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Coherent Features of Resonance-Mediated Two-Photon Absorption Enhancement by Varying the Energy Level Structure, Laser Spectrum Bandwidth and Central Frequency *

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The femtosecond pulse shaping technique has been shown to be an effective method to control the multi-photon absorption by the light–matter interaction. Previous studies mainly focused on the quantum coherent control of the multi-photon absorption by the phase, amplitude and polarization modulation, but the coherent features of the multi-photon absorption depending on the energy level structure, the laser spectrum bandwidth and laser central frequency still lack in-depth systematic research. In this work, we further explore the coherent features of the resonance-mediated two-photon absorption in a rubidium atom by varying the energy level structure, spectrum bandwidth and central frequency of the femtosecond laser field. The theoretical results show that the change of the intermediate state detuning can effectively influence the enhancement of the near-resonant part, which further affects the transform-limited (TL)-normalized final state population maximum. Moreover, as the laser spectrum bandwidth increases, the TL-normalized final state population maximum can be effectively enhanced due to the increase of the enhancement in the near-resonant part, but the TL-normalized final state population maximum is constant by varying the laser central frequency. These studies can provide a clear physical picture for understanding the coherent features of the resonance-mediated two-photon absorption, and can also provide a theoretical guidance for the future applications.

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The multi-photon absorption process has attracted considerable attention for its applications in biology and medicine, such as fluorescence spectroscopy, three-dimensional fluorescence imaging or photodynamic therapy.[1−4] Realizing the manipulation of the multi-photon absorption not only is an important fundamental research for understanding the multi-photon excitation process, but also can optimize the fluorescence properties of an atom or molecule system for promoting their related applications. Fortunately, the femtosecond pulse shaping technique has been shown to be an effective method to control the multi-photon absorption by manipulating the constructive or destructive interference between different optical pathways in atoms,[5,6] molecules,[7,8] rare-earth ions,[9,10] and condensed state systems.[11] Recently, with the development of the femtosecond pulse shaping technique, the shaped pulse with an almost arbitrary temporal distribution can be obtained by controlling the spectral phase, amplitude and polarization of the femtosecond laser field,[12] and therefore various schemes have been proposed and used to manipulate the multi-photon absorption.[13−17] For example, the (1+1) resonance-mediated two-photon absorption can be effectively enhanced or suppressed by the π phase or the amplitude modulation of femtosecond laser field.[13] The non-resonant two-photon absorption can be tuned by the cosine or sinusoidal phase modulation.[14,15] Moreover, the single-photon fluorescence of IR 125 can be tuned by the polarization modulation of the femtosecond laser field, which is increased with the laser polarization changing from linear through elliptical to circular.[16] The up-conversion luminescence of Dy^{3+}-doped glass sample can be controlled by the laser polarization and the π phase modulation.[17]

Previous studies mainly focused on the control of the multi-photon absorption by the phase, amplitude and polarization modulation of the femtosecond laser field.[13−17] However, the coherent features of the multi-photon absorption depending on the energy level structure, the spectrum bandwidth and the central frequency of the femtosecond laser field still lack systematic research. In this work, we further explore the coherent features of the resonance-mediated two-photon absorption in rubidium atoms by varying the energy level structure, spectrum bandwidth and central frequency of the femtosecond laser field. We find that the change of the intermediate state detuning can effectively influence the enhancement of the near-resonant part, which further affects the TL-normalized final state population maximum. Moreover, as the laser spectrum bandwidths increase, the TL-normalized final state population maximum can be effectively enhanced due to the increase of the enhancement in the near-resonant two-photon part. However, varying the laser central frequency can change the final state population, but not affect the

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TL-normalized final state population maximum.

The schematic energy-level diagram of the rubidium atom and the resonance-mediated two-photon excitation processes are shown in Fig. 1(a). The $^5S_{1/2}$, $^5P_{1/2}$ and $^5D_{3/2}$ states represent the ground state $|g\rangle$, the intermediate state $|i\rangle$ and the final state $|f\rangle$, respectively. Usually, the cw nanosecond laser field has the relatively lower laser intensity, and the final state can be populated by the ground and excited state absorption under the cw nanosecond laser field excitation. However, the femtosecond laser field has relatively higher laser intensity and wide spectrum bandwidth, and can effectively induce the resonance-mediated two-photon absorption. When the rubidium atom is excited by the femtosecond laser field, the transition process of $|g\rangle \rightarrow |f\rangle$ can be realized through the intermediate state $|i\rangle$, which is the resonance-mediated two-photon absorption. As the spectral bandwidth of the femtosecond laser field is relatively wide, the transition frequency of $|g\rangle \rightarrow |i\rangle$ is in the laser spectrum range, and the resonance-mediated two-photon absorption includes on-resonant and near-resonant parts. For the on-resonant process, the population in the ground state $|g\rangle$ is pumped to the intermediate state $|i\rangle$ and further is excited to the final state $|f\rangle$ by absorbing photons with the transition frequencies of $|g\rangle \rightarrow |i\rangle$ and $|i\rangle \rightarrow |f\rangle$. For the near-resonant process, the population in the ground state $|g\rangle$ is directly pumped to the final state $|f\rangle$ without going through the intermediate state $|i\rangle$ by simultaneously absorbing two photons. Therefore, the resonance-mediated two-photon transition probability in a rubidium atom excited by the femtosecond laser field in the frequency domain can be approximated as[6]

