Tailoring the photon emission patterns in nanostructures

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Abstract. We investigate photon emission in coupled quantum dots on the basis of symmetry considerations. With the help of a new theorem that we have proved, we reveal the origin of various emission patterns, which is the combinative symmetry in the time domain and the spectrum domain. We are able to tailor the emission patterns to obtain emission spectra with odd harmonics alone, even harmonics alone or both odd and even harmonic components, and even with the quenching of all harmonic components. These interesting emission patterns can be obtained in experiments by a careful design of nanostructures, which have many applications in optical–electric nanodevices.

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1. Introduction

Photon emission in nanostructures has a crucial role in modern electronic/optical devices. Generating high-order harmonics is one efficient up-conversion method to obtain the desired spectra (e.g. THz spectra) from sources at lower frequencies. In recent decades, much effort has been made in the study of the generation of high-order harmonics in atomic/molecular systems and nanostructures [1]–[6], as well as their applications. For instance, Ahn et al [1] recently proposed a method for THz wave generation by high-order harmonic waves on the basis of semiconductor nanostructures driven by acoustic waves. The scheme of an electrically pumped photonic-crystal THz laser was developed by Chassagneux [2]. A method for THz wave generation by gigahertz (GHz) waves was suggested in [3].

Different harmonic components have interesting applications. For example, even harmonics were used as a test wave to diagnose the fast time evolution of current density [7]. Yet odd harmonics were often observed in experiments on atomic and molecular systems. The appearance of odd harmonics was attributed to the particular inversion symmetry in the central potential. There have been several studies of the dynamical symmetry [8] and related selection rules [9] in high-order harmonics generation. Attempts were made previously to obtain emission spectra with even harmonic components by avoiding the selection rule [10]–[14]. Some studies focused on the use of symmetry breaking to obtain even harmonics in molecular and atomic systems [12]–[14]. Symmetry breaking may be realized in a molecule with two nuclei of different masses [13]. Even harmonics are found to appear in driven double quantum wells, where the potential is not of inversion symmetry. Also, radiation may occur at noninteger multiples of the fundamental frequency.

There have been many theoretical and experimental studies of high-order harmonics and most of them have focused on atomic and molecular systems [15]–[27]. In spite of many studies on even harmonics generation, it appears that the deep origin of different emission patterns is still unclear, and there is a lack of an effective way of generating emission spectra of a specific pattern. In this paper, we study the emission spectra of coupled quantum dots (QDs) and show the symmetry origin of various emission patterns as well as methods for the generation of different emission patterns including those with even harmonics. The main advantage of QDs (artificial atoms) and coupled quantum dots (CQDs) (artificial molecules) is their tunability. By carefully designing the material and the growth process or by adding appropriate gate voltages, etc, one is able to adjust the energy levels/energy gaps in CQDs. One can further design the structure of CQDs (the relative positions of QDs, the inter-dot distances and the height and width of the tunneling barriers) to tune the optical dipole between QDs. The optical coupling between QDs can also be changed by tuning the polarization of the incident light. Compared with a single QD with multiple energy levels, the CQD system has the advantage of more tunability. By making full use of the tunability of coupled nanostructures, we propose an effective approach to tailor emission patterns. We reveal the origin of the appearance of odd/even harmonics on the basis of symmetry considerations. It is found that the emission pattern is determined not just by the inverse symmetry but also by a new type of symmetry: the combinative symmetry in the time domain and spectrum domain. Based on our findings, we are able to obtain emission patterns with even harmonics alone, odd harmonics alone, or both even and odd harmonics, and even with the quenching of all harmonic components. Our methods for the generation of various emission patterns in nanostructures have many applications.
2. Theoretical formalism

We consider a CQD system with one energy level for each dot. The energy is $E_i$ for the state $|i\rangle$ in the dot $i$, $i = 1 \ldots N$. This CQD is driven by an external field $E = F \cos(\omega_0 t) \vec{u}$, where $\vec{u}$ is a unit vector.

