Model of generating either odd or even optical harmonics by varying the coupling parameters between source quantum dots

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We show that the high harmonics generation (HHG) in quantum dot structures can be changed from only odd orders to both odd and even orders by controlling the coupling parameters. We find that even harmonics cannot be generated in a multilevel system if the electronic levels can be classified into groups by the parity of oscillation modes (the Fourier components of Floquet states) and the radiative transitions within same group are forbidden. Otherwise both odd and even harmonics are possible. According to the two distinct dynamic behaviors of HHG, we obtain a straightforward judgement of HHG in any multilevel systems.

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The fundamental nature of high harmonics generation (HHG) is a process of frequency conversion induced by nonlinear interaction processes, which provides an important method to obtain coherent high-frequency radiation from low-frequency source. In the past decade, the HHG in atomic or molecular systems has been extensively studied due to its potential applications in x-ray coherent radiation and attosecond pulse lasers.[1–4] Most recently, the HHG study is extended to semiconductor quantum dots (QDs) and coupled QDs (CQDs), the so-called "artificial atoms and molecules."[5–7] One motivation of the study of the HHG in QDs is to find an efficient way of terahertz wave generation due to their controllable energy spectra and wave functions.

Generally, the HHG spectrum of an atomic or molecular system consists of only odd order harmonics. It is a quantum effect resulting from the geometric inversion symmetry of the potential, which conserves the parity of the eigenstates. Great efforts have been made for even-harmonic generation by breaking the inversion symmetry of atomic or molecular systems.[8–11] In real QD systems, especially in CQDs, the inversion symmetry is usually absent. However, it must be realized that the absence of geometric symmetry cannot ensure the even-harmonic generation. For example, there are only odd harmonics in the two-level QDs with or without the inversion symmetry.[5–7] In this work, we explore the dynamic behaviors of an electron in lateral-coupled triple-QD structures (TQDSs) under driving fields and reveal the selection rule of HHG in multilevel systems without the inversion symmetry.

In experiments, the energy-level structures and corresponding wave functions of TQDS can be modulated by controlling dot confinement and interdot tunneling. For a realistic lateral-coupled TQDS, the interdot tunneling is mainly determined by lateral confinement of each dot and distance between dots. Compared with the lateral confinement, the stronger vertical confinement gives an energy constant to the low-lying levels. Usually, in theoretical studies, a two-dimensional coupled parabolic potential [as shown in Fig. 1(a)] in effective mass approximation is adopted as follows:

$$V(x,y) = \frac{1}{2}m^* \min\left\{\omega_l^2\left[\left(x+\frac{1}{2}d_x\right)^2+y^2\right], \omega_m^2 \times[x^2+(y-d_y)^2], \omega_r^2\left[\left(x-\frac{1}{2}d_x\right)^2+y^2\right]\right\}, \quad (1)$$

where m^* is the effective mass of electron. ω_l, ω_m and ω_r are parabolic confining frequencies of the left, middle, and right dots, respectively. d_x is the distance between the left and right dots and the middle dot is d_y away from the others in the y direction. Using electronic states of the three single QDs $|\phi_j^k\rangle$ $(j=l,m,r)$ to construct a nonorthogonal basis, we can obtain the single-electron eigenstates $|i\rangle=\Sigma_{j,k}c_{ij}^k|\phi_j^k\rangle$ of the TQDS by solving a general eigenvalue problem.

