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Quantum coherent control of two-photon transitions by square phase-modulation*

Zhang Shi-An(张诗按), Wang Zu-Geng(王祖赓), and Sun Zhen-Rong(孙真荣)†

State Key Laboratory of Precision Spectroscopy,

Department of Physics, East China Normal University, Shanghai 200062, *China*

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A femtosecond laser pulse can be tailored to control the two-photon transitions using the ultra-fast pulse-shaping technique. This paper theoretically and experimentally demonstrates that two-photon transitions in molecular system with broad absorption line can be effectively controlled by square phase-modulation in frequency domain, and the influence of all parameters characterizing the square phase-modulation on two-photon transitions is systemically investigated and discussed. The obtained results have potential application in nonlinear spectroscopy and molecular physics.

Keywords: two-photon transitions, coherent control, pulse shaping
PACC: 3280K, 4255B

1. Introduction

Quantum coherent control has the ability to control the dynamics at various stages of a process as it evolves under the effect of a coherent laser source, which can be used to manipulate the light–matter interaction by interference phenomena in time or frequency domain and achieve desirable outcomes.[1−6] In the traditional approach, the coherent control can be realized through pulse-pair excitation by varying their time delay.[7−9] The recent progress in ultra-fast optics offers new opportunities for coherent control with the pulse shaping technique which manipulates the shape of a single laser pulse through the modulation of the spectral phases or amplitudes in the frequency domain.[10−12] This enables the quantum coherent control by only a single pulse excitation. Due to the great convenience and agility of the technique, it has attracted a wide attention of both experimentalists and theorists working in different areas.

Two-photon processes are a nonlinear optical effect, where two photons are simultaneously absorbed to excite a material system. For ultra-short laser pulse, due to broad band of the excitation light, the two-photon processes can be generated in all frequency components of the laser pulse as long as the sum of the two-photon energies equals the excited-state energy difference, as shown in Fig.1(a). Coherent control provides a method to adjust the quantum interferences between these different paths, and affects the rate of the whole process. In principle, the phase structure of the electric field determines the temporal order of the frequency components in the pulse and may be expected to have a profound effect on the two-photon processes. If the phase distribution can be artificially tuned, the two-photon processes can be controlled. Meshulach and Silberberg[1] firstly achieved experimentally the coherent control of the two-photon transitions by phase-only modulation in cesium gas. Then Dayan *et al*[6] experimentally demonstrated two-photon absorption and coherent control with broadband down-convert light in rubidium gas. In the atomic system, the energy level structure is comparatively simple, so two-photon processes in the experiment and theory can be well described. However, in molecular system, the two-photon processes are usually too complicated to model exactly. Meshulach and Silberberg[4] showed that the two-photon processes in the two-level molecular system with broad inhomogeneous absorption line

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†Corresponding author. E-mail: zrsun@phy.ecnu.edu.cn

depend on the intensity distribution of the exciting pulse. Therefore the two-photon transition probability in molecular system can be controlled by varying the temporal intensity distribution of the exciting pulse using the pulse-shaping technique.

The square spectral phase filter is a simple and effective method to realize coherent control and is widely applied on various nonlinear processes, such as two-photon absorption[6] and broadband frequency up-conversion.[13] In this paper, we focus on theoretically and experimentally investigating quantum coherent control of two-photon transitions in the molecular system with broad absorption line by square phase-modulation in frequency domain. By varying the parameters characterizing the square phase-modulation (modulation depth, modulation period, and initial phase), the two-photon transitions are effectively controlled. The physical mechanism for coherent control of two-photon transitions by square phase-modulation is discussed and analysed based on the temporal intensity distribution of shaped laser pulse.

