{agrees} \quad \text{well} \quad \text{with} \quad \text{the} \quad \text{numerical} \quad \text{simulation} \quad \text{as} \quad \text{shown} \quad \text{in} \quad \text{Figs.} \quad 4(b) \quad \text{and} \quad 4(d). \quad \text{Here}, \quad \text{one-photon-resonant} \quad \text{background} \quad \text{signals} \quad \text{that} \quad \text{were} \quad \text{position-insensitive} \quad \text{were} \quad \text{subtracted} \quad \text{for} \quad \text{clarity} \quad \text{by} \quad \text{means} \quad \text{of} \quad \text{numerical} \quad \text{fitting} \quad \text{with} \quad \text{the} \quad \text{focal} \quad \text{spot} \quad \text{profiles} \quad \text{of} \quad \text{counter-propagating} \quad \text{Gaussian} \quad \text{beams.}
\end{align*}\]

\[\begin{align*}
\text{FIG. 3: Photo images of the fluorescence signals:} \quad \text{(a,b) V-shape} \quad \text{spectral} \quad \text{phase} \quad \text{functions} \quad \Phi_V(\omega) \quad \text{are} \quad \text{used} \quad \text{respectively} \quad \text{with} \quad (a) \quad \alpha = -1.4 \quad \text{ps} \quad \text{and} \quad (b) \quad \alpha = +1.4 \quad \text{ps}. \quad \text{(c) The} \quad \text{spectral} \quad \text{phase} \quad \text{function} \quad \text{with} \quad \text{a} \quad \text{phase} \quad \text{step}, \quad \Phi(\omega) \quad \text{with} \quad \alpha = -1.4 \quad \text{ps}. \quad \text{[\Phi_V(\omega) \quad \text{and} \quad \Phi(\omega) \quad \text{are} \quad \text{defined} \quad \text{in} \quad \text{Eqs.} \quad (9) \quad \text{and} \quad (11), \quad \text{respectively}, \quad \text{and} \quad z_o = 420 \mu m \quad \text{in} \quad \text{(c).}].}
\end{align*}\]

\[\begin{align*}
\text{FIG. 4: (Color online) The phase-only modulation experiment:} \quad \text{(a,c) spatial} \quad \text{fluorescence} \quad \text{signals} \quad \text{for} \quad \Phi_V(\omega) \quad \text{and} \quad \text{the} \quad \text{numerical} \quad \text{simulation} \quad \text{at} \quad \text{step} \quad \text{phase} \quad \text{of} \quad 0 \quad \text{and} \quad \text{phase} \quad \text{step} \quad \text{of} \quad \text{1.2}\pi \quad \text{are} \quad \text{extracted} \quad \text{from} \quad \text{(a)} \quad \text{and} \quad \text{extracted} \quad \text{in} \quad \text{(c)} \quad \text{and} \quad \text{(d), respectively.}
\end{align*}\]

The behavior that the maximum peak signal in Fig. 4 exhibits at \(\beta = 1.2\pi\), rather than at \(\beta = \pi\), is due to the presence of the resonant amplitude term \(c_r(z)\) in Eq. (5). If we divide the non-resonant term \(c_{nr}(z)\) in Eq. (6) into \(c_{nr}^+\) and \(c_{nr}^-\), where \(c_{nr}^+\) and \(c_{nr}^-\) denote the positive and negative parts of \(c_{nr}(z)\) integrated for \(\omega > \omega_{ig}\) and \(\omega < \omega_{ig}\), respectively, the \(c_{nr}^-\) term is 90° phase-advanced and the \(c_{nr}^+\) term is 90° phase-delayed with respect to the \(c_r\) term. Figure 5 graphically depicts \(c_r\), \(c_{nr}^+\), and \(c_{nr}^-\) on the complex plane, where Fig. 5(a) corresponds to the case in which \(c_{nr}^-\) rotates by the applied step phase and the others remain unchanged. To ensure that the amplitude of the vector sum of all of the vectors reaches the maximum, the phase rotation angle has to be greater than \(\pi\) and smaller than \(1.5\pi\). This explains the step phase of \(1.2\pi\) for the maximum peak. A similar explanation applies to the case in which \(\beta\) and \(c_r\) partially rotate along with \(c_{nr}^-\), due to the fact that the size of the focal spot passing through the spatial light modulator is finite, and provides the same conclusion.

\[\begin{align*}
\text{FIG. 5: Transition amplitude change graphically depicted on the complex plane when} \quad \text{(a) } c_r, \quad \text{(b) } c_{nr}^+ + c_r, \quad \text{and} \quad \text{(c) } c_{nr}^- + c_r/2 \quad \text{is} \quad \text{phase-rotated, respectively.}
\end{align*}\]

In addition to the spectral phase modulation, we also tested spectral amplitude modulation, where the spectral components near the intermediate resonant transition \(\omega_{ig}\) were removed. In this case, we only have the non-resonant contribution \(c_{nr}(z)\) but not the resonant contribution \(c_r(z)\). For this, the resonant spectrum was blocked with a 120-µm-wide copper wire placed in the Fourier plane, which corresponds to 0.44 nm in wavelength. In this circumstance, the resonant transition from the 5S1/2 state to the 5P3/2 state is suppressed, so excitations only occur at the points where the two photons with frequencies \(\omega_{ig}\) and \(\omega_{eg} - \omega_{ig}\) respectively, collide with each other. The phase modulation of Eq. (13) from the first experiment was again applied, with \(\alpha = 1.0\) ps. As shown in Figs. 6(a) and 6(c), the phase modulation with \(\beta = \pi\) (red line) enhances and more tightly localizes the peaks than the bare \(V\)-shape phase does (light blue line). This result again compares with the theoretical prediction in Figs. 6(b) and 6(d). The maximal peak appears at \(\beta = \pi\) because of the complete removal of the resonant amplitude term.

V. CONCLUSION

In summary, we performed coherent control experiments with counter-propagating phase-modulated ultra-
short pulses for the two-photon excitations in the ladder-type three-state quantum system of atomic rubidium. The laser pulses designed on the basis of the spatial coherently control scheme to phase-flip the laser spectrum near the resonant intermediate transition, successfully produced spatially localized excitation patterns from the resonance-induced background along the focal region of the counter-propagating pulses. The resulting constructive interference phenomena among the constituent nonresonant two-photon transitions were theoretically confirmed and verified by numerical calculations. The presented scheme of coherent control has possible applications in Doppler-free frequency-comb spectroscopy of the resonant two-photon transitions. Since the phase-flipping across a particular intermediate resonance provides a two-photon transition enhancement selectively through the given intermediate state, it is hoped that this phase-programming method will be useful for atom and resonance specific laser spectroscopy.

FIG. 6: (Color online) The phase and amplitude modulation experiment: (a, c) spatial fluorescence signals for $\Phi(\omega)$ (V-shape + phase step) spectral functions with $\alpha = -1.0$ ps and (b,d) the numerical simulation. Signals at $\beta = 0$ (light blue line) and $\pi$ (red line) are extracted from (a) and (b) and shown in (c) and (d), respectively.

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