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NON-RESONANT TWO-PHOTON ABSORPTION CONTROL BY TWO TIME-DELAYED LASER PULSES

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In this paper, we theoretically investigate the control of the non-resonant two-photon absorption induced by two time-delayed laser pulses, and an analytical solution for the dependence of the two-photon transition probability on the time delay and relative carrier-envelope phase of the two laser pulses is achieved. We show that the two-photon absorption can be significantly enhanced or completely suppressed by varying the time delay between the two laser pulses or their relative carrier-envelope phase. We also show that the two-photon absorption can be selectively excited when two excited states are simultaneously excited. Furthermore, we discuss the two-photon absorption control in the molecular system and analyze the effect of the absorption bandwidth on the control efficiency of the two-photon absorption.

Keywords: Two-photon absorption; coherent control.

1. Introduction

Non-resonant two-photon absorption is a typical third-order optical process, where the two photons of identical or different frequencies are simultaneously absorbed to excite an atom or molecule from one state (usually the ground state) to a higher excited state. Comparing to one-photon absorption, the two-photon absorption has some important advantages, which can provide a method to activate chemical or physical processes with high spatial resolution in three dimensions, and therefore has been successfully applied in three-dimensional imaging,[1] microscopy,[2,3]

microfabrication,[4] three-dimensional data-storage,[5,6] optical power limiting,[7] and photodynamic therapy,[8] and so on.

Recently, coherent control strategy, which is dominant by the quantum constructive or destructive interference of different excitation pathways connecting the initial and final states, has attracted considerable attention because of its ability to manipulate the physical and chemical processes by light-matter nonlinear interaction.[9,10] As a typical nonlinear optical effect, the coherent control of the non-resonant two-photon absorption in atomic and molecular systems has been widely investigated in theory and experiment.[11−25] Femtosecond pulse shaping technique by modulating the spectral phase, amplitude and polarization in the frequency domain has proven to be effective method to control the two-photon absorption,[11−23] but the two-photon absorption by the shaped laser pulse can be suppressed but not enhanced. Recent study by Präkelt *et al.* showed that the two time-delay laser pulses are a well-established tool to control the two-photon absorption, which can be enhanced and also suppressed.[25] However, in their study only the effect of the time delay between the two laser pulses on the two-photon absorption was considered. In this paper, we further simplify and extend these theoretical works of non-resonant two-photon absorption control by the two time-delayed laser pulses, and obtain an analytical solution for the dependence of the two-photon transition probability on the time delay and relative carrier-envelope phase of the two laser pulses. Our results indicate that the two-photon absorption can be effectively controlled by varying the time delay between the two laser pulses or their relative carrier-envelope phase, which can be significantly enhanced or completely suppressed, and the two-photon absorption between the two excited states can be selectively excited. Furthermore, we extend our theoretical works to the molecular system, and show that the molecular absorption bandwidth will decrease the control efficiency of the two-photon absorption.

2. Theoretical Model

We consider the nonlinear interaction of two time-delayed laser pulses with a two-level system in the atomic system, as shown in Fig. 1. Here, $|g\rangle$ and $|f\rangle$ are the ground state and final excited state, and $|g\rangle \rightarrow |f\rangle$ is coupled by non-resonant two-photon absorption. $E_A(t)$ and $E_A(t-T_d)$ are the two time-delayed laser pulses, and their combined electric field $E(t)$ is expressed by

$$\begin{aligned} E(t) &= E_A(t) + E_A(t - T_d) \\ &= E_{A0}(t)\exp(-i\omega t) + E_{A0}(t - T_d)\exp[-i(\omega(t - T_d) + \Phi_{\mathrm{CEP}})], \end{aligned} \quad (2.1)$$

where T_d is the time delay between the two laser pulses, Φ_{CEP} is their relative carrier-envelope phase, and $E_{A0}(t)$ is the envelope of the laser pulse with $E_{A0}(t) = E_0\exp(-2\ln 2t^2/\tau^2)$, here E_0 is the laser amplitude and τ is the pulse duration. In the weak-field regime, the two-photon transition probability S_2 from