We choose the effective mass of GaAs as $m^*=0.067m_e$ and the parameters as $\hbar\omega_l=6.0$ meV, $\hbar\omega_m=7.2$ meV, and $\hbar\omega_r=6.4$ meV. And two typical TQDS I with $d_x=80$ nm,

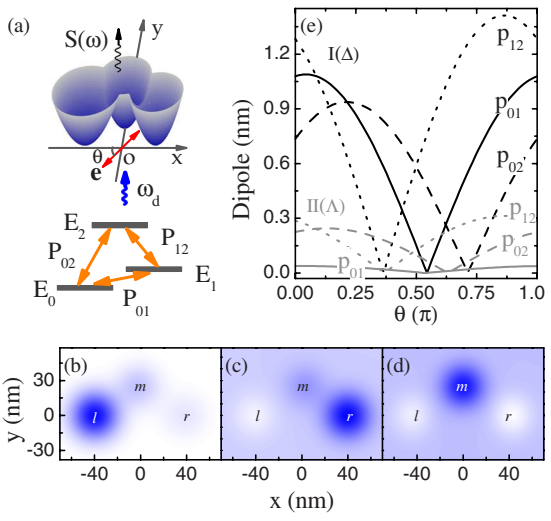

FIG. 1. (Color online) (a) Sketch of confining potential of a TQDS with driving field ω_d and coupling three-level structure. (b)–(d) Wave functions of the three levels for TQDS I. Marks l, m, and r indicate the dot centers, respectively. (e) The absolute value of projection of dipoles $\mathbf{p}_{ij}$ to spatial direction θ, black (gray) lines corresponding to TQDS I (II).

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d_y=25 nm and II with d_x=110 nm, d_y=25 nm are investigated. The calculated energies of the lowest three states of TQDS I (II) are, respectively, E_0=5.92(6.00) meV, E_1 =6.31(6.40) meV, and E_2=7.22(7.19) meV, as shown in Fig. 1(a). The next excited state is about 5 meV higher. A gigahertz linear-polarized field $\mathbf{E}(t)=F\mathbf{e}\cos\omega_d t$ with angular frequency $\omega_d=303.4\times10^9$ rad/s(0.2 meV) is used to drive the TQDS, where $\mathbf{e}$ is the polarization direction of the field. With such driving field frequency and moderate amplitude F, only the contribution of the lowest three states to HHG needs to be considered since the energy differences to higher states are much larger than ω_d and Rabi frequency of the field.

Since the size of the TQDS is quite smaller than the wavelength (about 6.2 mm) of the driving field, the dipole approximation is valid to describe the interaction of the field and the electron. The dipole between two states $\mathbf{p}_{ij}=\langle i|\mathbf{r}|j\rangle$ is determined by the distribution of the wave functions. In Figs. 1(b)–1(d), we plot the wave functions of states $|i\rangle$ (i =0,1,2) for TQDS I. It is clear that the electron is mainly localized in one dot but also extended to other two dots due to the tunneling effect. The extension of three wave functions leads to three dipoles $\mathbf{p}_{ij}$ with different polarized directions. In Fig. 1(e), we plot the projection of $\mathbf{p}_{ij}$ to spatial direction θ. From the maximums of three dipoles, it can be found that $\mathbf{p}_{ij}$ of TQDS I are along three direction $\theta=0.04\pi$, 0.21π, and 0.86π, respectively. Then the TQDS I is a Δ-type three-level structure. For TQDS II, $\mathbf{p}_{02}$ and $\mathbf{p}_{12}$ are along $\theta=0.13\pi$, and 0.87π, and $\mathbf{p}_{01}$ is quite close to zero due to the large distance d_x. In order to reveal the rules of HHG more clear, we set $\mathbf{p}_{01}$ to zero in the following discussions and then TQDS II is a Λ-type three-level structure.

In the dipole approximation, the interaction Hamiltonian of the microwave field with the three-level TQDS is written as

$$H(t)=\sum_{i=0}^{2}E_i|i\rangle\langle i|+(\hbar G_{01}|0\rangle\langle 1|+\hbar G_{02}|0\rangle\langle 2|+\hbar G_{12}|1\rangle\langle 2| + \text{H.c.})\cos\omega_d t, \quad (2)$$