$$S \propto |A_{\mathrm{On-Res.}} + A_{\mathrm{Near-Res.}}|^2, \tag{1}$$

with

$$A_{\mathrm{On-Res.}} \propto i\pi E_0(\omega_f - \omega_i)E_0(\omega_i) \cdot e^{i[\Phi(\omega_f-\omega_i)+\Phi(\omega_i)]}, \tag{2}$$

$$A_{\mathrm{Near-Res.}} \propto \wp \int_{-\infty}^{+\infty} d\omega \mathrm{E}_0(\omega_f - \omega) \cdot E_0(\omega) e^{i[\Phi(\omega_f-\omega)+\Phi(\omega)]}/(\omega_i - \omega), \tag{3}$$

where $\wp$ is Cauchy's principal value, $E(\omega) = E_0(\omega) \times \exp[i\Phi(\omega)]$ is the femtosecond laser field in frequency domain, and $E_0(\omega)$ and $\Phi(\omega)$ express the spectral amplitude and phase, respectively. It is obvious that the on-resonant term $A_{\mathrm{On-Res.}}$ in Eq. (2) is the two-photon excitation pathways via the intermediate state $|i\rangle$ with the resonant transition frequencies of ω_i and $\omega_f - \omega_i$. However, the near-resonant term $A_{\mathrm{Near-Res.}}$ in Eq. (3) includes all the two-photon excitation pathways without the intermediate state $|i\rangle$ with the transition frequencies of ω and $\omega_f - \omega$. Moreover, for the TL femtosecond laser field (i.e., $\Phi(\omega) = 0$), the on-resonant term $A_{\mathrm{On-Res.}}$ is an imaginary quantity (see Eq. (2)), while the near-resonant term $A_{\mathrm{Near-Res.}}$ is a real quantity (see Eq. (3)). Obviously, the on-resonant term $A_{\mathrm{On-Res.}}$ is constant and only related to the resonant transition frequencies of ω_i and $\omega_f - \omega_i$, but the near-resonant term $A_{\mathrm{Near-Res.}}$ is related to the opposite sign of $1/(\omega_i - \omega)$ for different laser spectrum components in Eq. (3), which induce a destructive interference for the TL femtosecond laser field. To realize the coherent interference between the near-resonant two-photon transition pathways to enhance the final state population, the π phase modulation is an effective method and used in many experiments.[13,17]

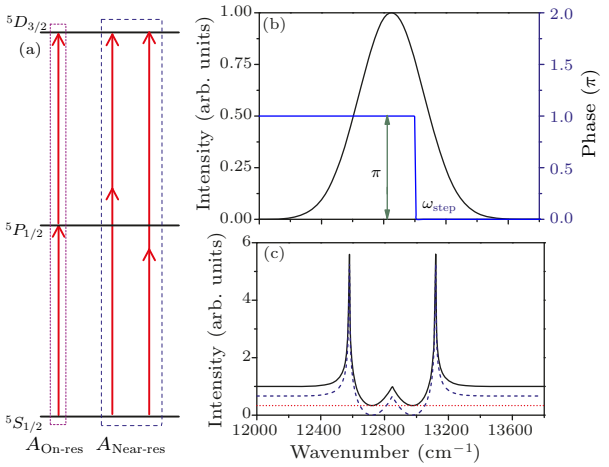

Fig. 1. (Color online) (a) The energy level diagram of the rubidium atom and the excitation processes by the resonance-mediated two-photon absorption, including on-resonant and near-resonant processes. (b) The femtosecond laser spectrum (black line) modulated by the π phase modulation. (c) The TL-normalized final state population (black solid line) as a function of the π phase step position, together with the contributions of on-resonant (red dotted line) and near-resonant two-photon absorption (blue dashed line).