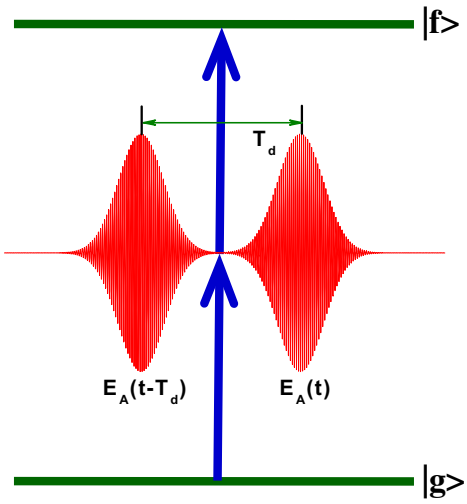

Fig. 1. (Color online) The schematic diagram of the non-resonant two-photon absorption induced by two time-delayed laser pulses $E_A(t)$ and $E_A(t - T_d)$.

the ground state $|g\rangle$ to the final excited state $|f\rangle$ can be described by second-order time-dependent perturbation theory, and is approximated by[11,12]

$$S_2 \propto \left| \int_{-\infty}^{+\infty} E^2(t) \exp(i\omega t) dt \right|^2, \tag{2.2}$$

where ω_{fg} is the transition frequency from the ground state $|g\rangle$ to the final excited state $|f\rangle$. Substituting Eq. (2.1) into Eq. (2.2), the two-photon transition probability S_2 can be written as

$$S_2 \propto |a_{\rm opt}(T_d) + a_{qm}(T_d)|^2, \tag{2.3}$$

with

$$a_{\rm opt}(T_d) = 2\exp(-T_d^2/2\tau^2)\exp[-i(\omega_{fg}T_d/2 + \Phi_{\rm CEP})], \tag{2.4}$$

and

$$a_{qm}(T_d) = 1 + \exp[-i(\omega_{fg}T_d + 2\Phi_{\rm CEP})], \tag{2.5}$$

where $a_{\rm opt}(T_d)$ and $a_{qm}(T_d)$ represent the optical and the quantum-mechanical contributions, respectively. One can see that the two-photon absorption probability S_2 depends on the time delay T_d and the carrier-envelop phase $\Phi_{\rm CEP}$. It is easy to be verified from Eq. (2.4) that the optical contribution $a_{\rm opt}(T_d)$ will disappear when the two time-delayed laser pulses are completely separated, thus Eq. (2.3) can be further simplified for different ranges of the time delay, and is written as

$$S_2(T_d \ll \tau) \propto 4 \times [1 + \cos(\omega_{fg}T_d/2 + \Phi_{\rm CEP})]^2, \tag{2.6}$$

$$S_2(T_d \gg \tau) \propto 2 \times [1 + \cos(\omega_{fg}T_d + 2\Phi_{\rm CEP})], \tag{2.7}$$

one can see from Eqs. (2.6) and (2.7) that the two-photon transition probability S_2 is periodically modulated by varying the time delay T_d and the carrier-envelope phase $\Phi_{\rm CEP}$, and the modulation periods are $2/\omega_{fg}$ and 2π for small time delay (i.e., $T_d \ll \tau$) and $1/\omega_{fg}$ and π for large time delay (i.e., $T_d \gg \tau$).

3. Results and Discussion

In our simulation, the parameters of the quantum system in Fig. 1 are set as follows. The transition frequency from the ground state $|g\rangle$ to the final excited state $|f\rangle\omega_{fg}$ is 25,000 $\rm cm^{-1}$, and the central frequency of the laser field is set to be $\omega_L = \omega_{fg}/2 = 12,500\,\rm cm^{-1}$ with the laser pulse duration τ [full width at half maximum (FWHM)] of 50 fs. Figure 2(a) shows the two-photon transition probability S_2 as the function of the time delay T_d for the carrier-envelop phase $\Phi_{\rm CEP} = 0$. The data are normalized so that a single laser pulse generates a unity signal. As can be seen, the two-photon transition probability S_2 is periodically modulated with the increase of time delay T_d, and the modulation period is different for small and large time delays. The modulation period is 2.66 fs around the small time delay (i.e., $T_d \ll \tau$), corresponding to $2/\omega_{fg}$ [see Fig. 2(b)], and is 1.33 fs around large time delay (i.e., $T_d \gg \tau$), corresponding to $1/\omega_{fg}$ [see Fig. 2(c)], which is consistent with above theoretical expectation. Since the modulation period is correlated with the transition frequency ω_{fg}, it can provide a feasible method to study the atomic or molecular excited state structure. This modulation of the two-photon transition

