Coherent control of two-photon transitions in a two-level system with broadband absorption

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Multiphoton transition probability can be manipulated by tailoring the ultrashort laser pulses. In this paper, we theoretically and experimentally investigate the influence of the absorption bandwidth and line shape on the coherent control of two-photon transitions in two-level system by manipulating the femtosecond laser pulse with the π spectral phase step. A coherent feature with a coherent peak and two coherent dips can be achieved if the absorption bandwidth is less than twice of the laser spectral bandwidth. The absorption bandwidth decides the coherent peak or dip intensity, and the absorption line shape affects the symmetry of the two coherent dips.

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Coherent control strategies by tailoring ultrashort laser pulses are tremendously successful to manipulate the physical and chemical processes and properties. With the development in femtosecond pulse-shaping technique, it is possible to obtain such a pulse with almost arbitrary temporal distribution by manipulating the spectral phase and/or amplitude in frequency domain [1–3]. For a simple quantum system, the predesigned pulse is easy to meet the experimental requirement [4–15]. For the complex case, the optimal feedback control based on learning algorithm is a rather perfect method to optimize the light field without any prior knowledge about the quantum system and achieve the desired outcomes [16–24]. So far, the coherent control by shaping femtosecond laser pulses has been widely applied in nonlinear optics, chemical reaction, material processing and quantum computing, and these various experiments are realized by open-loop or closed-loop schemes.

Two-photon transitions are a third-order nonlinear physical process, where two photons are simultaneously absorbed to excite a quantum system. For the broadband laser pulses, the two-photon transitions can occur in all frequency components of the laser pulses as long as the sum of the two-photon energies is equal to the energy difference between the excited state and the ground state. In previous study, it was indicated that the two or multiple-photon transition probability of the Rydberg atoms could be manipulated based on the two-photon Rabi oscillations with the microwave field [25,26]. Recently, the coherent control based on femtosecond pulse-shaping technique has been widely employed to manipulate the two-photon transitions by the quantum interference between these distinct competing routes and finally affect the total two-photon transition probability. The use of femtosecond laser pulses with the simple spectral phase pattern of sine, cosine, chirp or step-function shape to control the two-photon transitions has been reported [4–9]. Furthermore, the optimal feedback control based on learning algorithm has also been utilized to maximize the two-photon transitions [15–17].

The femtosecond laser pulses with the π spectral phase step are usually used to control two-photon transitions [5,6,9], and as well as may provide a feasible tool to investigate the coherence role in two-photon transitions. Silberberg *et al.* has demonstrated that, when the π phase step position is at the laser center frequency (i.e., the spectral phase structure is antisymmetric), two-photon transition probability in two-level atomic system with the very narrow absorption band (the narrow band limit) can be almost reconstructed as that for the transform limited pulses, but this coherent feature in molecular system with very broad absorption band (the broad band limit) will cancel out [5]. However, what and how to affect the coherence of two-photon transitions with the broad absorption band, are still not known by now. In this paper, we theoretically and experimentally study the influence of the absorption band on the coherent control of two-photon transitions in the two-level system by manipulating femtosecond pulses with the π spectral phase step, including of the absorption bandwidth and line shape. A coherent feature with a coherent peak and two coherent dips can be observed if the absorption bandwidth is less than twice of laser spectral bandwidth, and the dependence of the coherent feature on the absorption bandwidth and line shape will be detailedly discussed and analyzed in the context.

The two-photon resonant interaction of a laser field $E(t)$ with a two-level system is shown in Fig. 1(a), here $|g_i\rangle$ and $|f_i\rangle$ represent the ground and excited states, respectively. Assuming that the population is initially in the ground state and the femtosecond excitation laser pulse is much shorter than the lifetime of the excited state, and the two-photon transition probability in the two-level system induced by ultrashort laser pulse can be approximated by [5]

$$S_2 = \int_{-\infty}^{\infty} G(\omega_a) \left| \int_{-\infty}^{\infty} E^2(t) \exp(i\omega_a t) dt \right|^2 d\omega_a$$
$$= \int_{-\infty}^{\infty} G(\omega_a) \left| \int_{-\infty}^{\infty} E(\omega) E(\omega_a - \omega) d\omega \right|^2 d\omega_a,$$

where $E(\omega)=A(\omega)\exp[i\Phi(\omega)]$ is the Fourier transform of $E(t)$, $G(\omega_a)$ represents the distribution of the absorption band, and ω_a is the transition frequency from the ground state to the excited state. As can be seen from the above-mentioned equation, the two-photon transitions can occur in