Under the dipole approximation, our system is described by the Hamiltonian

$$H = \sum_i E_i |i\rangle \langle i| + \sum_{i \neq j} G_{ij} \cos(\omega_0 t) |i\rangle \langle j|,$$

where $G_{ij} = F \vec{u} \cdot \vec{u}_{ij}$ are Rabi frequencies and $\vec{u}_{ij} = (i|e\vec{r}|j)$ is the dipole between dot $i$ and dot $j$. The equation of motion for the density matrix is written in the form [28]

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [H, \rho] - \Gamma : \rho,$$

where the last term describes the possible dissipation effects (e.g. that from spontaneous phonon emission). We set $\hbar = 1$ in the analytical calculations below. The time-dependent mean dipole moment can be calculated as $P(t) = \langle e\vec{r} \rangle = \sum_{ij} \mu_{ij} \rho_{ij}(t)$. With the help of Fourier transformation, we can obtain the emission spectrum $S(\omega) = |\int dt \exp(i\omega t) P(t)|^2$. We now present a theorem on origin of the various emission patterns. Several examples will be given.

**Theorem.** For a quantum system described by the Hamiltonian (1), if there exists one symmetric operation $Q$, which is the time shift $\theta : t \rightarrow t+T/2$ (or $-t+T/2$, $T = 2\pi/\omega_0$) combined with another operation $\Omega$ in the spatial/spectrum domain, i.e. $Q = \Omega \cdot \theta$, such that the initial condition and the Hamiltonian are invariant (or $H \rightarrow -H$) and the dipole operator $\hat{P}$ has a definite parity, then the emission spectrum contains no odd/even component if operator $\hat{P}$ is even/odd.

The proof is given below.

**Proof.** We first consider the following case: under the operation $Q$, $H \rightarrow H$. The Schrödinger equation $i \frac{d}{dt} |\psi(t)\rangle = H |\psi(t)\rangle$ remains invariant under $Q$ transformation. If the initial condition remains unchanged (up to a phase), that is, if the initial condition $|\psi(t = 0)\rangle$ satisfies $|\tilde{\psi}(t = 0)\rangle \equiv Q |\psi(t = 0)\rangle \equiv e^{-i\vec{r}} |\psi(t = 0)\rangle$, then we have $|\tilde{\psi}(t)\rangle = e^{-i\vec{r}} |\psi(t)\rangle$. Therefore, $P(t) = \langle \tilde{\psi}(t)|\hat{P}|\tilde{\psi}(t)\rangle = \langle \tilde{\psi}(t)|\hat{P}|\psi(t)\rangle = \langle \psi(t)|\hat{P}|\psi(t)\rangle = \langle \psi(t+T/2)|\Omega^{-1} \hat{P} \Omega |\psi(t+T/2)\rangle = \pm \hat{P}|\psi(t+T/2)\rangle$, where ‘+’ for even $\hat{P}$ and ‘−’ stands for odd $\hat{P}$. Thus, we have $P(n\omega_0) \equiv \int dt \exp(n\pi \omega_0 t/2) P(t) = \pm (-1)^n P(n\omega_0)$.

Similarly, we consider the following case: under the operation $Q$, $H \rightarrow -H$. Now $P(t) = \langle \psi(t)|\hat{P}|\psi(t)\rangle = \langle \tilde{\psi}(t)|\hat{P}|\psi(t)\rangle = \langle \tilde{\psi}(-t+T/2)|\Omega^{-1} \hat{P} \Omega |\psi(-t+T/2)\rangle = \langle \psi(-t+T/2)| \pm \hat{P}|\psi(-t+T/2)\rangle = \pm \hat{P}|\psi(-t+T/2)\rangle$.

Thus, we have the fact that $P(t)$ is real. Hence, we also have $P(n\omega_0) = \pm (-1)^n P(n\omega_0)$.

So we reach the results

$P(n\omega_0) = 0$ for odd $n$, if $\hat{P}$ is even (under $\Omega$);
$P(n\omega_0) = 0$ for even $n$, if $\hat{P}$ is odd (under $\Omega$).

From the above proof, it is seen that the required initial condition is $|\psi(t = 0)\rangle = e^{i\vec{r}} Q |\psi(t = 0)\rangle = e^{i\vec{r}} \Omega |\psi(T/2)\rangle = e^{i\vec{r}} \Omega U(T/2) |\psi(0)\rangle$, where $U$ is the time evolution operator.
So the initial state needs to be an eigenvector of the operator $\Omega U(T/2)$. This initial condition is not very convenient for practical use since the time evolution operator is involved. Here we demonstrate that the initial condition could be replaced with $|\psi(t = 0)\rangle = e^{i\gamma} \Omega |\psi(t = 0)\rangle$, and the emission spectra show little change as long as $|E_i - E_j|/\omega_0$ is small.