2. Experiment

The layout of our experimental setup is shown in Fig.1(c). A Ti:sapphire mode-locked laser (Spectra-Physics Spitfire amplifier) is used as the excitation source with the pulse duration of about 50 fs, the repetition rate of 1 kHz, and the centre wavelength at 800 nm. The programmable 4-f pulse shaper is composed of a pair of diffraction gratings with 1200 lines/mm and a pair of concave mirrors with a 200-mm

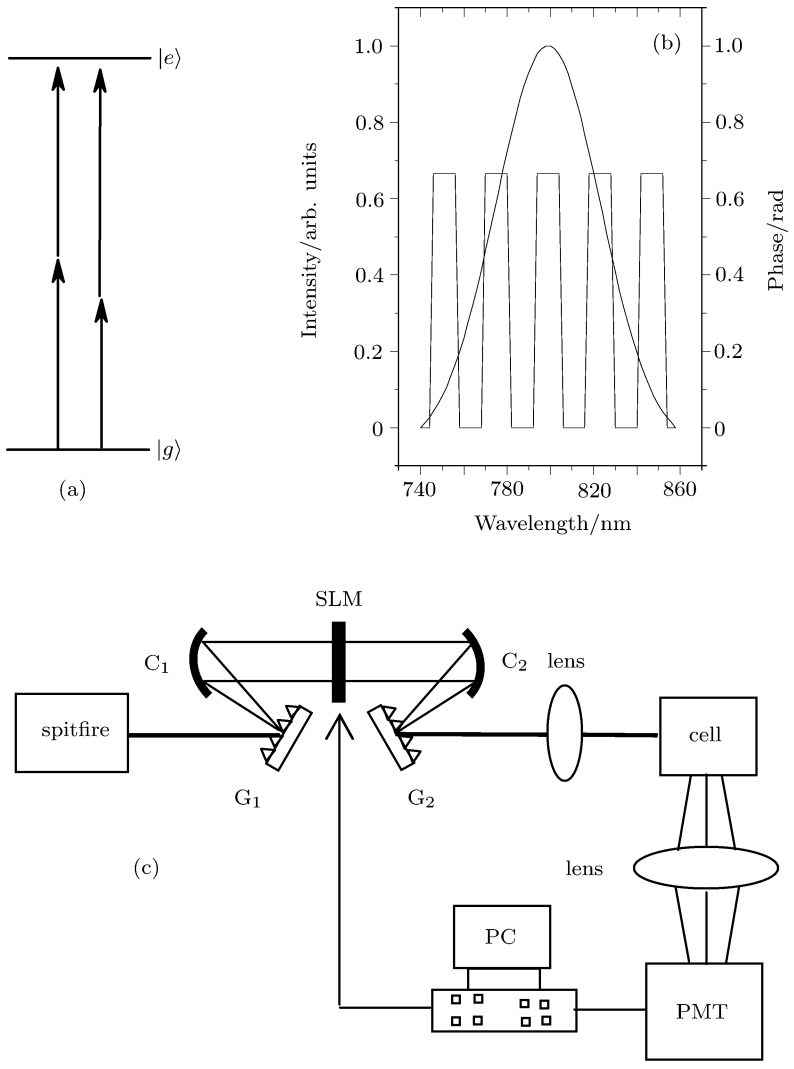


Fig.1. (a) Schematic energy-level diagram of two-photon transitions. (b) Schematic diagram of the square phase-modulation applied on laser spectrum. (c) Experimental arrangement for coherent control of two-photon transitions. The programmable liquid-crystal spatial light modulator (SLM-256) is used as an updatable filter for spectral manipulation.

focal length. A programmable one-dimensional liquid-crystal spatial light modulator array (SLM-256, CRI) is placed at the Fourier plane of the shaper and is used as an updatable filter for spectral manipulation of the incident pulses. The spatial light modulator (SLM) allows for independent control of the phase and the amplitude for each of its 128 pixels. A computer is served for updating the SLM. The intensity profile of the shaped laser pulse is measured by cross-correlating with an unshaped reference pulse.

To demonstrate coherent control of two-photon transitions in molecular system, we perform the experiment in Coumarin 515. The absorption and fluorescence spectra of Coumarin 515, together with the molecular structure, are shown in Fig.2. The maximum of the absorption band is at 410 nm, and its FWHM is about 56 nm. The central wavelength of two-photon fluorescence signal corresponds to 482 nm. The two-photon transition probabilities are directly monitored through measuring the two-photon fluorescence signal by a photomultiplier and amplified by a lock-in amplifier.