where $G_{ij}=(eF/\hbar)\langle i|\mathbf{e}\cdot\mathbf{r}|j\rangle$ (i,j=0,1,2) are the Rabi frequencies. For structure Λ, $G_{01}=0$ and the transition $|0\rangle\rightarrow|1\rangle$ is cutoff. The time evolution of the system can be described by the Liouville equation[12] $\dot{\rho}(t)=-\frac{i}{\hbar}[H(t),\rho(t)]+\Xi\rho(t)$, where $\rho(t)=|\psi(t)\rangle\langle\psi(t)|$ is the density matrix and $\Xi\rho(t)$ describes the relaxation processes, which mainly come from spontaneous emission of phonons leading to decay rates Γ_{ij} of excited levels. In calculation, $\Gamma_{10}=\Gamma_{21}=\Gamma_{20}$ =0.5 GHz are used. $\rho(t)$ is obtained by numerically solving the Liouville equation. For a system with several dipoles along different directions, the evolution of total dipole can be analyzed by projecting all dipoles to any two mutually perpendicular directions $\mathbf{e}_\parallel$ and $\mathbf{e}_\perp$. Along any direction $\mathbf{e}'$, the dipole moment $\mu(t)=\langle\psi(t)|\mathbf{e}'\cdot\mathbf{r}|\psi(t)\rangle=\Sigma_{ij}\mu_{ij}\rho_{ij}(t)$ with μ_{ij} $=\langle i|\mathbf{e}'\cdot\mathbf{r}|j\rangle$. By the Fourier transformation of $\mu(t)$ along $\mathbf{e}_\parallel$ and $\mathbf{e}_\perp$, the polarization of emission field can be obtained and the power spectrum is $S(\omega)=|\int dt\,\exp(-i\omega t)\mu_\parallel(t)|^2+|\int dt\,\exp(-i\omega t)\mu_\perp(t)|^2$.

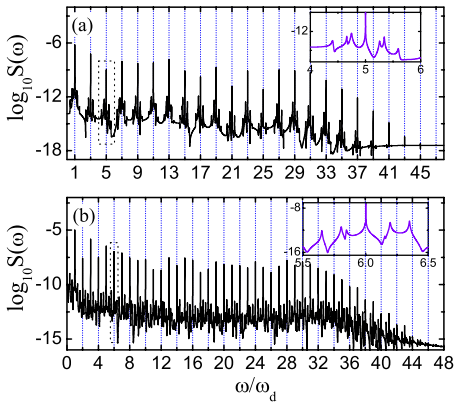

FIG. 2. (Color online) Logarithm of emission spectra as functions of ω/ω_d for structure (a) Λ and (b) Δ. Insets in (a) and (b): part of the emission spectrum enclosed in the dashed box.

In our calculation, we choose $\mathbf{e}$ of driving field at θ $=0.14\pi$ and 0.09π for structure Δ and Λ [see Fig. 1(e)], and use F=2.4 and 9.1 kV/cm, respectively, to make their Rabi frequencies approximately equal. Their emission spectra are shown in Figs. 2(a) and 2(b). Both of them consist of one series of harmonics at integer multiples of the fundamental frequency (ω_d). Around each harmonic, there are also three pairs of hyper Raman (HR)[13,14] peaks which is the typical property of three-level systems. The plateau of harmonics extends about to the 29th order, spanning the gigahertz region and reaching the terahertz region, for example, the 21st harmonic is about 1 THz. Comparing the intensity of harmonics in the plateau with that of fundamental frequency radiation (first order), the harmonic efficiencies of both structures are similar.

The most interesting phenomenon is that there are only odd harmonics in structure Λ but both of odd and even harmonics with comparable intensities in structure Δ. In fact, the further calculation shows that both odd and even harmonics can appear simultaneously in the spectra of structure Δ with comparable intensities no matter what polarized direction $\mathbf{e}$ of the driving field, even if it is perpendicular to one of dipoles. Although one dipole is not driven when $\mathbf{e}$ is perpendicular to it, the radiation can be transmitted through it, which results in even harmonics. Such nonsensibility on the polarized direction of the driving field suggests that the odd-even harmonic radiation is an intrinsic property of triple-coupling three-level systems. We will reveal the dynamic mechanism of odd-even HHG in the following discussions. We also inspect the polarization of the radiation, and find that it depends on the spatial configuration of the TQDS. The interesting phenomenon is that there are not only linear but also elliptical polarized harmonics in the spectrum, which is similar to the recent studies on multilevel molecular systems.[15] The polarization behaviors of HHG in multilevel TQDS will be investigated in details in another work.

