# **Boundary Analysis in Block Schemes for Control of Some Multi-Level Quantum Systems**

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*Abstract*— Two-photon absorption (TPA) is an important quantum process in the light-matter interaction. When more than one TPA pathway exists in a quantum system, it is necessary to utilize the interference of these pathways to increase the sum TPA amplitude. A simple block scheme (Phys. Rev. A 2013, 88, 053427) is proposed to maximize the constructive interference of two TPA pathways in a four-level diamondconfiguration quantum system. It can also be expanded to control multiple TPA pathways (Chin. J. Chem. Phys. 2015, 28, 426). The spectral boundaries in the scheme is related to the energy structure of the system. This work tries to give an analysis of the boundaries for general four-level and five-level quantum systems, which have two and three TPA pathways, respectively. How the boundaries and thus the number of blocks change with the energy structure will be explored.

### I. INTRODUCTION

THE control of quantum dynamics is of scientific impor-<br>tance both in physics and automation. Many strategies<br>have been prepared to monipulate quantum dynamics, such have been proposed to manipulate quantum dynamics, such as feedback control [1], [2], [3], [4] and coherent control [5], [6], [7]. Two-photon absorption (TPA) is a widely investigated fundamental process, and different strategies of coherent control can be employed to enhance the TPA amplitude [5], [8], [9], [10], [11]. Silberberg *et al.* [10] suggested that a resonant TPA rate can be enhanced via constructive interference between non-resonant contributions, and a  $\pi/2$ step scheme was proposed. Lee *et al.* [11] investigated the quantum interference control of a four-level diamondconfiguration quantum system, and the control idea was just to tune the interference between two TPA pathways. In their scheme, the whole spectrum is divided into eight blocks, and the seven spectral boundaries depend on the resonant frequencies and the dipole moments. The strategy can be generalized to more general quantum systems including  $N$ TPA pathways, and an 4N-block scheme could be a practical choice for coherent control of these pathways [12].

Considering a typical TPA process induced by a weak femto-second laser pulse  $\varepsilon(t)$ , the transition amplitude from the initial ground state  $|g\rangle$  to the final state  $|f\rangle$  is

$$
U = -\pi \frac{\mu_{fn}\mu_{ng}}{\hbar^2} E(\omega_{ng}) E(\omega_{fn})
$$

$$
+ i \frac{\mu_{fn}\mu_{ng}}{\hbar^2} \wp \int_{-\infty}^{\infty} \frac{E(\omega) E(\omega_{fg} - \omega)}{\omega_{ng} - \omega} d\omega.
$$
 (1)

where the  $E(\omega)$  is the Fourier transform of  $\varepsilon(t)$ ,  $\omega_{nq}$  and  $\omega_{fn} = \omega_{fq} - \omega_{nq}$  are the resonance frequencies, and  $\wp$  is the principal value of Cauchy. The right hand side of this equation clearly has two (resonant and non-resonant) terms.

For simplicity, the following definitions are adopted

$$
D_n = \frac{\mu_{fn}\mu_{ng}}{\hbar^2},\tag{2a}
$$

$$
\tilde{\omega} = \omega_{fg} - \omega. \tag{2b}
$$

For a general quantum system involving more than one TPA pathway, the sum transition amplitude can be written as

$$
U = -\pi \sum_{n} D_{n} E(\omega_{ng}) E(\omega_{fn})
$$

$$
+ i \sum_{n} D_{n} \wp \int_{-\infty}^{\infty} \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega} d\omega.
$$
 (3)

In the  $4N$ -block scheme [12] mentioned above, the spectral boundaries are the resonant frequencies  $\omega_{ng}$ ,  $\omega_{fn}$  and  $\omega_{fg}/2$ , some critical frequencies  $\omega_c$ , and  $\omega_{fg} - \omega_c$ . The critical frequencies  $\omega_c$  are the roots of the following equation with  $E(\omega)$  being transform limited (TL) pulses

$$
f(\omega) = \sum_{n} D_n \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega} = 0.
$$
 (4)

It is easy to see that  $f(\omega)$  is just the integral kernel of the non-resonant term, and the integration over different regions across these boundaries will change its plus-minus sign for TL pulses. The principle in the block scheme is to make the non-resonant terms over different spectral blocks align in-phase with the resonant terms for achieving maximal constructive interference.