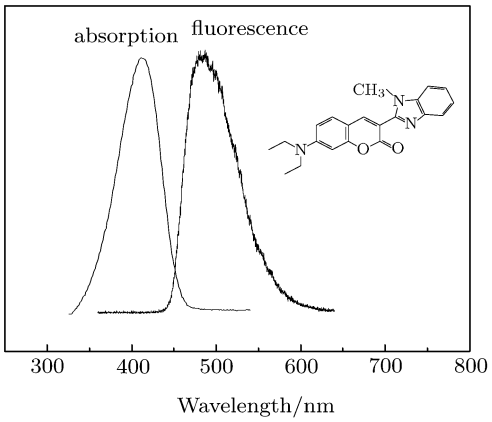


Fig.2. The absorption and fluorescence spectra of Coumarin 515, together with the molecular structure.

3. Results and discussion

Consider the two-photon interaction of an ultrashort pulse field $E(t)$ with a two-level system, as shown in Fig.1(a). In contrast to single-photon transition, all frequencies in the laser spectrum can induce two-photon transitions as long as the two-photon energy conservation is satisfied. Thus the two-photon transition probability in molecular system with broad absorption line induced by the ultra-fast laser pulse can be expressed by[4]

$$\begin{aligned} S_2 &= \int_{-\infty}^{\infty} g(\omega) \left| \int_{-\infty}^{\infty} E^2(t) \exp(\mathrm{i}\omega t) \mathrm{d}t \right|^2 \mathrm{d}\omega \\ &= \int_{-\infty}^{\infty} g(\omega) \left| \int_{-\infty}^{\infty} A(\Omega) A(\omega - \Omega) \right. \\ &\quad \left. \times \exp\left\{ \mathrm{i}\left[\Phi(\Omega) - \Phi(\omega - \Omega) \right] \right\} \mathrm{d}\Omega \right|^2 \mathrm{d}\omega \\ &= \int_{-\infty}^{\infty} I^2(t) \mathrm{d}t, \end{aligned} \tag{1}$$

where $g(\omega)$ is the line-shape function, which is related to the population distribution of the excited state, $E(\omega) = A(\omega)\exp[\mathrm{i}\Phi(\omega)]$ is the Fourier transform of $E(t)$, and $A(\omega)$ and $\Phi(\omega)$ are the spectral amplitude and the spectral phase, respectively. Eq.(1) indicates that two-photon transitions can occur in all pairs of photons with frequencies Ω and $\omega - \Omega$ (Ω and $\omega - \Omega$ lie within the laser spectrum) and depend on the temporal intensity distribution of the exciting pulse. For a given energy and power spectrum $A(\omega)$, it is easy to verify that the two-photon transition probability S_2 is maximum by the transform-limited pulse and minimum by the pulse trains with identical energy.

Since the two-photon transitions are correlated with spectral phase, we modify the spectral phase in frequency domain by square distribution (as shown in Fig.1(b)), and given by

$$\Phi(\Omega) = \frac{\alpha}{2} + \frac{2\alpha}{\pi} \sum_{l=0}^{\infty} \frac{\sin[(2l+1)(\beta\Omega + \varphi)]}{2l+1}, \tag{2}$$

where α, β and φ represent the modulation depth, modulation period and initial phase, respectively. Note that only phase-modulation does not affect the pulse energy or power spectrum. In our experiment, we study systemically the dependence of the two-photon transitions on the modulation depth α, modulation β and initial phase φ.

3.1. Coherent control by modulation depth α

Firstly, we investigate the two-photon fluorescence signal as a function of the modulation depth α. The experimental result is presented in Fig.3(a), together with the theoretical curve calculated from Eqs.(1) and (2). The theoretical curve is in good agreement with our experimental data. With the increase of α, the two-photon fluorescence signal is periodically modulated and reproduced for the transform-limited pulse at $\alpha = m\pi$ (m=1, 2, 3,$\cdots$). Further-

