Theoretical investigation of terahertz optical sidebands in a single quantum dot

Koo-Chul Je and Seung-Han Park*
National Research Laboratory of Nonlinear Optics, Department of Physics, Yonsei University, Seoul 120-749, Korea
(Received 22 October 2007; published 19 December 2007)

We present a theoretical investigation on the dynamics of excitons, excitonic distribution functions, and absorption spectra of a single quantum dot driven by terahertz electric fields using the femtosecond laser pulse. In particular, we observe optical sidebands at frequencies of $2n\hbar\omega_L$ away from the exciton resonance due to coherent quantum superposition of Floquet states induced by the terahertz field. These frequency-doubled sidebands can occur at twice the frequency typically observed in bulk, quantum well, and superlattice structures. We have also obtained the oscillating excitonic densities for Floquet states as a function of the terahertz field strengths.

DOI: 10.1103/PhysRevB.76.245318 PACS number(s): 78.67.Hc, 71.35.−y, 78.20.Jq, 78.70.Gq

Modern technologies such as micromachining, nanotechnology, and ultrafast optical techniques have boosted a new area of terahertz research in physics, chemistry, and biology. As a result, terahertz time-domain biological spectroscopy, terahertz biomedical imaging, terahertz remote sensing, and terahertz satellite communications have been successfully demonstrated.[1–4] In efforts to further develop terahertz radiation and related technologies, semiconductor nanostructures have attracted much attention since their optical properties are strongly modified and can be controlled by a terahertz electric field. This is primarily due to the efficient coupling of the terahertz electric fields to collective intraband excitations in semiconductors.[5–7]

At present, fascinating new phenomena for generation of optical sidebands in terahertz-driven semiconductors are widely investigated. Terahertz optical sidebands on a near-infrared (NIR) carrier beam are produced when the NIR field is resonant with an excitonic transition and a terahertz electric field resonantly couples to intersubband transitions or two excitonic transitions.[5–16] These optical phenomena, which have been recently generated in various semiconductor systems (i.e., bulk GaAs,[5] quantum wells,[6–11] and linear arrays of quantum dots[12]) using intense pulses of coherent terahertz radiation from a free electron laser, are mainly due to mixing of optical and terahertz frequencies.[6–10] Generating both even and odd sidebands requires breaking inversion symmetry in symmetric coupled quantum wells[5,10] and only even sidebands were observed from confined magnetoexcitons in multiple quantum wells.[11]

The dynamics including the terahertz field were investigated by introducing a moving coordinate frame with electron-hole operators in the semiconductor Bloch equations.[17,18] The resonant tunneling through time-dependent barriers and the nonperturbative method were also adopted to explain the sideband generation in quantum wells.[19] In order to understand the formation of optical sidebands in a single quantum dot, a Hamiltonian describing excitons in the presence of terahertz electric fields with a second-quantized formalism was employed.[20,21] The excitons consist of the products of electron and hole creation operators, of which states are determined by the excitonic stationary states and energy levels obtained using the Floquet theorem and the excitons have bosonic behavior at low densities. Thus, it is reported that the Floquet states strongly affect the formation of the optical sidebands.[21,22]

In this paper, we describe the dynamics of excitons which consist of the Floquet states of a single parabolic quantum dot system in the presence of time-dependent fields with a second quantization. We present that optical sidebands in the isolated single quantum dot can be generated at frequencies $2n\hbar\omega_L$ away from the excitonic resonance driven by a terahertz field with frequency ω_L, where n is an integer. We show that the generation of optical sidebands results from coherent quantum superposition of generated Floquet states by the terahertz fields. We also obtain the oscillating excitonic densities of the Floquet states with increasing the strength of the terahertz field.