Physically we can understand that in the case of linear time evolution systems, a very small change in the initial condition does not lead to a big change of the time evolution of the system. More precisely, from the Floquet theorem, we have $|\psi(t)\rangle = \sum_c c_a e^{-i\epsilon_a t} |u_a(t)\rangle$, with $|u_a(t)\rangle$ being the time periodic Floquet state and $c_a$ is determined by the initial condition. It can easily be seen that $P(\omega) = \sum_{a,a'} c_a^* c_a K_{a,a'}(\omega)$, where $K_{a,a'}(\omega) = \int dt e^{i\omega t} e^{-i(c_a - c_a')t} U_{a'}^*(t) \hat{H} u_a(t)$. So, for two different initial conditions with $c_a, \tilde{c}_a$ such that $|c_a - \tilde{c}_a| < \eta$, with $\eta$ being a sufficiently small parameter, it is easy to show that $|P(\omega) - \bar{P}(\omega)| < \sum_{a,a'} 2\eta K_{a,a'}$, where $\bar{P}(\omega)$ are the emission spectra corresponding to the initial conditions with $c_a, \tilde{c}_a$, respectively.

Now we show that the difference between the initial conditions satisfying $|\psi(t = 0)\rangle = e^{i\gamma} \Omega |\psi(t = 0)\rangle$ and those satisfying $|\psi(t = 0)\rangle = e^{i\gamma} \Omega |\psi(t = 0)\rangle$ can be small enough if $|E_i - E_j|/\omega_0 \ll 1$. The time evolution operator satisfies the equation $i dU/dt = \hat{H} U$ with the initial condition $U(t = 0) = I$, where $I$ is identical operator. $\hat{H} = \hat{\bar{E}} + G \cos(\omega_0 t) \hat{\mu}$, where $\hat{\bar{E}} = \text{diag}(E_1, \ldots, E_N)$. The symmetric matrix $\hat{\mu}$ can be diagonalized as $A^{-1} \hat{\mu} A = \text{diag}(\lambda_1, \ldots, \lambda_N)$. Using the transformation $U = AB\tilde{U} A^{-1}$, $B = \text{diag}(e^{-i\lambda_1 G(\omega_0 t)}, \ldots, e^{-i\lambda_N G(\omega_0 t)})$, $f(t) = \int_0^t d\tau \cos(\omega_0 \tau)$, one sees that $i d\tilde{U}/dt = \hat{\bar{E}} \tilde{U}$, where $\hat{\bar{E}} = B^{-1} A^{-1} \hat{\bar{E}} A B, \tilde{U}(t = 0) = I$. Then it is clear that $[\tilde{U}(T/2) - I]$ and $|U(T/2) - I| = |A[\tilde{U}(T/2) - I]A^{-1}|$ can be small enough if $|E_i - E_j|/\omega_0$ is small. Since $Q = \Omega U(T/2) = \Omega + \Omega U(T/2) - \Omega = \Omega + \Omega (U(T/2) - I)$, standard perturbative calculation shows that the correction of the initial condition due to the perturbation $\Omega (U(T/2) - I)$ is small and one can use the initial condition $|\psi(t = 0)\rangle = e^{i\gamma} \Omega |\psi(t = 0)\rangle$ for the cases when $|E_i - E_j|/\omega_0 \ll 1$. In the above proof, the dissipation has not been included; yet we would like to point out that if the dissipation is small, the main feature of emission spectra remains unchanged, as verified by our numerical calculations, shown in the next section.

We have obtained one selection rule in the perturbation regime ($|E_i - E_j|/\omega_0 \ll 1$) and it depends on the initial condition. What will happen in the nonperturbation regime? In the regime with large $|E_i - E_j|/\omega_0$ and $G/\omega_0$, most quasienergies are large and the corresponding quasienergy states in the expansion $|\psi(t)\rangle = \sum_a c_a e^{-i\epsilon_a t} |u_a(t)\rangle$ lead to fast oscillation behavior. Then, only the quasienergy state with the smallest quasienergy $\epsilon_{a_0}$ (modulo $\hbar \omega_0$) is dominant, i.e. $|\psi(t)\rangle = c_{a_0} e^{-i\epsilon_{a_0} t} |u_{a_0}(t)\rangle$. If there exists one symmetric operation $Q$ such that $QHQ^{-1} = H$, $Qi_{a_0}^{\alpha} Q^{-1} = i_{a_0}^{\alpha}$, then the state $Q|u_{a_0}\rangle$ is also a quasienergy state with quasienergy $\epsilon_{a_0}$. If the quasienergy states are nondegenerate, $Q|u_{a_0}\rangle$ is just $|u_{a_0}\rangle$ (up to a phase). Straightforward calculation shows that $P(t) = -P(t + T/2)$ (note that $\hat{P}$ is odd in this case). Therefore, there are no even harmonics. One should note that this selection rule is insensitive to the initial condition.