Intuitively, the structure Δ provides only one more transition path than structure Λ does in processes of electron-field interaction. However, it leads to their distinct HHG behaviors. To reveal the HHG mechanism of the two structures we adopt the Floquet theorem to investigate the evolution of dipole moment $\mu(t)$. Based on the Floquet theorem, the

quantum state of a time-periodic Hamiltonian such as Eq. (2) is written as $\psi(t)=\Sigma_\nu A_\nu e^{-i\varepsilon_\nu t/\hbar}\Phi_\nu(t)$, where Floquet state $\Phi_\nu(t)$ is periodic functions of t and ε_ν is the corresponding quasienergy with $\nu=\alpha,\beta,\gamma$. By expanding $\Phi_\nu(t)$ further into Fourier series $\Phi_\nu(t)=\Sigma_{n=-\infty}^{\infty}\Sigma_{i=0}^{2}C_i^n(\varepsilon_\nu)e^{in\omega_d t}|i\rangle$, the solution of $i(d/dt)\psi(t)=H(t)\psi(t)$ can be converted to an eigenvalue-eigenvector problem for an infinite dimensional Floquet Hamiltonian in the basis $|i,n\rangle$,[16]

$$(H_F)_{ij}^{nk}=H_{ij}^{n-k}+n\hbar\omega_d\delta_{ij}\delta_{kn}, \tag{3}$$

where H_{ij}^{n-k} are Fourier components of $H(t)$ of Eq. (2), i (j) is the index of electron level and n (k) is the index of Fourier basis. By diagonalizing H_F, one can obtain the eigenvectors $\Phi_\nu^m(t)$ and the corresponding eigenvalues $\varepsilon_{\nu,m}=\varepsilon_\nu+m\hbar\omega_d$ with ε_ν denoting the three smallest absolute values of $\varepsilon_{\nu,m}$, i.e., the three quasienergies. For structure $\Delta(\Lambda)$, $\varepsilon_\nu=-0.358(-0.299),-0.163(-0.044),-0.008(0.296)\hbar\omega_d$. In fact, the space spanned by $|i,n\rangle$ is the direct product space of electronic levels and harmonic oscillation modes. Then $|C_i^n|^2$ is just the probability of electronic occupation on level i with mode $n\omega_d$. For a direction $\mathbf{e}'$, the evolution of dipole moment $\mu(t)$ can be analyzed by using the following formula:

$$\begin{aligned}\mu(t)&=\langle\psi(t)|\mathbf{e}'\cdot\mathbf{r}|\psi(t)\rangle\\&=\sum_{n,n'=-\infty}^{\infty}\sum_{\nu,\nu'=\alpha}^{\gamma}A_\nu^*A_{\nu'}e^{-i(n-n')\omega_d t}e^{i(\varepsilon_\nu-\varepsilon_{\nu'})t/\hbar}\\&\quad\times\sum_{i,i'=0}^{2}C_i^{n^*}(\varepsilon_\nu)C_{i'}^{n'}(\varepsilon_{\nu'})\mu_{ii'}=\sum_k C_k e^{-i\omega_k t}.\end{aligned} \tag{4}$$