The Hamiltonian for an exciton in a single quantum dot structure driven by the terahertz field of intensity F_0 and frequency ω_L can be written as

$$H_{ex} = H_0 + ezF_0 \cos \omega_L t, \tag{1}$$

where H_0 is the effective-mass Hamiltonian for an electron-hole pair in a quantum dot and z is the relative distance between the electron and hole. The Hamiltonian of an interacting electron-hole pair is given by

$$H_0 = \frac{\hbar^2 p_e^2}{2m_e} + \frac{\hbar^2 p_h^2}{2m_h} - \frac{e^2}{\epsilon_0 |r_e - r_h|} + \frac{1}{2}\omega_0^2 (m_e r_e^2 + m_h r_h^2), \tag{2}$$

where p_e and p_h are the electron and hole momenta and ϵ_0 is the static dielectric constant. The last terms are the parabolic confinement potential of the electron and hole with a common parabola frequency of $\omega_0 \sim \hbar / m_r R_0^2$ (R_0 is the quantum dot radius and m_r is the reduced mass).

Using the coordinate $r = r_e - r_h$ and $R = \frac{m_e}{M} r_e + \frac{m_h}{M} r_h$, the effective Hamiltonian is separable in terms of the relative coordinate $\mathbf{r}$ and center-of-mass coordinate $\mathbf{R}$, that is, the relative momentum $\mathbf{p} = \hbar \nabla_r / i$ and the center-of-mass momentum $\mathbf{P} = \hbar \nabla_R / i$ as

$$p_e = p + P\frac{m_e}{M}, \quad p_h = -p + P\frac{m_h}{M}. \tag{3}$$

Substituting Eq. (3) in the effective Hamiltonian yields

$$H_{ex} = \frac{\hbar^2 P^2}{2M} + \frac{1}{2}MR^2\omega_0^2 + \frac{\hbar^2 p^2}{2m_r} - \frac{e^2}{\epsilon_0|r|} + \frac{1}{2}m_r r^2\omega_0^2 + ezF_0 \cos\omega_L t, \tag{4}$$

where $M=m_e+m_h$ is the total mass and $m_r=m_e m_h/M$. Therefore, the exciton wave function consists of the relative coordinate part and center-of-mass coordinate part which is the wave function of a harmonic oscillator and the exciton energy has the sum of the relative coordinate part and center-of-mass coordinate part. The eigenfunctions and eigenenergies of the Hamiltonian H_0 in the single parabola quantum dot are described in detail using the exact numerical diagonalization method with the basis set of the Hermite polynomials.[23]

In the presence of time-dependent terahertz fields with the z direction, the Hamiltonian is periodic in time with period $T=2\pi/\omega_L$, $H_{ex}(t+T)=H_{ex}(t)$. Thus, according to the Floquet theorem, the solution of the Schrödinger equation with the Hamiltonian periodic in time is given by[22]

$$\Psi(z,t) = \sum_{\alpha} e^{-i\epsilon_\alpha t/\hbar} \sum_{n=-\infty}^{\infty} C_{\alpha,n} e^{-in\omega_L t}\phi_\alpha(z), \tag{5}$$

where $\Psi(z,t)$ denotes the quasiexciton states. The excitonic states, $\phi_\alpha(z)$, without time-dependent interaction are chosen as the basis set with a finite number of basis states and obtained by the numerical matrix diagonalization of the relative Hamiltonian matrix, $\langle\alpha|H_0|\alpha'\rangle$.[23] The coefficients, $C_{\alpha n}$, are the nth Fourier coefficients in the expansion of α Floquet states and are called the Floquet modes of α state. The coefficients are determined by the infinite Floquet matrix eigenvalue equation as follows:

$$\sum_{\alpha,n}\left[(E_\alpha - \varepsilon - n\hbar\omega_L)\delta_{n,n'}\delta_{\alpha,\alpha'} + \frac{eF_0}{2}\aleph_{\alpha,\alpha'}(\delta_{n',n+1} + \delta_{n',n-1})\right]C_{\alpha,n} = 0, \tag{6}$$