To gain a clearer understanding of the physical picture, we study the case of double dots (with two levels), since some explicit solution could be obtained. The probability amplitudes $\beta, \alpha$ for one electron in states $|1\rangle$ and $|2\rangle$ satisfy the following equation,

$$
\begin{pmatrix}
\frac{d\beta}{dt} \\
\frac{d\alpha}{dt}
\end{pmatrix}
= \begin{pmatrix}
\frac{\Delta}{2} & G \cos(\omega_0 t) \\
-G \cos(\omega_0 t) & -\frac{\Delta}{2}
\end{pmatrix}
\begin{pmatrix}
\alpha \\
\beta
\end{pmatrix}.
$$

(3)

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where $\Delta = E_2 - E_1$. Define $A \equiv e^{iGf(t)\alpha}e^{iGf(t)\beta}$ and $B \equiv e^{-iGf(t)\alpha}e^{-iGf(t)\beta}$. Then we have the equations for $A$ and $B$,
\begin{align}
\frac{dA}{dt} &= \frac{\Delta}{2}e^{i2Gf(t)}B, \\
\frac{dB}{dt} &= \frac{\Delta}{2}e^{-i2Gf(t)}A.
\end{align}

The time-dependent dipole $P$ can be written as
\begin{equation}
P(t) = 2\mu_{12} \text{Re}\{\alpha^*\beta\} = \mu_{12} \text{Re}\{A^*A - B^*B\}/2.
\end{equation}

After some algebraic calculation, we have the equation for $P(t)$,
\begin{equation}
\frac{dP}{dt} = \Delta \text{Im}\{(a^* + b^*)(a - b)e^{i2Gf(t)}\} - \Delta^2 \int_0^t d\tau \cos[2Gf(t) - 2Gf(\tau)]P(\tau),
\end{equation}
where $a$ and $b$ are the probability amplitudes of the electrons in the two dots at initial time $t = 0$. We have made no approximation, and the above equation is exact. Here, we see that the first term, i.e. the initial condition, is important at $\Delta \ll 1$. The dependence of emission spectra on the initial state has also been noted in [29]. $P(t)$ becomes less sensitive to the initial condition at large $\Delta$, as we have discussed in the nonperturbation regime. For a particular type of initial condition $a = b$, i.e. $\rho_{11}(t = 0) = \rho_{22}(t = 0) = 0.5$, the first term on the right side of equation (6) vanishes. Then the equation can be solved for small $\Delta$ and one finds that there is no odd harmonic in the emission spectrum [30]. This agrees with our theorem, as shown in figure 1(b).

Figure 1. Emission spectra in double dots with different initial conditions. $\Delta = E_1 - E_2 = 1\text{ meV}$, $G_{12} = 850\text{ meV}$ and $\Gamma = 0.12\text{ GHz}$.
3. Numerical results and discussions

The above theorem is very helpful in ‘harmonic engineering’. It leads to many consequences and can be used in designing the optical emission patterns of coupled nanostructures. In the following, we give a few examples of the applications of our theory.

We perform our numerical calculations for typical GaAs dots. The energy level of each dot can be adjusted by using the confining potential. For instance, for the triple dots with the confining potential $\frac{1}{2}m^*\omega_i^2\overrightarrow{r}_i - \overrightarrow{r}_i^2$ ($i = 1, 2, 3$), with $\overrightarrow{r}_i$ being the position of dot $i$, $m^* = 0.067m_0$, $m_0$ the electron mass, $\hbar\omega_1 = 8.5$ meV, $\hbar\omega_2 = 8.0$ meV and $\hbar\omega_3 = 7.0$ meV, the energy spacings $E_1 - E_2 = 0.5$ meV and $E_2 - E_3 = 1.0$ meV are obtained. We use the typical value of dipole moment $e\ell_0$, $\ell_0 = 1.2$ nm, the incident laser intensity of the order of $10^{10}$ W cm$^{-2}$, and laser frequency $\omega_0/(2\pi) = 1.21 \times 10^{13}$ Hz. We choose the value of the relaxation rate $\Gamma = 0.12$ GHz, corresponding to the typical relaxation time of the order of nanoseconds. In the numerical calculations, the density matrix is obtained by solving equation (2) through the Runge–Kutta method with the time step of $0.01/\omega_0$, total steps of 1 500 000 and appropriate initial conditions. Emission spectra are obtained by numerically calculating the dipole through the evolution of density matrix elements $\rho_{ij}(t)$.

