Interband optical-absorption spectra of a finite quantum dot superlattice in a cylindrical nanowire

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Theoretical calculations of the interband optical-absorption spectra of a finite quantum dot superlattice in a wire (QDSLW) in the presence of a homogeneous dc or terahertz field are presented. The QDSLW has miniband structures of the electron states as general quantum well superlattices do and shows the two features common to one-dimensional structures, viz., strong excitonic absorption below and weak absorption above the band edge. The applied dc field causes a redshift of the location and a decrease in height of the main excitonic peak and leads to an increase in height of the minor peak next to the main peak. In the presence of a terahertz field, the Autler-Townes splitting of the main excitonic peak and the emergence of one-photon and two-photon gain peaks on the spectra are demonstrated. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2131190]

I. INTRODUCTION

Advances in material growth technologies have led to fabrication of various low-dimensional semiconductor nanostructures which have diverse potential applications in electronics and photonics. Quantum wells, quantum wires (QWs), quantum dots, and superlattices (SLs) have been investigated as low-dimensional objects for decades. Their transport and optical properties are well understood.[1–4] In recent years, the dynamic properties of electrons and holes in low-dimensional semiconductors driven by terahertz fields have attracted considerable attention. Many fascinating phenomena, e.g., photoassisted tunneling, dynamic localization, absolute negative conductivity, ac Stark effect, dynamic Franz-Keldysh effect, formation of sidebands, and nonlinear terahertz absorption and reflection were explored.[5–11] The investigation of the terahertz field effect on designed nanostructures constitutes an important future development. Recently, a structure consisting of a series of quantum dots interlaced along a nanowire has been grown.[12–16] This quantum dot superlattice structure has characteristics of quantum dots, QWs, and SLs. As the band offset of different materials confines the carriers in one of the quantum dots, the electron states have zero-dimensional (0D) characteristics of a quantum dot. However, by tunneling between adjacent quantum dots the carriers can travel along the wire axis. Thus, a miniband electronic structure of a SL is formed. Due to strong lateral confinement, the carrier motion in the plane perpendicular to the wire axis is confined so that the system has characteristics of one-dimensional (1D) structures. This structure has a wide range of potential applications in nanolasers, nanobarcodes, 1D waveguides, resonant tunneling diodes, and thermoelectric systems.[12–17]

In this paper, it is intended to investigate a system consisting of a finite quantum dot superlattice embedded in the central part of a finite nanowire. This quantum dot superlattice in a wire (QDSLW) structure is more practical than an infinite quantum dot superlattice, as is usually assumed. We calculate the linear optical-absorption spectra of a QDSLW in the presence of dc or terahertz electric fields. The optical-absorption coefficient is obtained by employing equation-of-motion techniques.[3] The equation of motion is extended to include the finite number of quantum dot potentials for electrons and holes. The translational invariance breaking down of the system is taken into account which is in contrast to the general approach of solving the electron and hole relative motion. This approach has been used to discuss the effects of external dc or ac fields on the interband optical absorption in quantum wells, QWs, and quantum rings.[18–23]

II. THEORY OF OPTICAL ABSORPTION IN A QDSLW

A finite cylindrical QW with infinite potential on the lateral and the two ending walls and finite modulating potential in the central part due to alloy alternation is shown in Fig. 1. The infinite potential at the lateral wall of the thin nanowire is adequate for freestanding nanowires.[14] In the

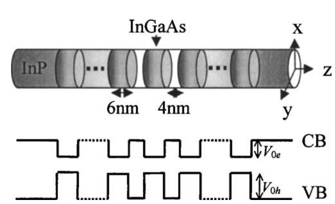

FIG. 1. Schematic representation of the QDSLW geometry and the corresponding potential profiles of electrons and holes. A superlattice of 20 $In_{0.53}Ga_{0.47}As$ quantum dots (InP barriers) embedded in the central part of a 300-nm-long rod of InP nanowire.

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presence of a homogeneous dc or terahertz field along the wire axis, the system is probed by a short infrared optical pulse.