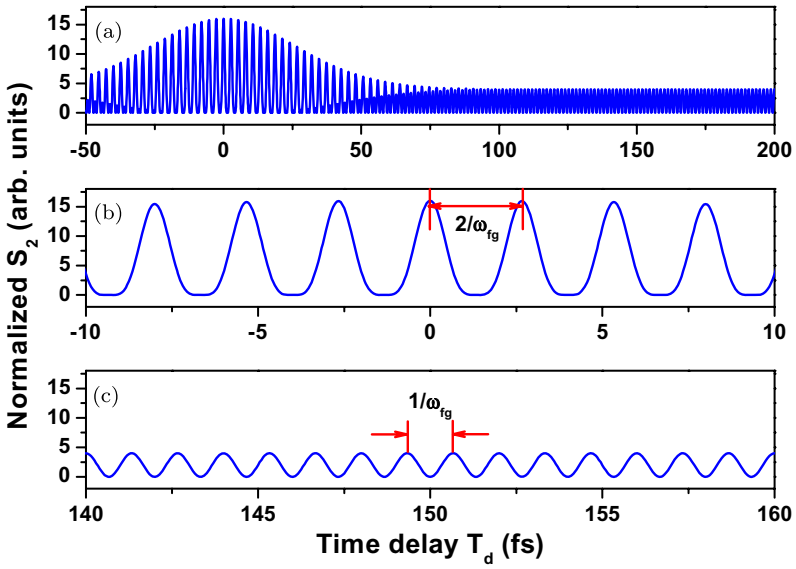

Fig. 2. (Color online) (a) The two-photon transition probability S_2 as the function of the time delay T_d for the carrier-envelope phase $\Phi_{\rm CEP} = 0$. (b) and (c) show the two-photon transition probability S_2 around small time delay (i.e., $T_d \ll \tau$) and large time delay (i.e., $T_d \gg \tau$), respectively.

probability S_2 can be intuitively explained as follows. The two laser pulses are overlapped at small time delay and therefore the optical contribution $a_{\rm opt}(T_d)$ is dominant due to the optical interference, while the optical contribution $a_{\rm opt}(T_d)$ will disappear when the two laser pulses are completely separated at large time delay, and so only quantum-mechanical contribution $a_{qm}(T_d)$ occurs. Furthermore, it can be seen that the two-photon transition probability S_2 can be significantly enhanced or completely suppressed by controlling the time delay T_d. That is to say, the two-photon absorption induced by a laser pulse can be enhanced or suppressed by applying another time-delayed laser pulse.

To demonstrate the important role of the carrier-envelope phase $\Phi_{\rm CEP}$ on the two-photon absorption, we present the two-photon transition probability S_2 as the function of the carrier-envelope phase $\Phi_{\rm CEP}$ for small and large time delays, as shown in Fig. 3. As can be seen, with the increase of carrier-envelope phase $\Phi_{\rm CEP}$, the two-photon transition probability S_2 is also periodically modulated, and the modulation period is 2π and π for small and large time delays, respectively. This result is also in good agreement with above theoretical prediction. Moreover, it can be found that the two-photon transition probability S_2 can be significantly enhanced or completely suppressed at any time delay T_d. In other words, the two-photon transition probability S_2 can be effectively controlled at any time delay T_d by varying the carrier-envelope phase $\Phi_{\rm CEP}$ or at any carrier-envelope phase $\Phi_{\rm CEP}$ by varying the time delay T_d. So, it can be concluded that the two-photon transition