where E_α denotes the eigenenergy of H_0 and $\aleph_{\alpha,\alpha'}=\langle\phi_\alpha|z|\phi_{\alpha'}\rangle$ are the dipole matrix elements coupling zero-field states. The quasiexciton wave functions $\Psi(z,t)$ are solved nonperturbatively by diagonalizing the Floquet matrix.[7,14] In practice, the Floquet matrix must be truncated up to $\pm N$ photons. N can be made arbitrarily large for an arbitrarily precise result at high-field strengths.[24] In this case, the eigenvalues and eigenvectors of Eq. (6) are numerically obtained by

$$\varepsilon_{\alpha,m} = E_\alpha + m\hbar\omega_L, \quad C^n_{\alpha,m} = J_{m+n}\left(\frac{eF_0 z}{\hbar\omega_L}\right). \tag{7}$$

The quasienergies $\varepsilon_{\alpha,m}$ refer to the m-photon representation with the corresponding eigenvectors $C^n_{\alpha,m}$ and J_n denotes the nth-order Bessel function of the first kind. Note that quantum dot models, including the parabolic model, affect the excitonic eigenenergy E_α, but $\Delta\varepsilon(=\varepsilon_{\alpha,m\pm1}-\varepsilon_{\alpha,m})$ does not vary. The total energy of the quasiexcitons is given by

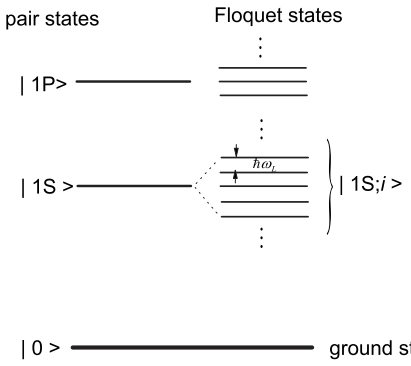

FIG. 1. Scheme of the Floquet states for pair states in a single quantum dot.

$$E_{ex}(\alpha,m,K) = \frac{\hbar^2 K^2}{2M} + E_g - \varepsilon_{\alpha,m}. \tag{8}$$

The eigenfunction of the most general electron-hole pair state is given by $A^+_\alpha|0\rangle=\Sigma c_{kk'}a^+_k b^+_{k'}|0\rangle$ where $a^+_k(a_k)$ and $b^+_{k'}(b_{k'})$ are the electron and hole creation (annihilation) operators, and $|0\rangle$ is the vacuum state for pair generation. One obtains $A^+_\alpha=\Sigma_{k,k'}\phi_\alpha(r)a^+_k b^+_{k'}$ and denoted original exciton states, $\phi_\alpha(r)$, are obtained by diagonalizing the matrix $\langle\alpha|H_0|\alpha'\rangle$. To investigate the excitonic dynamics in a single quantum dot under the terahertz fields, we describe a Hamiltonian in terms of excitonic operators of $A^\dagger_{\alpha,n}$ $(A_{\alpha,n})$ which create (destroy) excitons with Floquet states (α,n) as follows:

$$H = \sum_{\alpha,n}\hbar\Omega_{\alpha,n}A^\dagger_{\alpha,n}A_{\alpha,n} - \sum_{\alpha,n} d_{\alpha n}E(t)(A^\dagger_{\alpha,n} + A_{\alpha,n}), \tag{9}$$

where $\hbar\Omega_{\alpha,n}=\varepsilon_{\alpha,n}$ denote the quasienergies and the interband optical dipole moment $d_{\alpha n}$ couples the excitonic system to the external optical field $E(t)$. The time-dependent optical dipole moment in the spectral Floquet representation is given by

$$d_{\alpha n} = e^{-i(\varepsilon_\alpha + n\omega_L)t/\hbar}C_{\alpha,n}\int dz\langle\phi_\alpha(z,t=0)|r|0\rangle = e^{-i(\varepsilon_\alpha + n\omega_L)t/\hbar}C_{\alpha,n}\mu_{\alpha 0}, \tag{10}$$