The interband optical-absorption coefficient $\alpha(\omega)$ of the system can be obtained from the real and imaginary parts of the complex dielectric function $\epsilon(\omega)=\epsilon'(\omega)+i\epsilon''(\omega)$ as in Ref. 4,

$$\alpha(\omega)=\frac{\omega}{nc}\epsilon''(\omega), \tag{1}$$

where ω is the angular frequency and $n=\sqrt{(1/2)\{\epsilon'(\omega)+\sqrt{[\epsilon'(\omega)]^2+[\epsilon''(\omega)]^2}\}}$ is the index of refraction. The complex dielectric function $\epsilon(\omega)$ is related to the linear optical susceptibility $\chi(\omega)$ via $\epsilon(\omega)=1+4\pi\chi(\omega)$. The linear optical susceptibility $\chi(t;t')$ in time domain relates the incident optical field $E(t')$ with the induced optical polarization $P(t)$ by

$$P(t)=\int_{-\infty}^{t}\chi(t;t')E(t')dt'. \tag{2}$$

The optical pulse is assumed to be polarized along the QDSLW and has a Gaussian slow varying envelope $E(t)=E_0\exp(-t^2/\sigma^2)$ with σ the pulse width.

The optical polarization $P(t)$ of the QDSLW can be calculated from the time-dependent electron-hole envelope wave function $\Psi(z_e,z_h;t)$ as

$$P(t)=d^*\int_{-L/2}^{L/2}\Psi(z,z;t)dz, \tag{3}$$

where d^* is the complex conjugate of d (the projection of the interband dipole matrix element on the direction of the optical polarization), z_e and z_h are the coordinates of the electron and hole along the nanowire, respectively, and L is the length of the nanowire. Using the effective-mass approximation and the two-band model, the envelope function Ψ satisfies the inhomogeneous Schrödinger equation:

$$i\hbar\frac{\partial\Psi}{\partial t}=\hat{H}\Psi-d\delta(z_e-z_h)\delta(\rho)E(t)-i\hbar\gamma\Psi, \tag{4}$$

with

$$\hat{H}=-\frac{\hbar^2}{2m_e}\frac{\partial^2}{z_e^2}-\frac{\hbar^2}{2m_h}\frac{\partial^2}{z_h^2}+U_e(z_e)+U_h(z_h)+eF(z_e-z_h)-V_{\mathrm{Coul}}(z_e-z_h), \tag{5}$$

where $\hat{H}$ is the Hamiltonian of the electron-hole pair, $\hbar$ the reduced Planck constant, δ the Dirac's delta function, ρ the radial coordinate of the relative motion of the electron and the hole in the plane perpendicular to the wire axis, γ the dephasing constant of the optical polarization resulted from various scatterings in the system, and m_e and m_h the masses of the electron and hole, respectively. $U_e(z_e)$ and $U_h(z_h)$ denote the electron and hole confining potentials along the QDSLW, respectively. Moreover, e is the absolute value of the electron charge and V_{Coul} is Coulomb interaction between the electron and hole. The applied dc and ac electric fields along the QDSLW is $F=F_{\mathrm{dc}}+F_{\mathrm{ac}}\cos(\Omega t)$. The differences of the effective masses in the barrier layers and in the well layers are neglected, thus the barrier localization and inversion effect[24] due to the different carrier masses in barriers and wells are not accounted for here.

The infinite potential at the lateral wall of the thin nanowire implies the Bessel function of zero order for the envelope of the lateral ground-state wave function. Considering the fundamental electron and hole subbands only, the quasi-1D Coulomb interaction $V_{\mathrm{Coul}}(z_e-z_h)$ is obtained from the average of the three-dimensional (3D) Coulomb interaction with the lateral ground state:[25,26]

$$V_{\mathrm{Coul}}(z_e-z_h)=\kappa\int_0^R\int_0^R\int_0^{2\pi}\int_0^{2\pi}\frac{\rho_e\rho_h J_0^2(\alpha_0\rho_e/R)J_0^2(\alpha_0\rho_h/R)}{\sqrt{(z_h-z_e)^2+\rho_e^2+\rho_h^2-2\rho_e\rho_h\cos(\theta_h-\theta_e)}}d\rho_e d\rho_h d\theta_e d\theta_h, \tag{6}$$

where $\kappa=e^2/[4\pi^3\epsilon R^4 J_1^4(\alpha_0)]$, ϵ the dielectric constant, R the radius of the nanowire, $\theta_{e(h)}$ and $\rho_{e(h)}$ the polar angle and radial coordinate of electron (hole), respectively, J_0 and J_1 the Bessel functions, and α_0 the zero of J_0.