The intensity of possible radiation frequency mode $\omega_k=(n-n')\omega_d-(\varepsilon_\nu-\varepsilon_{\nu'})/\hbar$ is determined by the coefficient C_k which is written as

$$\begin{aligned}C_k=\sum_{\nu,n}\{&[C_0^{n^*}(\varepsilon_\nu)C_2^{n'}(\varepsilon_{\nu'})+C_2^{n^*}(\varepsilon_\nu)C_0^{n'}(\varepsilon_{\nu'})]\mu_{02}\\&+[C_1^{n^*}(\varepsilon_\nu)C_2^{n'}(\varepsilon_{\nu'})+C_2^{n^*}(\varepsilon_\nu)C_1^{n'}(\varepsilon_{\nu'})]\mu_{12}\\&+[C_0^{n^*}(\varepsilon_\nu)C_1^{n'}(\varepsilon_{\nu'})+C_1^{n^*}(\varepsilon_\nu)C_0^{n'}(\varepsilon_{\nu'})]\mu_{01}\}.\end{aligned} \tag{5}$$

The high-order harmonics and HR radiation, respectively, correspond to the cases when $\nu=\nu'$ and $\nu\neq\nu'$.

The Fourier components of field-electron interaction terms of $H(t)$ in Eq. (2) provide the off-diagonal elements (coupling terms) of H_F. For structure Λ, the basis vectors $|0,n\rangle$ and $|1,n'\rangle$ are not coupled due to $G_{01}=0$. Then by rearranging the sequence of the bases, the matrix H_F of structure Λ can be reduced to a form of a direct sum of two matrices,

$$H_F=H_{F1}\oplus H_{F2}=\begin{pmatrix}\vdots&\vdots&\vdots&\vdots&\vdots&\\ \cdots & E_0+(n-1)\hbar\omega_d & 0 & \hbar G_{02}/2 & 0 & 0 & \\ \cdots & 0 & E_1+(n-1)\hbar\omega_d & \hbar G_{12}/2 & 0 & 0 & \cdots\\ \cdots & \hbar G_{02}/2 & \hbar G_{12}/2 & E_2+n\hbar\omega_d & \hbar G_{02}/2 & \hbar G_{12}/2 & \cdots\\ \cdots & 0 & 0 & \hbar G_{02}/2 & E_0+(n+1)\hbar\omega_d & 0 & \cdots\\ \cdots & 0 & 0 & \hbar G_{12}/2 & 0 & E_1+(n+1)\hbar\omega_d & \cdots\\ &\vdots&\vdots&\vdots&\vdots&\vdots&\end{pmatrix}$$

$$\oplus\begin{pmatrix}\vdots&\vdots&\vdots&\vdots&\vdots&\\ \cdots & E_0+n\hbar\omega_d & 0 & \hbar G_{02}/2 & 0 & 0 & \\ \cdots & 0 & E_1+n\hbar\omega_d & \hbar G_{12}/2 & 0 & 0 & \cdots\\ \cdots & \hbar G_{02}/2 & \hbar G_{12}/2 & E_2+(n+1)\hbar\omega_d & \hbar G_{02}/2 & \hbar G_{12}/2 & \cdots\\ \cdots & 0 & 0 & \hbar G_{02}/2 & E_0+(n+2)\hbar\omega_d & 0 & \cdots\\ \cdots & 0 & 0 & \hbar G_{12}/2 & 0 & E_1+(n+2)\hbar\omega_d & \cdots\\ &\vdots&\vdots&\vdots&\vdots&\vdots&\end{pmatrix}, \tag{6}$$

where H_{F1} and H_{F2} are in the subspaces with basis $\{\cdots|2,n\rangle,|0,n+1\rangle,|1,n+1\rangle,|2,n+2\rangle\cdots\}$ and $\{\cdots|2,n-1\rangle,|0,n\rangle,|1,n\rangle,|2,n+1\rangle\cdots\}$, respectively. In this direct sum space, the eigenvectors of H_F can be written as $|\varphi_i^1\rangle\oplus|\varnothing^2\rangle$ and $|\varnothing^1\rangle\oplus|\varphi_i^2\rangle$, where $|\varphi_i^{1(2)}\rangle$ are eigenvectors of $H_{F1(2)}$, and $|\varnothing^{1(2)}\rangle$ are zero vectors of the subspaces. For structure Δ, however, the vectors $|i,n\rangle$ and $|j,n'\rangle$ $(i\neq j$ and $n-n'=\pm1)$ are directly coupled so that H_F cannot be reduced to a form of a direct sum of two matrices as in the case of structure Λ. This can leads to their distinct dynamical behaviors.