The number of blocks is  $4N$  when Eq. (4) has  $N - 1$ real roots, and cases can be different when imaginary roots appear. In this work, we will explore how the boundaries and thus the number of blocks change with the energy structure for some simple multi-level systems. Lee's scheme only considers the dominant non-resonant contributions by just making  $\sum_{n} D_n \frac{1}{\omega_{ng}-\omega} = 0$ . We would propose another block<br>scheme to maximize the sum amplitude of two TPA pathways scheme to maximize the sum amplitude of two TPA pathways by taking all non-resonant contributions into account.

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Fig. 1. (Color online) A five-level model system, and the three different colored arrows denote the three TPA pathways.

In Lee's scheme, for a four-level system involving two TPA pathways, the equation  $\sum_n D_n \frac{1}{\omega_{ng}-\omega} = 0$  is linear and thus certainly has one real root, which leads to a eightblock control scheme. In this work, we prove that there are always two real roots when we extend Lee's scheme to a five-level system with three TPA pathways, which leads to a twelve-block control scheme. Situation is quite different for the new block scheme, and number of roots of Eq. (4) are discussed for a four-level system with two pathways.

## II. BOUNDARY ANALYSIS IN LEE'S SCHEME FOR A FIVE-LEVEL SYSTEM

For a five-level quantum system having three TPA pathways (shown as Fig. 1), the boundary equation is

$$
\sum_{n=a,b,c} \frac{D_n}{\omega_{ng} - \omega} = 0,\tag{5}
$$

which leads to

$$
A\omega^2 - B\omega + C = 0,\t\t(6)
$$

with

$$
A = \sum_{n=a,b,c} D_n,
$$
 (7a)

$$
B = \sum_{n=a,b,c} \left( D_n \sum_{n'=a,b,c}^{n' \neq n} \omega_{n'g} \right), \qquad (7b)
$$

$$
C = \sum_{n=a,b,c} \left( D_n \prod_{n'=a,b,c}^{n'\neq n} \omega_{n'g} \right). \tag{7c}
$$

The discriminant  $\Delta = B^2 - 4AC$  can also written as a quadratic function about  $\omega_{aa}$ ,

$$
\Delta = A' \omega_{ag}^2 + B' \omega_{ag} + C', \tag{8}
$$

Here  $A'$ ,  $B'$ ,  $C'$  are defined as

$$
A' = (D_b + D_c)^2,
$$
  
\n
$$
B' = 2(D_b + D_c) [D_a (\omega_{bg} + \omega_{cg}) + D_b \omega_{cg} + D_c \omega_{bg}]
$$
  
\n
$$
-4(D_b \omega_{cg} + D_c \omega_{bg}) \sum_{n} D_n,
$$
\n(9b)

$$
C' = [D_a (\omega_{bg} + \omega_{cg}) + D_b \omega_{cg} + D_c \omega_{bg}]^2
$$
  
- 4D\_a \omega\_{bg} \omega\_{cg} \sum\_{n=a,b,c} D\_n. (9c)

Then the discriminant can be finally written as

$$
\Delta = A' \left( \omega_{ag} + \frac{B'}{2A'} \right)^2
$$
  
+ 
$$
4 \left( \frac{\omega_{bg} - \omega_{cg}}{D_b + D_c} \right)^2 \prod_{n=a,b,c} D_n \sum_{n=a,b,c} D_n, \qquad (10)
$$

which is always equal or larger than zero. It is obvious that  $\triangle = 0$  indicates  $\omega_{bg} = \omega_{cg}$ , and  $\omega_{ag} + \frac{B'}{2A'} = 0$  will further lead to  $\omega_{ag} = \omega_{bg} = \omega_{cg}$ . That means Eq. (5) always has two real roots for a five-level quantum system when any two of the intermediate energy levels are not degenerate.

#### III. NEW BLOCK SCHEME FOR A FOUR-LEVEL SYSTEM

In Lee's scheme, only the dominant non-resonant contributions are considered to obtain the boundaries making  $f(\omega)$ being zero. In this section, a new scheme is proposed to obtain the accurate boundaries by taking all non-resonant contributions into account. The Cauchy principle value in Eq. (3) can be further simplified by the variable transformation  $\omega = \omega_{fg} - \tilde{\omega},$ 

$$
\wp \int_{-\infty}^{\infty} \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega} d\omega
$$
  
\n
$$
= \wp \left( \int_{-\infty}^{\omega_{fg}/2} \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega} d\omega + \int_{\omega_{fg}/2}^{\infty} \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega} d\omega \right)
$$
  