Since we are interested in the response to the continuous-wave (cw) optical fields, we perform a Fourier transform of Eq. (2) and obtain

$$\tilde{P}(\omega)=\sum_{n=-\infty}^{\infty}\tilde{\chi}_n(\omega;\omega-n\Omega)\tilde{E}(\omega-n\Omega). \tag{7}$$

The coefficients $\tilde{\chi}_n(\omega;\omega-n\Omega)$ in Eq. (7) are related to $\chi(t;t')$ as described in Ref. 27. To extract $\tilde{\chi}_n(\omega;\omega-n\Omega)$, the method proposed by Maslov and Citrin[27] is adapted here. By setting N short optical pulses $E_l(t)$ arriving at well-defined phases of the terahertz field, where $E_l(t)=E[t-(lT/N)]$ with $l=0,1,\ldots,N-1$ and $T=2\pi/\Omega$ the period of the terahertz field, and solving the inhomogeneous Schrödinger equation for each short optical pulse $E_l(t)$ to obtain each associated $P_l(t)$, the susceptibility $\tilde{\chi}_0$ in the frequency domain is then calculated as

$$\tilde{\chi}_0(\omega;\omega)=\frac{1}{N}\sum_{l=0}^{N-1}\frac{\tilde{P}_l(\omega)}{\tilde{E}_l(\omega)}, \tag{8}$$

where $\tilde{E}_l(\omega)$ and $\tilde{P}_l(\omega)$ are the Fourier transform of $E_l(t)$ and $P_l(t)$, respectively.

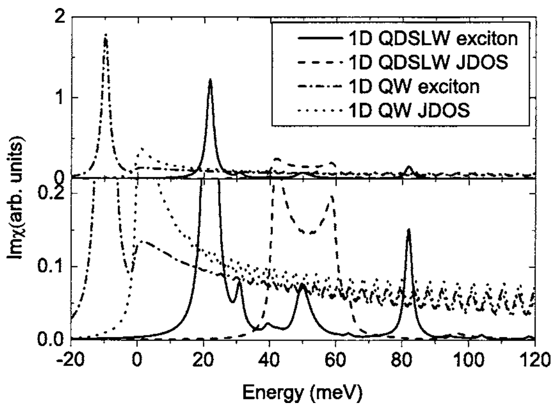

FIG. 2. Comparison of the exciton-dominated optical-absorption spectrum and joint density of states in an ideal 1D QDSLW and in a plain 1D QW.

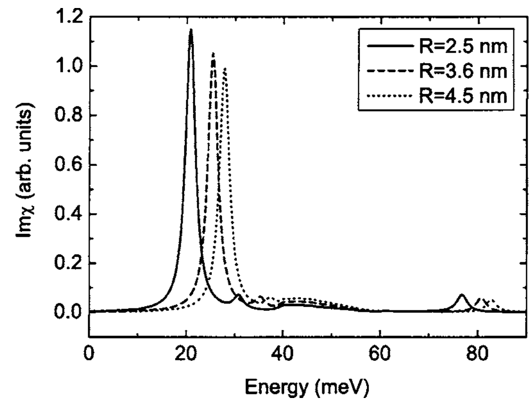

FIG. 3. Size dependence of the exciton dominant optical-absorption spectra in the QDSLW. The radius of the cylindrical QDSLW is R=2.5, 3.6, and 4.5 nm, respectively.

III. NUMERICAL RESULTS AND DISCUSSIONS

A SL consisting of 20 periods of $In_{0.53}Ga_{0.47}As$ cylindrical quantum dots in an InP nanowire is investigated. The thicknesses of the well layers and the barrier layers are 6.0 and 4.0 nm, respectively, and the radius of the cylindrical wire is $R=3.6$ nm. The SL lies in the center of the InP nanowire of length $L=300$ nm. The effective mass of electron and the heavy hole is $m_e=0.042m_0$ and $m_h=0.429m_0$ (m_0 is the mass of free electron), respectively. The effective masses are considered to be the same for the well and barrier materials. Due to band-gap energy difference of the materials, the potential barriers $V_{0e}=245.0$ meV (for the conduction electron) and $V_{0h}=367.6$ meV (for the valence heavy hole). The electron (hole) potential $U_{e(h)}=0$ inside the InGaAs dots and $U_{e(h)}=V_{0e(h)}$ in the InP barriers. The background dielectric constant is $\varepsilon=13.9$, the interband dipole moment $d=0.3\ e$ nm, the width of the Gaussian optical pumping pulse $\sigma=15$ fs, and the dephasing time $\gamma^{-1}=500$ fs.[28]