As shown in Fig. 1(b), the femtosecond laser field in frequency domain (black solid line) is modulated by the π phase. For the π phase modulation, the phase modulation depth is π, and the phase step position is ω_{step}. One can see that the phase is set as π for the frequency components $\omega \leq \omega_{\mathrm{step}}$ (i.e., $\Phi(\omega \leq \omega_{\mathrm{step}}) = \pi$), while the phase is set to be zero for the other frequency components (i.e., $\Phi(\omega > \omega_{\mathrm{step}}) = 0$). In our theoretical simulation, the central frequency of the femtosecond laser field is 12850 cm^{-1} with the pulse width of 30 fs, and the transition frequencies of $|g\rangle \rightarrow |i\rangle$ and $|g\rangle \rightarrow |f\rangle$ are 12580 and 25700 cm^{-1}. Figure 1(c) shows the TL-normalized resonance-mediated two-photon absorption by the π phase modulation based on Eq. (1), together with the contributions of on-resonant (red dotted line) and near-resonant two-photon absorption (blue dashed line) according to Eqs. (2) and (3). All the data are normalized by the population excited by the TL pulse, and therefore the TL-normalized final state population can directly reflect the resonance-mediated two-photon absorption control. Hereafter, the same method is utilized, and we use the TL-normalized final state population maximum to ex-

press the maximum value of the TL-normalized final state population by the π phase modulation. As can be seen, the TL-normalized resonance-mediated two-photon absorption and the near-resonant two-photon absorption can be enhanced or suppressed by varying the phase step position, while the on-resonant two-photon absorption remains unchanged. Moreover, the near-resonant two-photon absorption can be effectively enhanced with the π phase step position at the resonant frequencies of 12850 or 13120 cm^{-1} due to the sign inversion of $e^{i[\Phi(\omega_f^{-\omega})+\Phi(\omega)]}$ in Eq. (3), and then the resonance-mediated two-photon absorption can obtain the maximum value. Obviously, the π phase modulation is an effective method to realize enhancement of the resonance-mediated two-photon absorption, and here we further study the coherent features of the multi-photon absorption depending on the energy level structure, the laser spectrum bandwidth and laser central frequency.

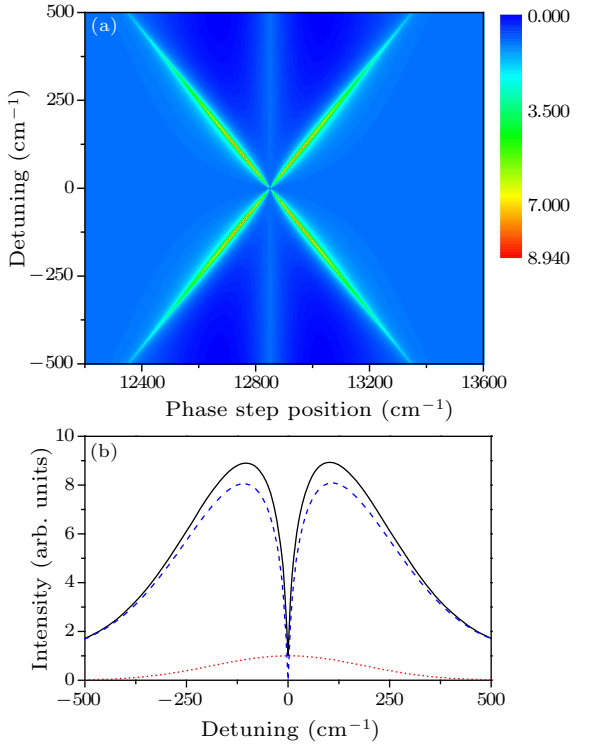

Fig. 2. (Color online) (a) A contour plot of the TL-normalized final state population as functions of the π phase step position and the intermediate state detuning changing from -500 to 500 cm^{-1}. (b) The TL-normalized final state population maximum P_{fmax} (black solid line) by varying the intermediate state detuning, together with the contributions of on-resonant (red dotted line) and near-resonant two-photon absorption (blue dashed line).