more, an interesting feature is that a weaker peak appears at $\alpha = (n+1)\pi/2$ (0, 1, 2,$\cdots$). Since the two-photon transition probability depends on the temporal intensity distribution of the exciting pulse, we measure the temporal intensity distribution of the shaped pulse by cross-correlating with an unshaped reference pulse. The experimental results are presented in Fig.3(b), here, we only present the pulse distribution for α=0, $\pi/4$ and $\pi/2$. Due to periodic phase modulation, the shaped pulse is a sequence of the pulses. With increase of the modulation depth, the shaped pulse also shows a periodic evolution. At $\alpha = m\pi$ (m=1, 2, 3,$\cdots$), the shaped pulse reconstructs the transform-limited pulse. At $\alpha = (n+1)\pi/2$ (0, 1, 2,$\cdots$), the energy of the central pulse in the pulse train is zero and the energy of the side pulses is maximum. According to above theory, now it is easy to understand that the intense peaks arise from reconstructing the transform-limited pulse at $\alpha = m\pi$ (m=1, 2, 3,$\cdots$), and the weak peaks arise from the energy of the central pulse in the train entirely transferred into the side pulses at $\alpha = (n+1)\pi/2$ (0, 1, 2,$\cdots$).

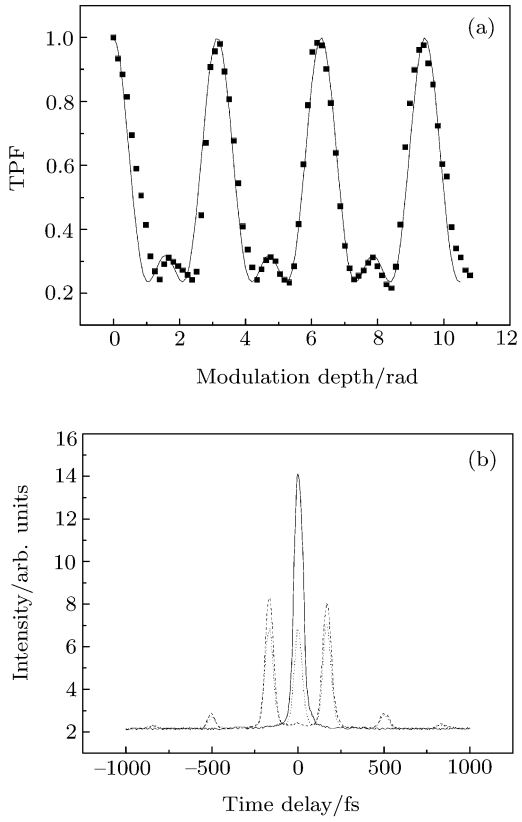


Fig.3. (a) Experimental (square) and calculated (line) two photon fluorescence signal as function of the modulation depth α for square phase-modulation, β=8, φ=0. (b) The experimental temporal intensity distribution of the shaped pulse for α=0 (solid line), $\pi/4$ (dot line), and $\pi/2$ (dash line).

3.2. Coherent control by modulation period β

In this section, we investigate the two-photon fluorescence signal as a function of the modulation period β. The experimental result is presented in Fig.4(a), together with the theoretical curve. We can observe that the two-photon fluorescence signal shows an initial fast following by a slow decay with the increase of β. Similarly, we also measure the temporal intensity distribution of the shaped laser pulse. Here, we only present the pulse distribution for β=0, 8, and 16, as shown in Fig.4(b). We note that, with the increase of β, the separation between two pulses in the pulse train increases, the energy of the central pulse decreases and the energy of side pulses increases. So it can be deduced that the evolution process of two-photon fluorescence signal with the increase of β results from the energy of the central pulse in the pulse train gradually transferred into the side pulses since the two-photon transition probability is minimum by the pulse trains with identical energy.