\n
$$
= \wp \left( \int_{\omega_{fg}/2}^{\infty} \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega_{fg} + \tilde{\omega}} d\tilde{\omega} + \int_{\omega_{fg}/2}^{\infty} \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega} d\omega \right)
$$
  
\n
$$
= \wp \left( \int_{\omega_{fg}/2}^{\infty} \frac{E(\omega) E(\tilde{\omega})}{\tilde{\omega} - \omega_{fn}} d\tilde{\omega} + \int_{\omega_{fg}/2}^{\infty} \frac{E(\omega) E(\tilde{\omega})}{\omega_{ng} - \omega} d\omega \right)
$$
  
\n
$$
= \wp \int_{\omega_{fg}/2}^{\infty} E(\omega) E(\tilde{\omega}) \left( \frac{1}{\omega - \omega_{fn}} + \frac{1}{\omega_{ng} - \omega} \right) d\omega. (11)
$$

The new spectral boundaries  $\omega_c$  are roots of the following equation

$$
f_T(\omega) = \sum_n D_n \left( \frac{1}{\omega - \omega_{fn}} + \frac{1}{\omega_{ng} - \omega} \right)
$$

$$
= \sum_n \frac{D_n \lambda_n}{(\omega - \omega_{ng})(\omega - \omega_{fn})} = 0.
$$
 (12)

where  $\lambda_n = \omega_{fn} - \omega_{ng}$ . As seen in Eq. (11), the integration region is  $(\omega_{fq}/2, \infty)$ , so this new scheme considers all the photon pairs adding up to  $\omega_{fn}$  while Lee's scheme only considers the dominant ones.



Fig. 2. (Color online) A four-level system with two TPA pathways denoted by left and right arrows. The horizontal dashed line corresponds to the frequency of  $\omega_{fg}/2$ , with  $\omega_{fg}$  being the frequency difference between levels  $|a\rangle$  and  $|f\rangle$ . levels  $|g\rangle$  and  $|f\rangle$ .

Only the case of  $N = 2$  will be analysed below since the above equation is more complex than that in Lee's scheme, that is  $\sum_n D_n \frac{1}{\omega_{ng}-\omega} = 0$ . Two intermediate levels are  $|a\rangle$  and  $|b\rangle$  as shown in Fig. 2. Then Eq. (12) becomes

$$
\sum_{n=a,b} \frac{D_n \lambda_n}{(\omega - \omega_{ng})(\omega - \omega_{fn})} = 0.
$$
 (13)

The numerator is

$$
f_1 = D_a \lambda_a \left( \omega - \omega_{bg} \right) \left( \omega - \omega_{fb} \right) + D_b \lambda_b \left( \omega - \omega_{ag} \right) \left( \omega - \omega_{fa} \right).
$$
 (14)

It is a quadratic function and can be rewritten as

$$
f_1 = \bar{A}\omega^2 + \bar{B}\omega + \bar{C}.\tag{15}
$$

with

$$
\bar{A} = \sum_{n=a} D_n \lambda_n,\tag{16a}
$$

$$
\bar{B} = -\omega_{fg} \sum_{n=a,b} D_n \lambda_n,
$$
\n(16b)

$$
\bar{C} = D_a \lambda_a \omega_{bg} \omega_{fb} + D_b \lambda_b \omega_{ag} \omega_{fa}.
$$
 (16c)

The corresponding discriminant is

$$
\Delta = \bar{B}^2 - 4\bar{A}\bar{C}
$$
  
=  $\lambda_a \lambda_b (D_a \lambda_a + D_b \lambda_b) (D_a \lambda_b + D_b \lambda_a).$  (17)

If  $D_a = D_b$ , the equation can be simplified as

$$
\Delta = D_a^2 \lambda_a \lambda_b (\lambda_a + \lambda_b)^2. \tag{18}
$$

It is obvious the sign of discriminant depends only on the product of  $\lambda_a \lambda_b$  in this case as shown in Fig. 3. It has to be noted that Eq. (13) will becomes  $\overline{C}=0$  when  $\lambda_b = -\lambda_a$ , which is not true. Therefore, the line  $\lambda_b = -\lambda_a$  corresponds to no real roots.



Fig. 3. (Color online) The schematic diagram showing the regions of the discriminant  $\triangle$  with positive ("+") or negative ("-") signs. The red solid lines corresponds to  $\Delta = 0$ , which has only one real root. The line  $\lambda_b = -\lambda_a$ corresponds to no real roots.