The absorption spectra of this QDSLW calculated in the absence and in the presence of Coulomb interaction are shown in Fig. 2. In this and following figures, the zero in the energy abscissa correponds to the band gap of InGaAs, i.e., 1259 meV. In the absence of Coulomb interaction, the spectrum is actually the joint density of states (JDOS), while in the presence of Coulomb interaction, the calculated spectrum shows exciton-dominant optical absorption. The upper panel shows an overall survey of the optical-absorption spectra, whereas the lower panel is the enlarged lower part of the upper panel to show the details. The JDOS of the ideal 1D QDSLW shows the miniband structure. Two minibands are observed in the considered energy range. The first miniband and mini-band-gap are about 20 and 35 meV wide whereas the second miniband is wider than the first miniband. Due to the 1D property of the structure, two broadened singularities with a $1/\sqrt{\omega}$ divergence occur at the miniband edges corresponding to the center and the edge of the mini-Brillouin-zone. In the presence of Coulomb interaction, the optical absorption is dominated by exciton peaks. The two exciton peaks below the first miniband correspond to the 1s and 2s excitons formed from the electron-hole pair of first valence and conduction minibands. The main excitonic peak is observed at about 20 meV below the first miniband. The exciton peak in the first mini-band-gap corresponds to the 1s exciton formed from the electron-hole pair of second valence and conduction minibands. Furthermore, the Coulomb interaction leads to the change of absorption from minibands to peaks emerging at the centers of the minibands. The intensity of absorption almost completely shifts to the main exciton peak from the miniband, whereas the absorption continuum above the first miniband edge is identically zero (except for some minipeaks). The compared optical absorption in an ideal 1D uniform QW is also shown in Fig. 2. The JDOS of the plain QW shows the $1/\sqrt{\omega}$ singularity at the edge of the band gap and a series of peaks above the band gap. The series of peaks are resulted from the quantized levels due to finite length of the QW. Also, the main excitonic peak is below the band gap at about 10 meV and the continuum above the band gap is smaller than the free electron-hole pair. In contrast to 2D and 3D cases, where the Coulomb interaction between the electron and the hole increases the continuum optical absorption, the Coulomb interaction in this 1D QW suppresses the continuum of interband absorption intensity. Very strong exciton absorption below the band gap and weak interband absorption above the band gap in a plain 1D QW have been experimentally observed[29] and theoretically investigated.[30]

In Fig. 3, the size dependence of the optical-absorption spectra of the QDSLWs with radii R=2.5, 3.6, and 4.5 nm is presented. Increasing R, the amplitude of the exciton peak decreases and its location with respect to miniband edge decreases. This size dependence can be understood by noting that the electron and hole have more opportunity apart from with increasing R, thus the overlapping of their wave functions reduces, and, therefore, the Coulomb interaction between them decreases. Note that a similar size dependence of photoluminescence from a plain InP nanowire by the radial quantum confinement has been reported.[31]

The optical-absorption spectra around the main exciton peak in the presence of an external dc electric field are shown in Fig. 4. Increasing the dc field, the location of the main exciton peak shifts to the lower-energy side and decreases in magnitude. Meanwhile, a minor peak appears increasingly and redshifts. The redshift of the minor peak is less than the main peak and the distance between the two

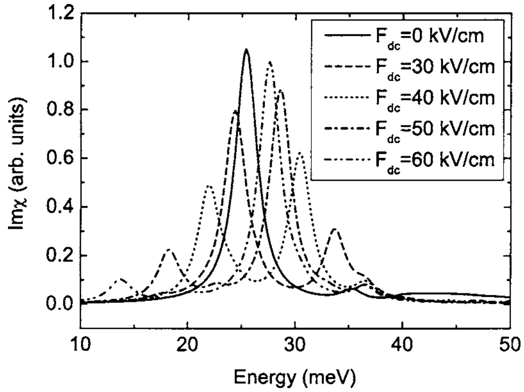

FIG. 4. Optical absorption in the QDSLW in the presence of a dc field F_{dc} along the wire with different field strengths. With the field strength increasing, the main exciton peak shift to the lower-energy side and its magnitude decreases and the minor peak next to the main exciton peak increases and redshifts.