3.1. Double dots

We first consider the system of double dots with two levels. One can define $Q$ as the time shift $\theta: t \rightarrow t + T/2$ combined with $\Omega_1: c_1 \rightarrow -c_1$ (here and in the following $c_j, j = 1, \ldots, N$, refers to the annihilation operator for state $|jj\rangle$ in dot $j$). It can be easily seen that under such a transformation, $H$ is invariant, $\hat{P} = G(c_1^2c_3 + c_2^2c_1)$ is odd, and the initial condition $\rho_{22} = 1$ is invariant under transformation $\Omega_1$. So there is no even component and only odd peaks appear, as shown in figure 1(a) and noted in many previous publications.

As our theory shows, emission patterns depend on the initial condition. We can apply our theory to $\Omega_2: c_1 \leftrightarrow c_2$ for the initial condition $\rho_{11} = \rho_{22} = 0.5$. In this case, $\hat{P}$ is even. Thus there is no odd harmonic and only even harmonics appear, as seen in figure 1(b). (Note that these peaks may appear to be very small split from exact even components.) This also agrees with our previous analytical perturbation results. It is worth noting that even the peak for $\omega_0$ (corresponding to Rayleigh peak, which is often very pronounced in the usual emission spectra) disappears due to the particular symmetry. As seen in figure 1(c), there are both odd and even components for the initial condition $\rho_{11} = 0.3$, $\rho_{22} = 0.7$. This pattern is related to the fact that both of the symmetries related to $\Omega_1 \cdot \theta$ and $\Omega_2 \cdot \theta$ are broken.

3.2. Triple dots

We first show that we can use different initial conditions to realize quite different emission patterns. These results are displayed in figure 2. It is seen that the emission spectra may show odd only, even only or both even and odd components. In particular, the emission may be completely quenched under appropriate conditions. These results are based on our theorem. The symmetry property related to $\Omega_1: c_2 \rightarrow -c_2$ leads to the disappearance of even harmonics in figure 2(a). The symmetry $Q = \Omega_2 \cdot \theta$, $\Omega_2: c_1 \leftrightarrow c_3$ results in the spectrum without odd harmonics, as shown in figure 2(b). Quite interestingly, both transformations with $\Omega_1$ and $\Omega_2$ are satisfied in figure 2(c), which leads to the complete quenching of emission. This interesting
phenomenon is independent of the frequency of the incident light. Thus it is different from the quenching of emission due to coherent trapping. Actually, the electron has appreciable occupation probability for each dot. It is the symmetry-induced cancelation between the dipoles $P_{21}$ and $P_{23}$ that leads to complete quenching of emission. The breaking of both symmetries $\Omega_1$ and $\Omega_2$ leads to an emission with odd and even components, as shown in figure 2(d).

Figure 3 shows the dependence of emission patterns on the structure of nanosystems. Figures 3(a) and (c) show systems with chain structures and figures 3(b) and (d) show systems with loop structures. Systems (c) and (d) are of symmetric energy levels, which is absent from systems (a) and (b). The different emission patterns are the direct consequences of the symmetries generated by $\Omega_1$ for (a), $\Omega_2$ for (d), $\Omega_1$ and $\Omega_2$ for (c) and neither $\Omega_1$ nor $\Omega_2$ for (b).

3.3. Quadruple dots

Our theorem can be applied to more general/complex nanostructures. Here we give one example of tailoring the emission patterns of quadruple dots. A careful design of the structures of CQDs or the initial conditions leads to interesting emission spectra, as shown in figure 4. The generators of the related symmetries are $\Omega_1$: $c_2 \rightarrow -c_2$, $c_4 \rightarrow -c_4$ for figure 4(a); $\Omega_2$: $c_2 \leftrightarrow c_3$, $c_1 \leftrightarrow c_4$ for figures 4(b) and (d); and none for figure 4(c).