where $\mu_{\alpha 0}=\langle\phi_\alpha(z,t)|r|0\rangle$. The Floquet states of pair states in a single quantum dot are schematically shown in Fig. 1. The indices 0 and α refer to the ground and pair states and $e=(\alpha,n)$ states denote the Floquet states. The excitonic operators obey the Pauli commutation rules[20,21]

$$[A_{\alpha,n},A^\dagger_{\alpha',n'}] = (\delta_{\alpha\alpha'}\delta_{nn'} - 2\chi A^\dagger_{\alpha',n'}A_{\alpha,n}),$$

$$[A_{\alpha,n},A_{\alpha',n'}] = [A^\dagger_{\alpha,n},A^\dagger_{\alpha',n'}] = 0, \tag{11}$$

where χ parameters describe phase-space filling.

We introduce the definitions of interband and intraband correlations as follows:

$$X_{e0} \equiv \langle A_{\alpha,n}\rangle, \quad X_{ee'} \equiv \langle A^\dagger_{\alpha,n}A_{\alpha',n'}\rangle. \tag{12}$$

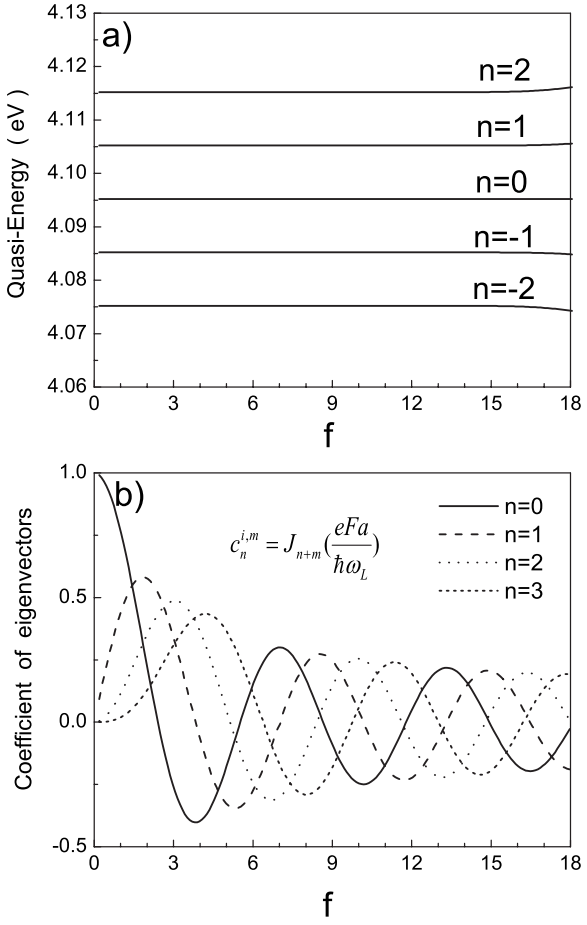

FIG. 2. (a) Quasienergies of $1S$ exciton with ladder indices $n=0, \pm 1, \pm 2$ and (b) their eigenvectors with $m=0$ as a function of terahertz field strengths.

Then, the polarization $P(t)$ and excitonic distribution function $F_e(t)$ can be defined as $\Sigma_e d_{e,0} E(t)(X_{e0}^* + X_{e0})$ and X_{ee}, respectively, and the absorption coefficient is obtained by $\alpha(\omega) \propto \mathrm{Im}\left[\frac{P(\omega)}{E(\omega)}\right]$. The resulting dynamical equations of motion for X_{e0} and $X_{ee'}$ are

$$i\frac{dX_{e0}}{dt} = -(\Omega_e - i\gamma_2)X_{e0} + \frac{d_e E(t)}{\hbar}(1 - F_e) - \sum_{e'} \frac{2d_{e'0}E(t)}{\hbar} X_{e'e},$$