To study the influence of the intermediate state detuning on the final state population, we calculate the TL-normalized final state population as functions of the π phase step position and the intermediate state detuning from -500 to 500 cm^{-1} in Fig. 2(a). Here the intermediate state detuning is defined by the function of $\Delta d = \omega_f/2 - \omega_i$. Obviously, by varying the intermediate state detuning, the line types of TL-normalized final state population by the π phase modulation are different. The maximum value of the TL-normalized final state population can change, and the phase step position of the maximum value can move. Moreover, we further calculate the TL-normalized final state population maximum (black solid line) by varying the transition frequency of the intermediate state with the phase step position at resonant frequency of $|g\rangle \to |i\rangle$ or $|i\rangle \to |f\rangle$, together with the contributions of on-resonant (red dotted line) and near-resonant two-photon absorption (blue dashed line), and the theoretical results are shown in Fig. 2(b). It is noted that the line types of TL-normalized final state population maximum are symmetrical. As can be seen, with the intermediate state detuning Δd increasing from 0 to 500 cm^{-1} (or from 0 to -500 cm^{-1}), the TL-normalized final state population maximum first increases and then decreases, which is mainly induced by the increase and decrease of the near-resonant two-photon absorption. When the detuning Δd is close to 0 cm^{-1}, the structure of energy levels is highly symmetrical, and the near-resonant transition probability amplitude based on Eq. (3) is smaller due to the same sign of $e^{i[\Phi(\omega_f^{-\omega})+\Phi(\omega)]}$ for all the spectrum components. Therefore, the TL-normalized final state population maximum is relatively weaker and mainly contributed by the on-resonant part, which cannot be enhanced by the π phase modulation. As the detuning Δd is close to 500 cm^{-1} (or -500 cm^{-1}), the resonant transition frequencies are far away from the central spectrum components of the femtosecond laser field, and $1/(\omega_i - \omega)$ in Eq. (3) is relatively smaller, which induces the decrease of the near-resonant two-photon absorption and the TL-normalized final state population maximum. When the detuning Δd approaches to 105 cm^{-1} (or -105 cm^{-1}), the TL-normalized final state population maximum can be effectively increased due to the near-resonant two-photon absorption enhancement. Therefore, selecting a suitable energy level structure can obtain the effective enhancement of the TL-normalized final state population maximum.

Next, we further demonstrate the dependence of the final state population on the spectrum bandwidth of the femtosecond laser field, we calculate and show the TL-normalized final state population as functions of the π phase step position and the laser spectrum bandwidth in Fig. 3(a). Clearly, the TL-normalized final state population maximum is first constant and then increases with the increase of the laser spectrum bandwidth, while the phase step position of the maximum value is stable at the resonant transition frequencies, which is correlated with the energy level structure of the atom system. In addition, the TL-normalized final state population maximum P_{fmax} (black line) by varying the laser spectrum bandwidth is shown in Fig. 3(b), together with the contributions of on-resonant (red dotted line) and near-resonant two-photon absorption (blue dashed line). Obviously, the TL-normalized final state population maximum P_{fmax} is constant for the laser spectrum bandwidth changing from 10 to 200 cm^{-1}. As the laser spec-

trum bandwidth changes from 10 to 200 cm^{-1}, the resonant transition frequency of $|g\rangle \to |i\rangle$ or $|i\rangle \to |f\rangle$ is not in the laser spectrum range due to the narrow laser spectrum bandwidth, and therefore the final state is mainly populated by the non-resonant two-photon absorption, which cannot be enhanced by the π phase modulation. As the laser spectrum bandwidth increases, the transition frequency of $|g\rangle \to |i\rangle$ or $|i\rangle \to |f\rangle$ is in the spectrum range of the femtosecond laser field, and the near-resonant two-photon absorption can be effectively enhanced by the π phase modulation, which can induce the enhancement of the TL-normalized final state population. Moreover, the contribution of the near-resonant part for the final state population increases with the laser spectrum width, and results in the rise of the TL-normalized population maximum P_{fmax}. Therefore, the laser spectrum bandwidth can provide a feasible scheme to manipulate the contribution of the near-resonant part, and can further affect the TL-normalized final state population maximum.