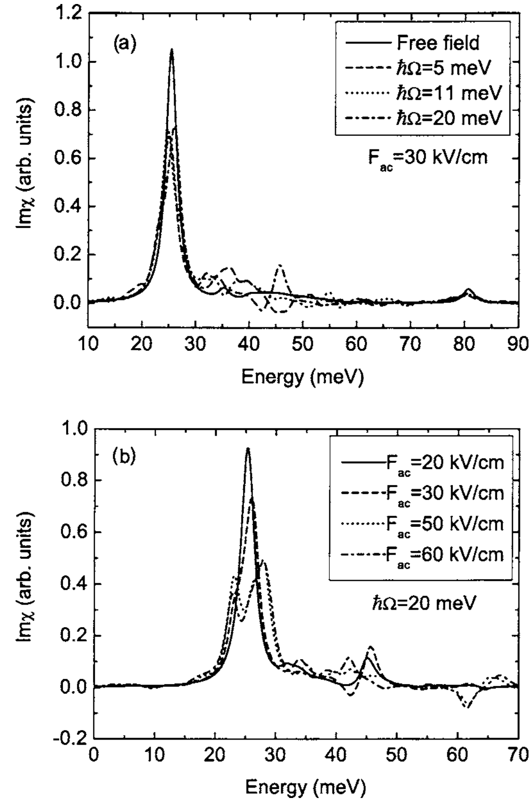

FIG. 5. Optical absorption in the QDSLW in the presence of a terahertz field F_{ac} along the wire. (a) Different frequencies $\hbar\Omega=5$, 11, and 20 meV, with field strength fixed at $F_{ac}=30$ kV/cm; (b) Different field strengths $F_{ac}=20$, 30, 50, and 60 kV/cm, with frequency fixed at $\hbar\Omega=20$ meV. Autler-Townes splitting of the exciton peak and one-photon and two-photon gain spectra at different field strengths.

peaks spreads. At certain field strengths, the two peaks are same in height. This is prominently different from the Franz-Keldysh effect in uniform QWs where oscillatory modulation of the spectra and strong below-gap absorption are reported.[20,32] It is also different from general quantum well SLs where clear Wannier-Stark ladders are observed.[33]

The effects of an external terahertz field on the interband optical-absorption spectra are shown in Fig. 5. Figure 5(a) shows the absorption spectra for different frequencies of the terahertz field with field strength fixed at $F_{ac}=30$ kV/cm. The main exciton peak decreases, broadens, and becomes asymmetric in the presence of an external terahertz field. There appears some small gain and absorption peaks in the spectra. These small absorption peaks are replicas of the excitonic states dressed by terahertz photons.[34] The excitonic absorption spectra for different field strengths F_{ac} with frequency fixed at $\hbar\Omega=20$ meV are shown in Fig. 5(b). The oscillator strength is almost completely transferred from the minibands to the exciton. Increasing the field strength, the main excitonic peak decreases, broadens, and splits up. The asymmetric splitting is the well-known Autler-Townes splitting and is due to the coherent coupling of the lowest and higher exciton states. Furthermore, when the field strength increases, there exist one-photon and two-photon gain spectra. The one-photon gain exists when $F_{ac}=30$ kV/cm and the two-photon gain exists when $F_{ac}=50$ and 60 kV/cm. The similar Autler-Townes splitting and two-photon gain in plain QWs under a terahertz field are recently reported by Hughes.[21] And the asymmetric Autler-Townes splitting is also investigated in transient four-wave mixing spectra from laser-driven excitonic Fano resonance in biased SLs.[35]

IV. CONCLUSIONS

The interband optical absorption in a $In_{0.53}Ga_{0.47}As/InP$ QDSLW in the presence of a homogeneous dc or terahertz field is investigated. The QDSLW has a miniband structure as a general quantum well SL does. The QDSLW shows very strong excitonic absorption below the lower miniband edge and weak interband absorption above the edge. The absorption spectra of the QDSLW in the presence of a dc field are distinct from the Franz-Keldysh effect in a plain QW and from a general quantum well SL which shows the Wannier-Stark ladders. In the presence of a terahertz field, the QDSLW shows the Autler-Townes splitting of the main exciton peak and one-photon and two-photon gain peaks in the absorption spectra.

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