$$i\frac{dX_{ee'}}{dt} = (\Omega_e - \Omega_{e'} - i\gamma_{ee'})X_{ee'} + \frac{E(t)}{\hbar}(d_{e'}X_e^* - d_e X_{e'}), \quad (13)$$

where interband and intraband dephasings γ_2 and $\gamma_{ee'}$ have been included phenomenologically. If $e=e'$, then $\gamma_{ee'}=T_1$ (excitonic population decay time), whereas if $e \neq e'$, then $\gamma_{ee'}=T_2$ (intraband dephasing time). It was found to enhance numerical stability without changing our results. We note that, to second order in the optical field, the exciton-exciton interaction does not appear in the dynamical equations.

The system we are modeling is a single CdS quantum dot with radius $R_0=1.3$ nm. The following parameters in the single CdS quantum dot were used for the numerical calculation:[25] electron mass $m_e(\mathrm{CdS})=0.18m_0$, hole mass $m_h(\mathrm{CdS})=0.7m_0$ (m_0 is the electron mass in free space), and dielectric constant $\epsilon_d(\mathrm{CdS})=8.5$. We modeled the sample to be optically excited by a 20 fs Gaussian pulse and employed an interband dephasing time of $\gamma_2=0.5$ ps and intraband dephasing time $\gamma_{ee}=\infty$ and $N=20$ photons.

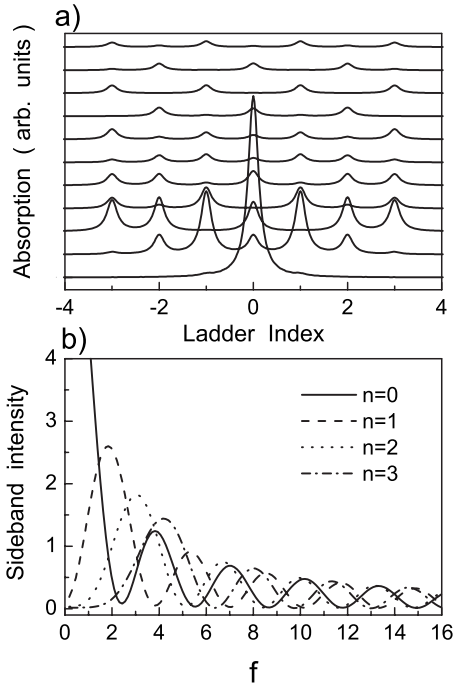

FIG. 3. (a) Absorption spectra and (b) sideband intensities with ladder indices of $n=0,1,2,3$ as a function of terahertz field strengths f. From bottom to top: $F_0=10,100,200,300,400,500,600,700,800,900,1000$ kV/cm. ($f=0.185,1.853,3.706,5.559,7.412,9.265,11.118,12.971,14.824,16.677,18.536$.)

In Fig. 2(a), we present the quasienergy ladders of the $1S$ exciton state obtained by numerical diagonalization of the Floquet matrix as a function of driving terahertz field strength of $f=eF_0R/\hbar\omega_L$. The quasienergies are almost equally spaced by $\hbar\omega_L$ up to $f \simeq 17$. Photon representations with large photon numbers are needed for more accurate results of the Floquet states as the intensity of the terahertz field increases. The equally spaced quasienergies will present the Stark ladders on the optical spectrum as shown in semiconductor superlattices.[6,8,9,16] Figure 2(b) shows results of the calculation for the $n=0,1,2,3$ Fourier components of the eigenvector, corresponding to the $1S$ exciton level in the zero-photon representation of $N=20$ photons, as a function of the strength of the external terahertz field, f. The coefficients of eigenvectors $n=0,1,2,3$ with zero-photon representation behave like the Bessel functions of J_0, J_1, J_2, J_3, respectively. The Bessel functions are approximate solutions of the Floquet equation for intense driving terahertz fields. Therefore, the Bessel functions are closely related to the optical transition of the Floquet states induced by the terahertz field in the single quantum dot.