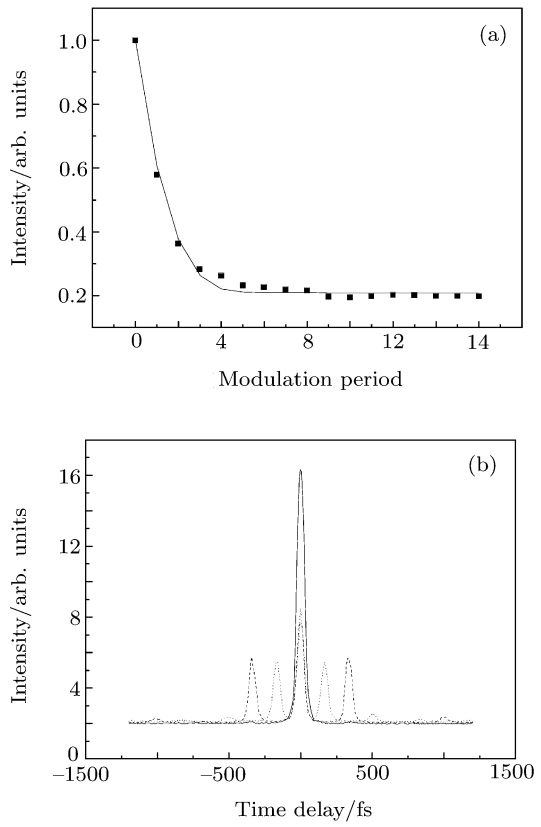


Fig.4. (a) Experimental (square) and calculated (line) two photon fluorescence signal as function of the modulation period β for square phase modulation, α=8, φ=0. (b) The experimental temporal intensity distribution of the shaped pulse for β=0 (solid line), 8 (dot line), and 16 (dash line).

3.3. Coherent control by initial phase φ

To emphasize the importance of initial phase, finally we investigate the two-photon fluorescence signal as a function of the initial phase φ. The experimental and theoretical results are presented in Fig.5(a). The two-photon fluorescence signal is minimum at $\varphi = \pi$. In our experiment, the two-photon fluorescence signal does not completely vanish at certain initial phase φ, which is different from the observation in gas with narrow absorption line.[4] The phenomenon can arise from the fact that part of the photons in the exciting pulses always satisfies with the condition of the two-photon transitions in the molecular system with broad absorption line when φ is increased. The temporal intensity distribution of the shaped laser pulse for φ=0 and π is shown in Fig.5(b). We can find that the energy of the central pulses decreases and the energy of side pulse increases for $\varphi = \pi$. So we reasonably believe that the decrease of the two-photon fluorescence signal at $\varphi = \pi$ also results from the energy of the central pulse in the pulse train transferred into the side pulses.

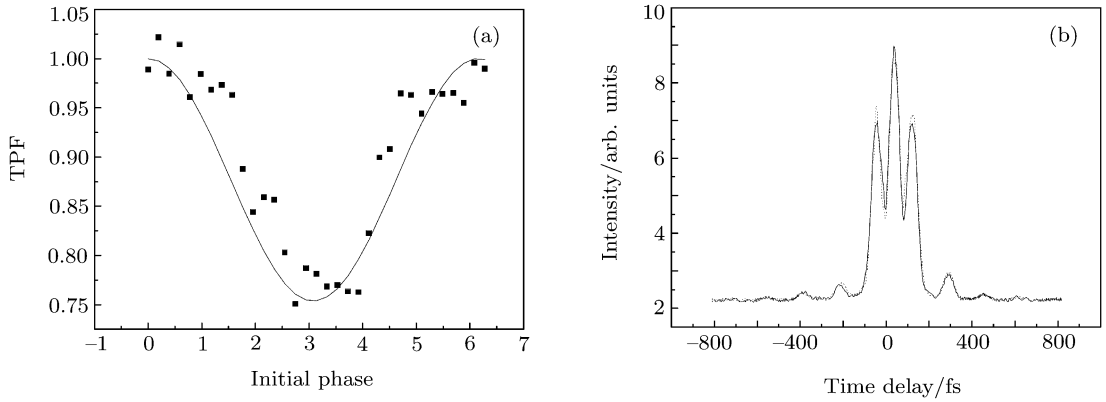


Fig.5. (a) Experimental (square) and calculated (line) two photon fluorescence signal as function of the initial phase φ for square phase-modulation, α=8, β=4. (b) The experimental temporal intensity distribution of the shaped pulse for φ=0 (solid line) and π (dot line).

4. Conclusion

In summary, we have theoretically and experimentally investigated the quantum coherent control of two-photon transitions in the molecular systems with broad absorption line by square phase-modulation in frequency domain. Experimental results and theoretical calculations indicate that the two-photon transition probability depends on the temporal intensity distribution of the exciting pulse. By varying the parameters characterizing the square phase-modulation (modulation depth, modulation period, and initial phase), the two-photon transitions are effectively controlled, which are realized by the energy transfer between the central pulse in the pulse train and the side pulses. Our research will be very useful in nonlinear spectroscopy and molecular physics.

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