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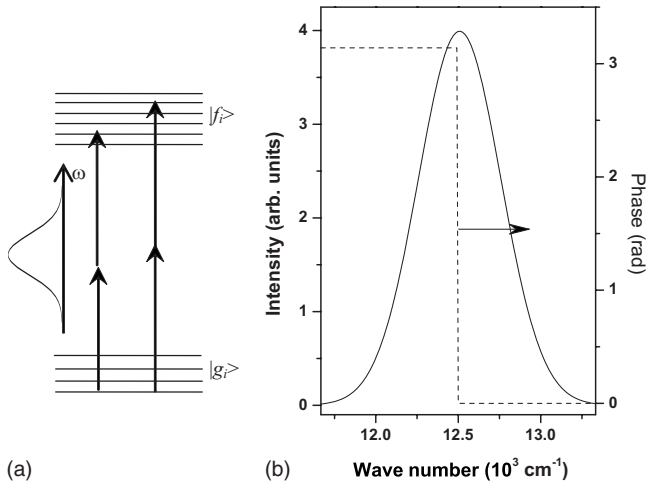

FIG. 1. (a) The schematic energy-level diagram of two-photon transitions in two-level system, the population is initially in the ground state. (b) The schematic diagram of a π spectral phase step applied on the femtosecond laser spectrum.

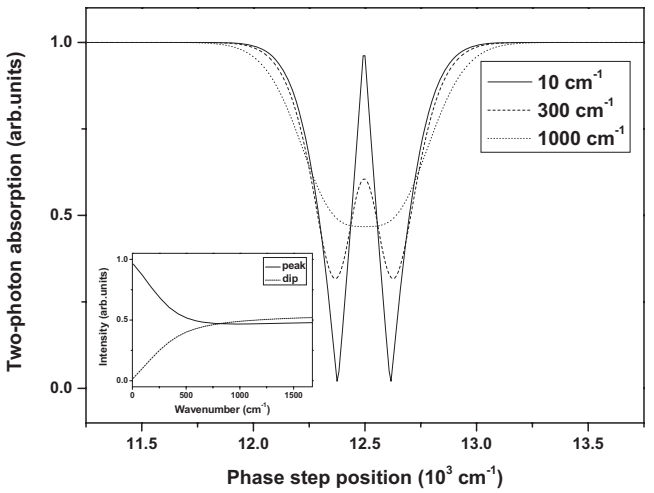

FIG. 2. The calculated results of two-photon transitions in two-level system for Gaussian absorption with bandwidth of 10 cm^{-1} (solid line), 300 cm^{-1} (dashed line) and 1000 cm^{-1} (dotted line). Inset shows the coherent peak (solid line) and dip (dashed line) intensities as the function of the absorption bandwidth.

all pairs of the photons with the frequencies ω and ω_a-ω (ω and ω_a-ω lie within the laser spectrum), and the S_2 is proportional to the sum of all individual transitions. For a given energy and power spectrum, it can be verified that, S_2 is the maximum for the transform limited pulse $\Phi(\omega)=\Phi(\omega_a\text{-}\omega)=0$, and will be reduced but not be enhanced for the other shaped pulse. The manipulation of S_2 has been widely investigated by the spectral phase modulation or closed-loop optimization [5–9,15–17]. In this paper, we focus on investigating the influence of the absorption band $G(\omega_a)$ on S_2 by manipulating femtosecond laser pulses with the π spectral phase step, including of the absorption bandwidth and lineshape, and the simple spectral phase modulation is shown in Fig. 1(b).