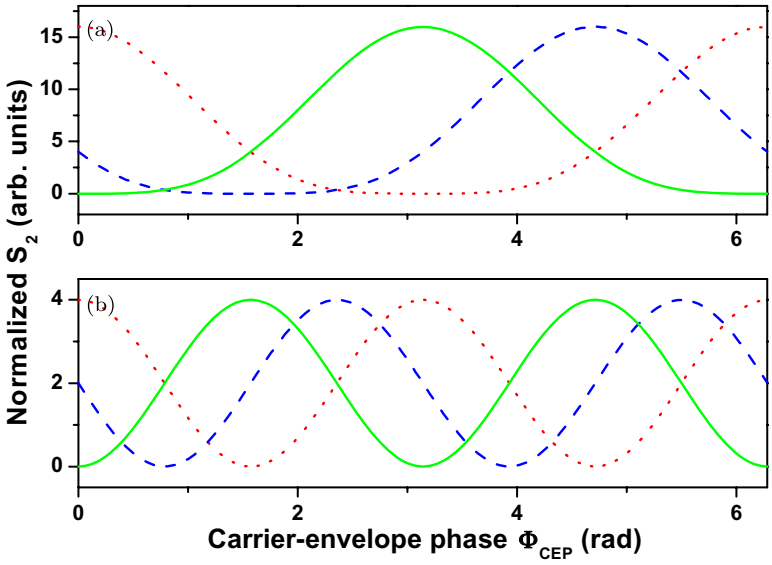

Fig. 3. (Color online) The two-photon transition probability S_2 as the function of the carrier-envelope phase $\Phi_{\rm CEP}$ for the time delays: (a) $T_d = 2.66$ (red dotted line), 3.325 (blue dashed line) and 3.99 fs (green solid line), and (b) $T_d = 146.8$ (red dotted line), 147.1 (blue dashed line) and 147.43 fs (green solid line).

probability S_2 depends on not only the time delay between the two laser pulses T_d, but also their relative carrier-envelope phase Φ_{CEP}.

As shown in Fig. 2, the modulation period of the two-photon transition probability S_2 depends on the transition frequency ω_{fg}, and therefore it can provide a feasible scheme to realize the selective excitation of the multiple excited states since the modulation periods are different for different excited states. We consider the quantum system including two excited states $|f_1\rangle$ and $|f_2\rangle$ with the transition frequencies of 24,500 and 25,500 cm^{-1}. Figure 4 shows the two-photon transition probability S_2 of the excited states $|f_1\rangle$ (a) and $|f_2\rangle$ (b) as the function of the time delay T_d with the carrier-envelope phase $\Phi_{\mathrm{CEP}} = 0$. One can see that the modulation periods of the two-photon transition probability S_2 for the two excited states $|f_1\rangle$ and $|f_2\rangle$ are different due to the quantum-mechanical contribution $a_{qm}(T_d)$. The two excited states $|f_1\rangle$ and $|f_2\rangle$ are simultaneously enhanced (or suppressed) around the time delay $T_d = 133.4\,\mathrm{fs}$, and the excited state $|f_1\rangle$ is enhanced (or suppressed) while the excited state $|f_2\rangle$ is suppressed (or enhanced) around the time delay $T_d = 116.4\,\mathrm{fs}$. Obviously, the two time-delayed laser pulses are a well-established tool to realize the selective excitation of the two-photon absorption.

Finally, we consider the other case of non-resonant two-photon absorption in molecular system with a broad absorption line. The two-photon absorption is proportional to the sum of each individual transition, and so can be written as

$$S_2 \propto \int_0^\infty A(\omega_{fg})|a_{\mathrm{opt}}(T_d) + a_{qm}(T_d)|^2 d\omega_{fg}, \tag{3.1}$$

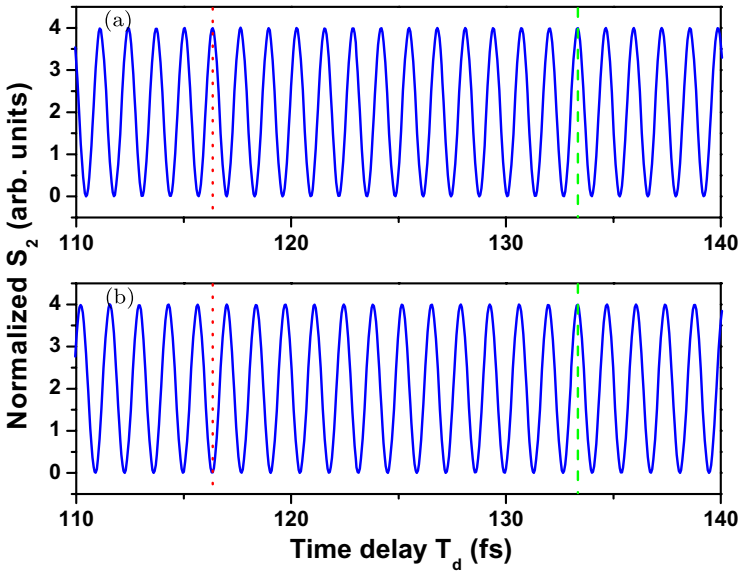

Fig. 4. (Color online) The two-photon transition probability S_2 as the function of the time delay T_d for the transition frequencies $\omega_{fg} = 24{,}500$ (a) and 25,500 cm^{-1} (b).