Figure 3(a) displays the excitonic absorption spectra of the single quantum dot for different f with a fixed terahertz frequency of $\hbar\omega_L=10$ meV. All optical sidebands are re-

placed with the $\hbar\omega_L$ spacing. The generation of optical sidebands is due to the absorption of free $1S$ excitons. Especially, for high f, there are regimes where even and odd sidebands do not exist simultaneously. Consequently, the optical sidebands appear with spacing $2\hbar\omega_L$ in the high $f \geq 11$ regime. Figure 3(b) shows the sideband intensities for ladder indices $n=0,1,2,3$ as a function of f. This behavior is closely related to the dipole moment of the Floquet states. Exciton peak with $n=0$ is changed by the zero-order Bessel functions. The sideband intensities with ladder index n are changed by the absolute value of the n-order Bessel function. Odd sidebands appear around f for $J_n(f)=0$ with even integers n and even sidebands appear around f for $J_n(f)=0$ with odd integers n. This means that the intensities of the optical odd sidebands have maximum values around f where the intensities of the optical even sidebands are zero. Consequently, the optical sidebands can be spaced by $2\hbar\omega_L$ over $f \geq 11$, which is similar to intersubband optical transitions induced by the terahertz field in quantum wells[26] and relates to the symmetry of the system under space inversion. The generation of optical sidebands can be explained by coherent quantum superposition of generated Floquet states by the terahertz field.

Figure 4 shows the distributions of the $1S$ quasiexcitons at $t=2$ ps for different terahertz intensities with the laser pulse tuned to the undriven $1S$ exciton energy. The distribution of excitons decreases with increasing terahertz intensity since the stronger terahertz field causes more nonresonant excitation. This change in exciton distribution is closely related to the optical dipole moment of the Floquet states, which are determined by solutions of the Floquet equation.

In summary, we have studied the dynamics of excitons using Floquet theorem in a single quantum dot driven by terahertz fields. The generation of optical sidebands and oscillating excitonic densities for the Floquet states results from coherent quantum superposition of generated Floquet states by the terahertz fields which create excitonic quasienergy levels with spacing $\hbar\omega_L$ and excitonic Floquet states such as Bessel functions. The intensities of the optical sidebands are strongly coupled to the terahertz field strength.

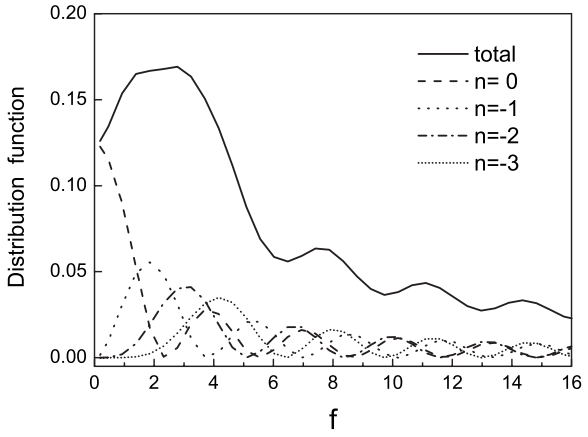

FIG. 4. Quasiexciton densities with ladder indices of $n=0$, -1, -2, -3 and total exciton density (solid line) at $t=2$ ps as a function of terahertz field strengths.

This research was supported by the Ministry of Science and Technology of Korea through the National Research Laboratory Program (M1-0203-00-0082), and by the Brain Korea 21 project of the Ministry of Education.