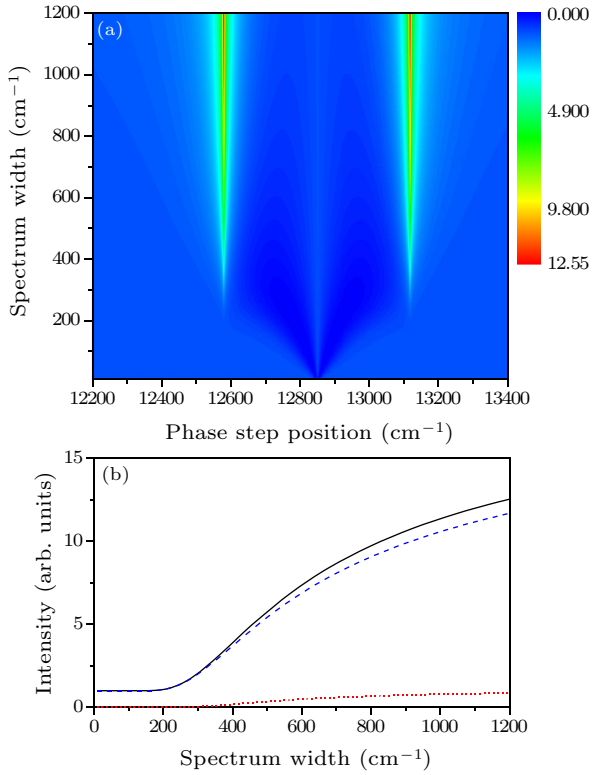

Fig. 3. (Color online) (a) A contour plot of the TL-normalized final state population as functions of the π phase step position and the laser spectrum bandwidth changing from 10 to 1200 cm^{-1}. (b) The TL-normalized final state population maximum P_{fmax} (black solid line) by varying the laser spectrum bandwidth, together with the contributions of on-resonant (red dotted line) and near-resonant two-photon absorption (blue dashed line).

The central frequency of the femtosecond laser field is an important parameter to study the control of the final state population. We calculate and show the TL-normalized final state population as functions of the π phase step position and the laser central frequency in Fig. 4(a). It can be seen that the maximum value of TL-normalized final state population is constant, and the step position of the maximum value is not changed by varying the laser central frequency. To study its physical mechanism, we calculate and show the final state population of the shaped pulse (black solid line) with the phase step position at the resonant transition frequencies and the TL pulse (red dashed line) by varying the laser central frequency in Fig. 4(b). As can be seen, with the increase of the laser central frequency, the final state population excited by the shaped pulse and the TL pulse simultaneously increases and decreases. Here the pulse width of the femtosecond laser field is 30 fs, the spectrum width is wide enough to induce the resonance-mediated two-photon absorption, and therefore the TL-normalized final state population is unchanged by varying the laser central frequency. These results indicate that the central frequency of the femtosecond laser field can change the final state population, but cannot affect the TL-normalized final state population maximum P_{fmax}.

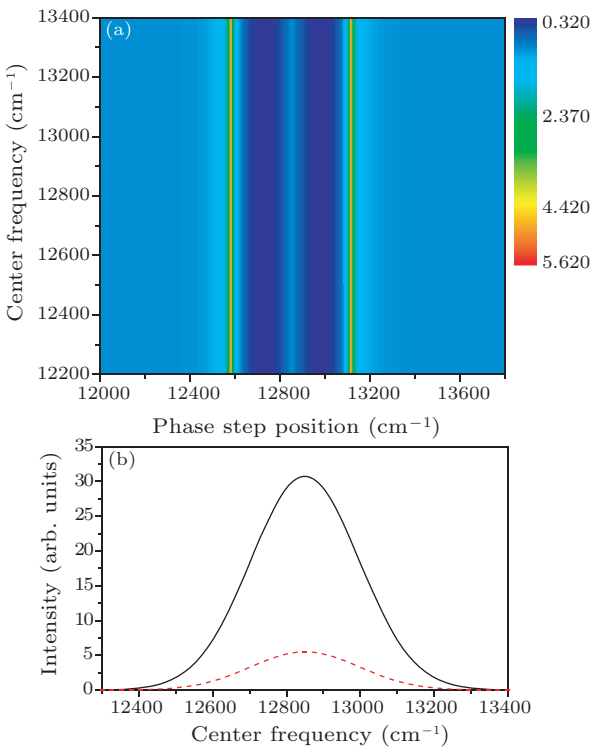

Fig. 4. (Color online) (a) A contour plot of the TL-normalized final state population as functions of the π phase step position and the laser central frequency changing from 12200 to 13400 cm^{-1}, respectively. (b) The final state population of the shaped pulse (black solid line) and the TL pulse (red dashed line) by varying the laser central frequency.

In summary, we have demonstrated that the resonance-mediated two-photon absorption in rubidium atoms can be effectively enhanced by the π phase modulation, and we further study the coherent features of the resonance-mediated two-photon absorption by varying the energy level structure, spectrum bandwidth and central frequency of the femtosecond laser field. Our theoretical calculations show that the

enhancement of the near-resonant part can be effectively modulated by varying the energy level structure, which can further affect the TL-normalized final state population maximum. In addition, with the increase of the laser spectrum bandwidth, the TL-normalized final state population maximum can be effectively enhanced due to the contribution of the near-resonant part, but it is not affected by varying the laser central frequency. These studies can provide a clear physical picture for understanding the coherent features of the resonance-mediated two-photon absorption, and can also provide a theoretical guidance for the future applications.

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