In Fig. 3, we present the expansion coefficients C_i^n of Floquet state $\Phi_\alpha(t)$ for structures Λ and Δ. The nonzero C_i^n localize in the range of $n=-23-20$ for structure Λ and $n=-32-26$ for Δ as shown in Figs. 3(a) and 3(c). The range of

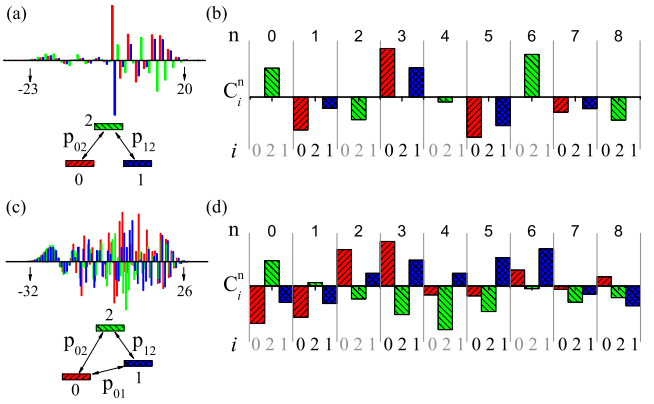

FIG. 3. (Color online) Fourier expansion coefficients and its zoomed in parts of Floquet state $\Phi_\alpha(t)$ of structure [(a) and (b)] Λ and [(c) and (d)] Δ.

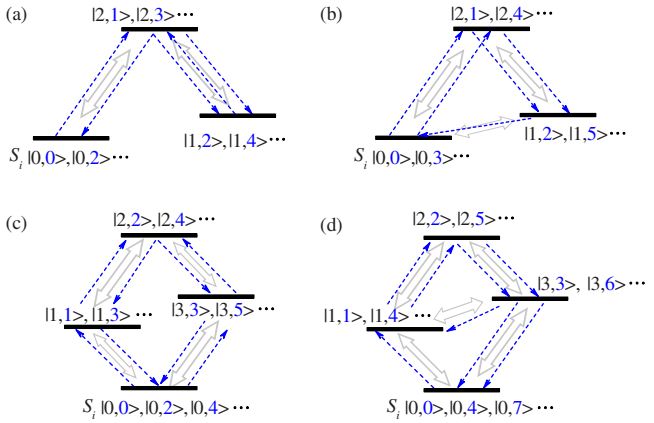

FIG. 4. (Color online) Scheme of the absorption processes for two typical three-level structures [(a) and (b)] and two four-level structures [(c) and (d)], respectively. Grey double arrows indicate the coupling between levels, and arrows with dashed line indicate possible transitions with absorbing a photon. Here, $|i,N\rangle$ is a virtual state representing electron absorbing N photons and occupying state $|i\rangle$.