TABLE I SIGN ANALYSIS OF THE DISCRIMINANT IN DIFFERENT REGIONS

Regions	$\lambda_a$	$\lambda_b$	$\frac{D_b}{D_a}$ $\lambda_b$ $\lambda_a$ $^+$	$\frac{D_a}{D_b}\lambda_a$ $\lambda_b$ $^+$	

If  $D_a \neq D_b$ , the equation can be changed as

$$
\Delta = D_a D_b \lambda_a \lambda_b \left( \lambda_b + \frac{D_a}{D_b} \lambda_a \right) \left( \lambda_b + \frac{D_b}{D_a} \lambda_a \right). \tag{19}
$$

$$
\mathbb{N} \tag{20}
$$

The signs of the discrimination in different regions can be obtained by making  $\triangle = 0$ ,

$$
\begin{cases}\n\lambda_a = 0, \\
\lambda_b = 0, \\
\lambda_b = -\frac{D_a}{D_b}\lambda_a, \\
\lambda_b = -\frac{D_b}{D_a}\lambda_a.\n\end{cases}
$$
\n(21)

So the signs of the discriminant can be analyzed as in Tab. I, and the results are plotted in Fig. 4. Here it is assumed that  $D_a > D_b$  without loss of generality. Similarly, Eq. (13) will also lead to  $\overline{C} = 0$  when  $\lambda_b = -\frac{D_a}{D_b}\lambda_a$ , and thus the line  $\lambda_b = -\frac{D_a}{D_b} \lambda_a$  corresponds to no real roots.

#### IV. NUMERICAL SIMULATIONS

In this section, two examples are, respectively, employed to illustrate the control schemes in Section II and Section



Fig. 4. (Color online) The schematic diagram showing the regions of discriminant  $\triangle$  with positive ("+") or negative ("-") signs, where  $\lambda_a$  =  $\omega_{fa} - \omega_{ag}$  and  $\lambda_b = \omega_{fb} - \omega_{bg}$ . The red solid lines corresponds to  $\wedge = 0$  which has only one real root. The line  $\lambda_b = \omega_a$  or corresponds  $\Delta = 0$ , which has only one real root. The line  $\lambda_b = -\frac{D_a}{D_b}\lambda_a$  corresponds to no real roots. to no real roots.

III. A laser pulse with a Gaussian envelope is employed to drive the system from state  $|g\rangle$  to state  $|f\rangle$  in the examples

$$
E(\omega) = B \exp\left\{-\frac{(\omega - \omega_0)^2}{2\delta^2}\right\} e^{i\phi(\omega)},\tag{22}
$$

where  $B = 6.0 \times 10^{-4}$ , small enough to make sure that the light-matter interaction is perturbative,  $\omega_0 = 0.05856$  and  $\delta = 5.7518 \times 10^{-4}$ .

In the control schemes, without loss of generality, we all assume  $\phi(\omega)=0$  when  $\omega \in (-\infty, \omega_{fg}/2)$ , and only half of the spectral blocks are shown in Figs. 5, 6, 7 and 8.

#### *A. Example 1*

In Section II, it is proved that there are always two real roots for a general non-degenerate five-level system involving three TPA pathways, which leads to twelve blocks in the control scheme. The following parameters (atomic units) are used for a five-level system:  $\omega_{ag} = 0.0595$ ,  $\omega_{bg} = 0.0601$ ,  $\omega_{cg} = 0.0607, \ \omega_{fg} = 0.1180, \ \bar{D}_a = 1.1590, \ \bar{D}_b = 1.1615,$ and  $D_c = 1.2508$ . The two real roots are  $\omega_1 = 0.0597500$ and  $\omega_2 = 0.0604345$ .

In Lee's scheme, the phases of the other blocks are shaped as

$$
\phi(\omega) = \begin{cases} +\frac{\pi}{2} & (\omega_{fg}/2, \omega_{ag}) \bigcup (\omega_1, \omega_{bg}) \bigcup (\omega_2, \omega_{cg}) \\ -\frac{\pi}{2} & (\omega_{ag}, \omega_1) \bigcup (\omega_{bg}, \omega_2) \bigcup (\omega_{cg}, \infty) \end{cases},
$$
\n(23)

which is shown in Fig. 5. The total TPA amplitude can be enhanced 5.39 times compared with transform limited pulses as listed in Tab. II.

#### *B. Example 2*

In the new block scheme for a four-level system involving two TPA pathways, there are three cases for the discriminant,



Fig. 5. (Color online) Phase modulating strategy for a five-level system in Lee's block scheme.