Considering the simple and common absorption band with the Gaussian shape, i.e., $G(\omega_a)=A_0\exp[-2(\omega_a-\omega_0)^2/\Delta\omega^2]$, where ω_0 is the absorption central frequency and $\Delta\omega$ represents the absorption bandwidth. Assuming that the absorption central frequency is 25 000 cm^{-1} and the laser central frequency is 12 500 cm^{-1} with the bandwidth [full width at half maximum (FWHM)] of 500 cm^{-1}. We first demonstrate the influence of the absorption bandwidth on two-photon transitions pumped by the femtosecond pulses with the π spectral phase step, and the calculated results are shown in Fig. 2 for the absorption bandwidth of 10 cm^{-1} (solid line), 300 cm^{-1} (dashed line) and 1000 cm^{-1} (dotted line). These results are normalized by the transform limited pulse excitation. As can be seen from Fig. 2, the two-photon transition probability is almost eliminated at the π phase step position of about 12 375 cm^{-1} and 12 625 cm^{-1} and reconstructed as that for the transform limited pulse at the π phase step position of 12 500 cm^{-1} for the very narrow absorption band (solid line), but this coherent feature will vanish for the very broad absorption band (dotted line). The disappearance of the coherent feature depends not only on the absorption bandwidth but also on the laser spectral bandwidth. If the absorption bandwidth is more than twice of the laser spectral bandwidth, the coherent feature will cancel out. The intensities of the coherent peak (solid line) and dip (dashed line) as function of the absorption bandwidth are shown in the inset of Fig. 2. The coherent peak shows a slow decrease process but the coherent dip shows a slow increase process with an increase of the absorption bandwidth, and finally both will approach to a constant.

For the symmetric Gaussian absorption line shape, the two coherent dips show the symmetry with the longitudinal axis at the coherent peak (as shown in Fig. 2). However, this symmetry will be destroyed for the asymmetrical absorption line shape (as shown in Fig. 3). The left dip is lower than the right one (solid line) for the absorption line shape with the distribution of the red components (the low frequency components) more intense than the blue components (the high

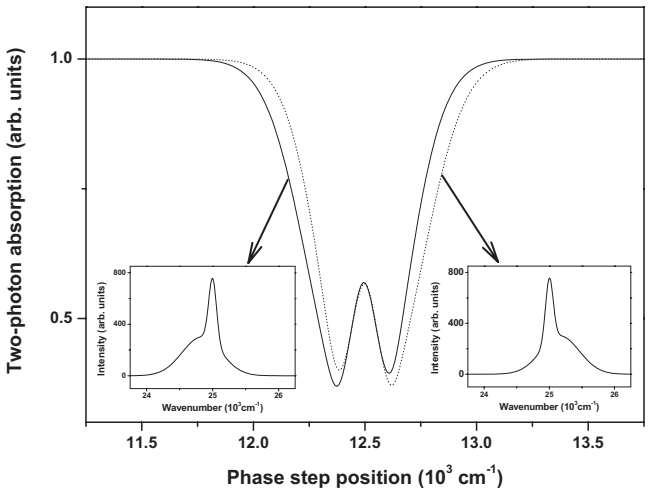

FIG. 3. The calculated results of two-photon transitions in two-level system with asymmetrical absorption line shape of the red frequency components more intense than the blue components (solid line) and the blue frequency components more intense than the red components (dotted line).