*shpark@yonsei.ac.kr

[1] P. H. Siegel, IEEE Trans. Microwave Theory Tech. **50**, 910 (2002); M. Toyoshima, J. Opt. Netw. **4**, 300 (2005).
[2] B. Fischer, M. Hoffmann, H. Helm, R. Wilk, F. Rutz, T. Kleine-Ostmann, and M. Koch, Opt. Express **13**, 5205 (2005).
[3] B. B. Hu and M. C. Nuss, Opt. Lett. **20**, 1716 (1995).
[4] T. Löffler, K. Siebert, S. Czasch, T. Bauer, and H. G. Roskos, Phys. Med. Biol. **47**, 3847 (2002).
[5] M. A. Zudov, J. Kono, A. P. Mitchell, and A. H. Chin, Phys. Rev. B **64**, 121204(R) (2001).
[6] S. G. Carter, V. Ciulin, M. S. Sherwin, M. Hanson, A. Huntington, L. A. Coldren, and A. C. Gossard, Appl. Phys. Lett. **84**, 840 (2004).
[7] M. Y. Su, S. G. Carter, M. S. Sherwin, A. Huntington, and L. A. Coldren, Phys. Rev. B **67**, 125307 (2003).
[8] M. Y. Su, S. G. Carter, M. S. Sherwin, A. Huntington, and L. A. Coldren, Appl. Phys. Lett. **81**, 1564 (2002).
[9] S. G. Carter, V. Ciulin, M. Hanson, A. S. Huntington, C. S. Wang, A. C. Gossard, L. A. Coldren, and M. S. Sherwin, Phys. Rev. B **72**, 155309 (2005).
[10] C. Phillips, M. Y. Su, M. Hanson, M. S. Sherwin, J. Ko, and L. Coldren, Appl. Phys. Lett. **75**, 2728 (1999).
[11] J. Kono, M. Y. Su, T. Inoshita, T. Noda, M. S. Sherwin, S. J. Allen Jr., and H. Sakaki, Phys. Rev. Lett. **79**, 1758 (1997).
[12] M. Pacheco and Z. Barticevic, Phys. Rev. B **55**, 10688 (1997).
[13] A. P. Jauho and K. Johnsen, Phys. Rev. Lett. **76**, 4576 (1996).
[14] K. Johnsen and A. P. Jauho, Phys. Rev. Lett. **83**, 1207 (1999).
[15] S. G. Carter, V. Birkedal, C. S. Wang, L. A. Coldren, A. V. Maslov, D. S. Citrin, and M. S. Sherwin, Science **310**, 651 (2005).
[16] K.-C. Je, S.-H. Park, and Y. Kim, Phys. Rev. B **64**, 075111 (2001).
[17] S. Hughes and D. S. Citrin, Phys. Rev. B **59**, R5288 (1999).
[18] S. Hughes and D. S. Citrin, Phys. Rev. B **58**, R15969 (1998).
[19] S. Hughes, Phys. Rev. B **63**, 153308 (2001); S. Hughes and D. S. Citrin, Opt. Lett. **25**, 493 (2000).
[20] M. M. Dignam and M. Hawton, Phys. Rev. B **67**, 035329 (2003).
[21] M. Hawton and D. Nelson, Phys. Rev. B **57**, 4000 (1998).
[22] J. H. Shirley, Phys. Rev. **138**, B979 (1965).
[23] O. Ciftja and M. Faruk, J. Phys.: Condens. Matter **18**, 2623 (2006).
[24] K.-C. Je, T.-Y. Hwang, H.-Y. Ju, and S.-H. Park, J. Korean Phys. Soc. **49**, 2373 (2006).
[25] *Semiconductors. Physics of Group IV Elements and III-V Compounds*, edited by O. Madelung, M. Schultz, and H. Weiss, Landolt-Börnstein, New Series, Group III, Vol. 17, Pt. a (Springer, Berlin, 1982).
[26] M. Y. Su, C. Phillips, J. Ko, L. Coldren, and M. S. Sherwin, Physica B **272**, 438 (1999).