Fig. 6. (Color online) Phase modulating strategy when  $\triangle > 0$  for a four-level system in the new block scheme.

and numerical examples are given correspondingly in the following. The results are listed in Tab. III. Compared with TL pulses, the total TPA amplitude can be, respectively, enhanced 7.76, 3.97 and 8.25 times for the three cases.

*1)* Case  $\Delta > 0$ : The following parameters are used in this case:  $\omega_{ag} = 0.0595, \omega_{bg} = 0.0601, \omega_{fg} = 0.1180, D_a =$ 1.1590 and  $D_b = 1.1615$ . Here  $\omega_{bg} > \omega_{ag} > \omega_{fg}/2$ . The roots of Eq. (12) are  $\omega_1 = 0.0597413$  and  $\omega_2 = 0.0582586$ .

The phase-shaping strategy is

$$
\phi(\omega) = \begin{cases} +\frac{\pi}{2} & (\omega_{fg}/2, \omega_{ag}) \bigcup (\omega_1, \omega_{bg}) \\ -\frac{\pi}{2} & (\omega_{ag}, \omega_1) \bigcup (\omega_{bg} + \delta, \infty) \end{cases}, \quad (24)
$$

which is shown in Fig. 6.

*2) Case*  $\Delta = 0$ : The following parameters are used in this case:  $\omega_{ag} = 0.0590, \ \omega_{bg} = 0.0601, \ \omega_{fg} = 0.1180,$  $D_a = 1.1590$ , and  $D_b = 1.1615$ . Here  $\omega_{ag} = \omega_{fg}/2 < \omega_{bg}$ .

The phase-shaping strategy is

$$
\phi(\omega) = \begin{cases} +\frac{\pi}{2} & (\omega_{fg}/2, \omega_{bg}) \\ -\frac{\pi}{2} & (\omega_{bg}, \infty) \end{cases},
$$
 (25)

which is shown in Fig. 7.

#### TABLE II

TPA AMPLITUDES OF A FIVE-LEVEL SYSTEM WITH LEE'S BLOCK SCHEME. FOR SIMPLICITY, ALL THE VALUES ARE REDUCED BY A FACTOR OF  $B^2$ .

Control pulses	$\cup$ r	$\cup$ n.r.		
TL pulse	-4.723	6.216i	$-4.723 + 6.216i$	7.807
optimal pulse	$-4.723$	$-37.392$	$-42.115$	42.116

TABLE III TPA AMPLITUDES OF A FOUR-LEVEL STATE SYSTEM WITH THE NEW BLOCK SCHEME. FOR SIMPLICITY, ALL THE VALUES ARE REDUCED BY A FACTOR OF  $B^2$ .





Fig. 7. (Color online) Phase modulating strategy when  $\Delta = 0$  for a four-level system in the new block scheme.

*3) Case*  $\Delta$  < 0*:* The following parameters are used in this case:  $\omega_{ag} = 0.05799, \ \omega_{bg} = 0.0601, \ \omega_{fg} = 0.1180,$  $D_a = 1.1590$ , and  $D_b = 1.1615$ . Here  $\omega_{ag} < \omega_{fg}/2 < \omega_{bg}$ .

The phase-shaping strategy is

$$
\phi(\omega) = \begin{cases}\n-\frac{\pi}{2} & (\omega_{fg}/2, \omega_{fa}) (\omega_{bg}, \infty) \\
+\frac{\pi}{2} & (\omega_{fa}, \omega_{bg})\n\end{cases},
$$
\n(26)

which is shown in Fig. 8.

## V. CONCLUSIONS

In this work, we perform boundary analysis in two types of multi-block schemes for some multi-level quantum systems. Lee's scheme only considers the dominant non-resonant contributions in obtaining the block boundaries, while our new block scheme takes all non-resonant contributions into account to seek for the boundaries. For Lee's block scheme, it is proved that there are always two real roots for a general five-level system having three TPA pathways, which leads to a twelve-block control scheme. For the new block scheme, the boundary equation is more complex, and a four-level



Fig. 8. (Color online) Phase modulating strategy when  $\triangle \lt 0$  for a four-level system in the new block scheme.

system involving two TPA pathways is given as an example. Different cases, in which the corresponding discriminants are of different signs lead to different number of spectral blocks. Numerical simulations show that, compared with TL pulses, both the two types of multi-block schemes can enhance the total TPA amplitude effectively.

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