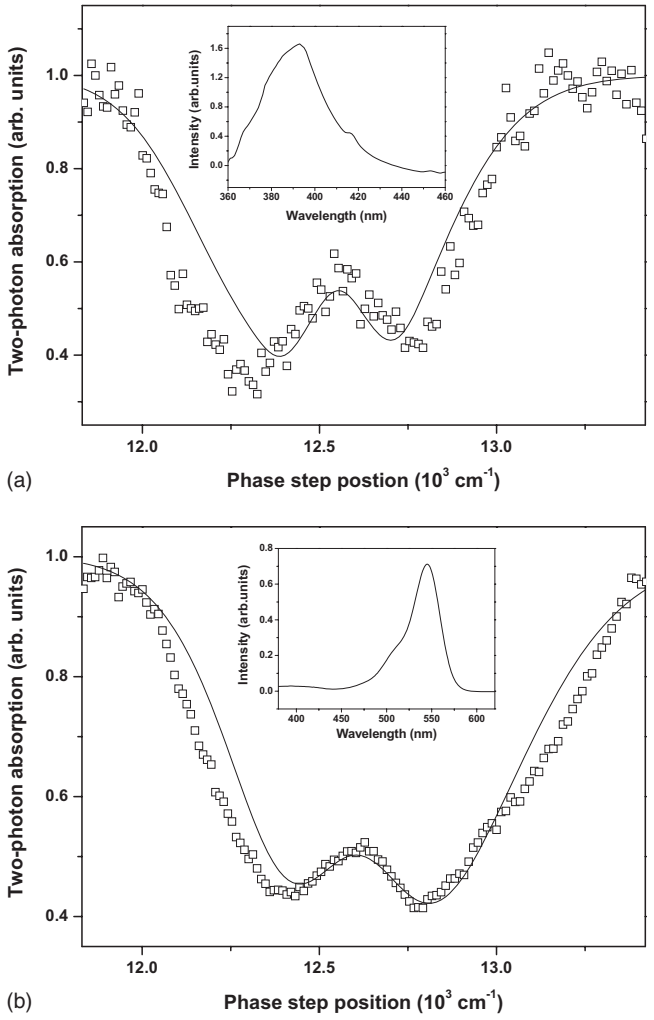

FIG. 4. The experimental (square) and theoretical (solid) results for two-photon transitions in the Er-doped glass (a) and Rhodamine B (b) pumped by the femtosecond pulses with the π spectral phase step.

frequency components), and the right dip is lower than the left one (dotted line) for the absorption line shape with the distribution of the blue components more intense than the red components. Moreover, the intensity ratio between the two coherent dips depends on the asymmetrical degree of the absorption line shape, and the greater asymmetry results in the larger intensity ratio.

In order to validate the influence of the absorption bandwidth and line shape on the coherent control of two-photon transitions with the π spectral phase step, we perform the experiment in Er-doped glass and Rhodamine B with different absorption bandwidth and line shape [as shown in inset of Figs. 4(a) and 4(b)]. The absorption band for Er-doped glass has the shape with the blue components more intense than the red components and the bandwidth (FWHM) of about 26 nm. However, the absorption band for Rhodamine B has the shape with the red components more intense than the blue components and the bandwidth (FWHM) of about 53 nm. In our experiment, a mode-locked Ti:sapphire amplifier is tuned to output about 50 fs laser pulses with the center wavelength at 795 nm and the spectral bandwidth (FWHM) of about 32 nm. The programmable 4 f pulse shaper is composed of a pair of diffraction grating of 1200 lines/mm and a pair of concave mirror with 200 mm focal length. A programmable one-dimensional liquid-crystal spatial light modulator (SLM-256, CRI) is placed at the Fourier plane of the shaper and used as an updatable filter for the π spectral phase step. The two-photon transitions are directly detected through measuring the two-photon fluorescence (TPF) signal by a photomultiplier (PMT) and lock-in amplifier.

The temporal electric filed of the π-step modulated pulse is double half-pulse structure. In the experiment, we apply appropriate voltages on the SLM to induce a spectral phase step in the laser spectrum, and the intensity profile of the shaped pulse is measured by cross correlating with an unshaped reference pulse. If the two subpulses have same intensity, it indicates that the output pulse is modulated by the π spectral phase step. The TPF signal of Er-doped glass and Rhodamine B as function of the π spectral phase step are measured and shown in Fig. 4, together with the theoretical calculation according to their corresponding absorption spectra. The absorption bandwidth of Rhodamine B is larger than that of Er-doped glass, so the coherent peak for Rhodamine B is lower than that for Er-doped glass. Moreover, due to the asymmetrical absorption band, the left coherent dip is lower than the right one for Er-doped glass and the right coherent dip is lower than the left one for Rhodamine B. It is obvious that the experimental results are excellent agreement with theoretical calculation. Furthermore, in Silberberg's experiment [5], the coherent feature in Coumarin 6H is not observed, and the reason is that its molecular absorption bandwidth of 56 nm is much more than twice of the laser spectral bandwidth of 14 nm. Therefore, it is crucial to select a suitable laser spectral bandwidth in order to achieve the coherent feature of two-photon transitions by the π spectral phase step.

In summary, we have demonstrated the effects of the absorption bandwidth and lineshape on the coherent control of two-photon transitions in two-level system pumped by the femtosecond pulses with the π spectral phase step. The theoretical calculation indicates that, a coherent feature with a coherent peak and two coherent dips can be obtained when the absorption bandwidth is within twice of the laser spectral bandwidth, and the coherent peak or dip intensity depends on the absorption bandwidth and the symmetry of the two coherent dips is correlated with the absorption line shape. Finally, the theoretical results are experimentally confirmed in Er-doped glass and Rhodamine B systems with different absorption bandwidth and line shape.

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