Our theorem on the essential role of symmetry in emission patterns emphasizes dynamic symmetry (spatial–temporal symmetry) of the system, instead of spatial symmetry alone. For a system with spatial inversion symmetry $\Omega_0$, there may be odd harmonics alone since the Hamiltonian is invariant under the symmetric operation $Q = \Omega_0 \cdot \theta$, and $\hat{P}$ is odd under $\Omega_0$. It is...
clear that dynamic symmetry is more basic. Our theory shows that the emission spectra depend not only on Floquet states (in particular their parity) but also on the initial state. Moreover, we have found nanostructures with more general symmetries that lead to more interesting emission patterns. Our studies suggest effective methods for generation of emission spectra with odd harmonics alone, even harmonics alone or both odd and even harmonics, and even with the quenching of all components. While previous works only studied the generation of even harmonics in addition to odd harmonics (i.e. both odd and even harmonics appear), our theory can be applied to more complex configurations.

Our theory can be used to explain a series of experimental results. The symmetry $Q = \Omega_0 \cdot \theta$ (with $\Omega_0$ a spatial inversion symmetry) prevents the appearance of even harmonics. If the symmetry is broken, additional even harmonic components appear. The appearance of even harmonic components in the experiments [31] is a consequence of breaking the spatial symmetry. In other types of experiments with helium or plasma plumes (containing nanoparticles, carbon nanotubes, etc) driven by a two-color laser [32], even harmonics appeared and were as strong as, or even stronger than, odd harmonics, in spite of the very weak second-harmonic driving field (for example, the ratio between the fundamental and second harmonics of the driving field was 12 : 1, or even 50 : 1). These experimental results can also be understood within our theory. The additional driving field of the form $\cos(2\omega_0 t)$ has different parity under the temporal transformation $\theta: t \rightarrow t + T/2$ compared with that for the field of the form $\cos(\omega_0 t)$. The second-harmonic driving term, despite being small, breaks the symmetry and

Figure 3. Emission spectra in triple dots with the same initial condition $\rho_{22} = 1.0$ but with different structures. The thin lines between the levels (in each dot) indicate the optical couplings between the dots with the value $G = 250$ meV. Panels (a) and (c) are the cases with chain structure. Panels (b) and (d) are the cases with loop structure. In (a) and (b), $E_1 - E_2 = 0.5$ meV, $E_2 - E_3 = 1$ meV; in (c) and (d), $E_1 - E_2 = E_2 - E_3 = 1$ meV. $\Gamma = 0.12$ GHz.
Figure 4. Emission spectra in quadruple dots with different structures/initial conditions. The thin lines between the levels (in each dot) indicate the optical couplings between the dots with value $G = 315$ meV. Panels (a) and (b) are the cases with the loop structure. Panels (c) and (d) are the cases with additional cross-couplings. $E_1 - E_2 = E_3 - E_4 = 0.4$ meV, $E_2 - E_3 = 1$ meV. $\Gamma = 0.12$ GHz.

Thus allows additional even harmonic components. Possible experiments with nanostructures have much more tunability. The symmetric configuration could be realized by a careful design of the nanostructures. For instance, the energy levels can be adjusted by changing the confining potential. They could also be tuned by applying appropriate gate voltages. The optical couplings among the dots could be tuned by changing the inter-dot barriers or the polarization of the incident field. In some situations, one may need weaker conditions. For instance, one may obtain the emission spectra with even harmonics only by using the configuration of triple-dot (loop structure) with degenerate levels for arbitrary inter-dot optical coupling. (It is a consequence of the symmetry generated by $Q = \Omega \cdot \theta$ with $\Omega: c_i \rightarrow -c_i$.) Another issue is the preparation of the appropriate initial state. Some initial states, such as those where electrons stay in the lowest-energy state, can be obtained easily. One may use some specially designed laser pulse to prepare other types of initial states [12]. As was seen in section 2, a small derivation of the initial conditions may not change the picture. For instance, the main features of emission spectra remain unchanged if the derivation of occupation probability for the initial states is less than 5%.
4. Summary

Based on the new theorem (on symmetry in the time domain and the energy spectrum domain) that we have proved, we provide methods to tailor photon emission patterns in driven nanostructures by tuning the symmetry of the system. In addition to emission spectra with only odd harmonics or both odd and even harmonics, we are able to obtain emission spectra with only even harmonics. Under suitable conditions, the photon emission can even be fully quenched. Our methods for tailoring emission spectra apply to general coupled nanostructures.

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