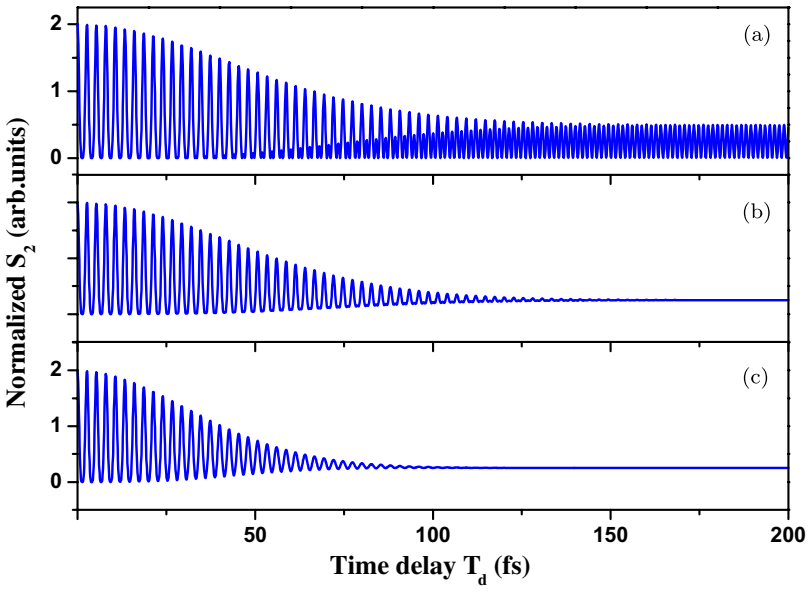

Fig. 5. (Color online) The two-photon transition probability S_2 as the function of the time delay T_d for the two-photon absorption bandwidth of 10 (a), 200 (b), and 500 cm^{-1} (c).

where $A(\omega_{fg})$ is the two-photon absorption line-shape function. We consider the simple and common molecular absorption with Gaussian shape, i.e., $A(\omega_{fg}) = A_0 \exp[-2(\omega_{fg} - \omega_0)^2/\Delta\omega^2]$, here ω_0 is the absorption central frequency and $\Delta\omega$ is the absorption bandwidth. Figure 5 presents the two-photon transition probability S_2 as the function of time delay T_d for the absorption bandwidth $\Delta\omega$ of 10 (a), 200 (b), and 500 cm^{-1} (c). As can be seen, the two-photon transition probability S_2 is periodically modulated for the very narrow absorption bandwidth [see Fig. 5(a)], this observation is the same as that obtained in Fig. 2, but the periodical modulation at large time delay will disappear for the broad absorption bandwidth [see Figs. 5(b) and 5(c)], and the two-photon transition probability S_2 become a constant. It can be found that the beginning time delay T_d that this periodic modulation disappears is decreasing with the increase of the absorption bandwidth. Since the two-photon absorption in the molecular system is the sum of each individual transition and the modulation period of each transition probability depends on its transition frequency ω_{fg}, always half transition probabilities are coherent enhancement while the other half are coherent suppression at a large time delay when the molecular absorption bandwidth is enough broad, and therefore their total transition probability will be constant. We theoretically find that this periodical modulation due to the quantum-mechanical contribution depends on both the molecular absorption bandwidth and laser spectral bandwidth, and it will cancel out when the molecular absorption bandwidth is more than twice of the laser spectral bandwidth.

4. Conclusion

In conclusion, we have presented a theoretical study on the non-resonant two-photon absorption control induced by two time-delayed laser pulses, and the dependence of the two-photon transition probability on the time delay and relative carrier-envelope phase of the two laser pulses was analytically obtained. It was showed that the two-photon absorption created by a laser pulse can be significantly enhanced or completely suppressed by applying another laser pulse and varying its time delay or relative carrier-envelope phase. It was also showed that the two-photon absorption between two excited states can be selectively excited since the modulation period of the two-photon absorption depends on the transition frequency of the excited state. Moreover, the two-photon absorption control in the molecular system was studied, and demonstrated that the absorption bandwidth will greatly decrease the control efficiency of the two-photon absorption.

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