the localization corresponds to the range of the emission spectrum and is determined by the Rabi frequencies. From Figs. 3(b) and 3(d), it can be seen that the C_i^n distributions of structures Λ and Δ are very different. For structure Λ, C_2^n is nonzero only for even n while C_0^n and C_1^n are nonzero only for odd n. This is in agreement with the fact that H_F of structure Λ can be reduced to the direct sum of H_{F1} and H_{F2}. According to Eq. (5), such distribution of C_i^n leads to the fact that $C_k=0$ for $n-n'=$even, thus the even harmonics are forbidden. In fact, the C_i^n distribution of structure Λ means that the three electronic levels can be divided into two groups which are $\{|0\rangle,|1\rangle\}$ and $\{|2\rangle\}$, respectively: one with oscillation modes of odd multiple ω_d and the other with even multiple ω_d. Since $\mathbf{p}_{01}=0$, only radiations between levels of different groups are allowed which lead to only odd HHG. For structure Δ, all three C_i^n $(i=0,1,2)$ are possible nonzero for a certain n because the H_F cannot reduce to block-diagonalized form, and then the above classification of levels is impossible. This makes C_k nonzero for both odd and even $n-n'$, i.e., both odd and even HHG are possible. It should be pointed out that the levels of structure Δ can be also classified as that of structure Λ if the polarization of the driving field is perpendicular to one of the dipoles, however the radiations within same group are allowed since $\mathbf{p}_{01}\neq 0$ which makes even harmonics possible.

By now we have discussed HHG of two typical three-level systems and can proceed to a general discussion of HHG in multilevel systems. Our three-level TQDS is a system without inversion symmetry, however there can be two different coupling structures leading to distinct HHG. It suggests that the broken symmetry cannot ensure the generation of even harmonics. From the discussion of Floquet theory, we see that the HHG of a specific coupling structure can be deduced from the distribution of vibration modes on each level. In fact, the analysis of HHG from Floquet theory can become clearer in a fully quantized description of the field interaction with multilevel systems.

From the viewpoint of full quantization, the whole process of a typical HHG is that the system absorbs n photons with energy $\hbar\omega_d$ and then radiates a photon with energy $n\hbar\omega_d$ which conserves the energy. In an intermediate step, the electron absorbs or emits a photon and transit to a virtual state which can be not energy conservation. In Figs. 4(a) and 4(b), we present the diagram of some possible absorption processes of two three-level structures discussed above. $|i,N\rangle$ is a virtual state representing electron absorbing N photons and occupying state $|i\rangle$. For structure Λ, from the initial state $|0\rangle$, the electron can only occupy the level $|2\rangle$ with odd photon absorptions and occupy $|0\rangle$ or $|1\rangle$ with even photon absorptions. Since the radiation of even harmonics from $|1\rangle$ to $|0\rangle$ is forbidden, there will be only odd harmonics. However for structure Δ, the electron can reach any level with both odd and even photon absorptions, then both odd and even harmonic generations become possible. This result can be extended to any coupling structures. Taking four-level structures in Figs. 4(c) and 4(d) as examples, the levels are divided into two groups $\{|0\rangle,|2\rangle\}$ and $\{|1\rangle,|3\rangle\}$ in loop-coupling case [Fig. 4(c)] and the radiative transitions within same group are forbidden, then there will be only odd harmonics. On the contrary, there will be both odd and even harmonics in the case of Fig. 4(d) which is also confirmed by our calculation.

In fact, such intuitive description is consistent with Floquet theory. As discussed in Ref. 16, the fully quantized basis $|i,N+n\rangle$ (N is the photon number) is approximately isomorphic to the basis $|i,n\rangle$ of H_F when N is very large. Then the classification of electronic levels according to the absorbed photon numbers is consistent with that according to the oscillation modes.

In conclusion, we have realized a control of HHG in TQDS which can be used as a possible design of terahertz source. By adjusting coupling structures of the multilevel systems, the HHG can be tuned from only odd order harmonics to both odd and even orders. By Fourier expanding the Floquet states, we found that there is only odd HHG if the electronic levels can be divided into two groups by the parity

of oscillation modes and the radiative transitions within same group are forbidden. Otherwise both odd and even HHG are possible. In a full-quantum viewpoint, the parity of the oscillation modes coincides with the parity of absorbed photon numbers, which leads to a straightforward judgement of the odd-even HHG in noninversion-symmetric multilevel systems. Our studies are important for understanding the dynamic behaviors of HHG in multilevel systems and helpful for the design controllable terahertz sources in nanostructures.

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