Fano resonances in the optical spectra of semiconductor quantum structures

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The appearance of Fano resonances in the fine structure of the optical absorption spectra is predicted for all semiconductor quantum structures with one or two degrees of freedom. The resonances are explained by Coulomb coupling of the bound and scattering states of an exciton series belonging to different subband pairs but occurring at the same photon energy. To demonstrate this effect, two examples, the quantum well and the quantum-well wire, are treated numerically. The optical spectra show Fano resonances.

I. INTRODUCTION

In the optical spectra of quasi-one- and quasi-two-dimensional semiconductor quantum structures, the important transitions are usually classified according to the pairs of electron and hole subbands that are coupled. However, such a classification is only approximately valid. Indeed, the Coulomb interaction between electrons and holes couples all subbands. This holds especially for the exciton series belonging to different subband pairs. Since partially bound states of higher subband pairs and scattering states of lower subband pairs are energetically degenerate, a resonant Coulomb interaction between the discrete excitonic states and the continuum of electron-hole pair states occurs. The resulting Fano effect[1,2] should produce an asymmetric line shape of the exciton lines in the optical spectra of the low-dimensional system. The linewidth of those lines can be considerably larger than the homogeneous broadening.

In recent months, Fano resonances have been observed in photoluminescence excitation spectra of GaAs-$Ga_xAl_{1-x}As$ quantum wells[3] (QW's) and in linear absorption experiments on GaAs under high magnetic fields.[4]

The possibility for the occurrence of a Fano resonance has already been discussed for quantum wells,[5,6] when the ground state of the light-hole exciton lies in the heavy-hole continuum, but has not been observed experimentally. Though the Fano model[1] explains the basic features of these lines, it is less useful for determining the complete optical spectrum of a microstructure, because of the complicated geometry. Various analytical and numerical attempts have been made in order to simplify the problem and to determine an approximate result, which is valid in a neighborhood of the expected Fano resonance.[7–10] Very recently Willcox and Whittaker have found Fano resonances in numerically calculated absorption spectra of quantum wells.[11]

The interplay of Coulomb and confinement effects gives rise to optical spectral with a rich fine structure. For instance, the quantization of the center-of-mass motion is responsible for additional peaks.[12,13] Most calculations of excitons in quantum wells neglect the coupling of subband pairs, even when the Schrödinger equation is solved by numerical integration.[14]

In this paper, we show analytically that Fano resonances are a *typical* feature of the absorption continuum of low-dimensional semiconductor structures. Two examples, the quantum well and the quantum-well wire, are treated numerically in order to visualize this general result. In particular, we present complete spectra of quantum-well wires which, for the first time, include continuum states.

II. GENERAL THEORY

The linear optical susceptibility of a quantum mechanical system is given by

$$\chi(\omega) = \sum_{\Lambda} \frac{\left|\langle \Psi | \Phi_\Lambda \rangle\right|^2}{E_\Lambda - \hbar(\omega + i\epsilon)} \; ; \quad |\Psi\rangle = \hat{T}|\, i\,\rangle\,, \tag{1}$$

where $|\, i\,\rangle$ is the initial state and $\hat{T}$ is the operator of the optical transition. The quantities E_Λ and $|\Phi_\Lambda\rangle$ are eigenvalues and normalized eigenstates of a stationary Schrödinger equation:

$$\left[-\boldsymbol{\nabla}\cdot\frac{\hbar^2}{2m}\boldsymbol{\nabla} + W(x_1,\ldots,x_n)\right]\Phi_\Lambda(x_1,\ldots,x_n)$$

$$= E_\Lambda\,\Phi_\Lambda(x_1,\ldots,x_n)\,. \tag{2}$$

Here, m is the effective mass tensor.

Fano resonances can occur when both discrete and continuum states are involved in Eq. (2) and, furthermore, are coupled. We assume that in some directions ($\boldsymbol{\xi}$) the motion is unlimited, whereas the motion in the other directions ($\boldsymbol{\eta}$) is bounded on both sides, which implies a potential of the form,

$$W(\boldsymbol{\xi},\boldsymbol{\eta}) = V(\boldsymbol{\xi},\boldsymbol{\eta}) + U(\boldsymbol{\eta})\ ;\ \ \boldsymbol{\xi} = (x_1,\ldots,x_m)\ ;$$
$$\boldsymbol{\eta} = (x_{m+1},\ldots,x_n)\ ;\ \ 0 < m < n,$$

$$\lim_{\|\boldsymbol{\eta}\|\to\infty} U(\boldsymbol{\eta}) = +\infty\ ;\ \ V(\boldsymbol{\xi},\boldsymbol{\eta}) < 0\ ;$$
$$V(\boldsymbol{\xi},\boldsymbol{\eta}) \to 0 \ \text{ for } \ \sqrt{\|\boldsymbol{\xi}\|^2 + \|\boldsymbol{\eta}\|^2} \to \infty, \tag{3}$$

$m^{-1} = m_{\boldsymbol{\xi}}^{-1} \oplus m_{\boldsymbol{\eta}}^{-1}$; $m_{\boldsymbol{\xi}}^{-1}, m_{\boldsymbol{\eta}}^{-1}$ positive definite.

The eigenvalues of the equation for the $\boldsymbol{\eta}$ coordinates,

$$\left[-\boldsymbol{\nabla}_{\boldsymbol{\eta}} \cdot \frac{\hbar^2}{2m_{\boldsymbol{\eta}}} \boldsymbol{\nabla}_{\boldsymbol{\eta}} + U(\boldsymbol{\eta})\right] \varphi_\lambda(\boldsymbol{\eta})$$

$$= E_\lambda\, \varphi_\lambda(\boldsymbol{\eta})\ ;\ \ \lambda = 0,\ldots,\infty\,,$$

are discrete and unlimited. For each λ a set of functions, $\psi_{\lambda\mu}$ and $\psi_{\lambda E}$, describing approximately the motion along $\boldsymbol{\xi}$, can be defined according to,

$$\left[-\boldsymbol{\nabla}_{\boldsymbol{\xi}} \cdot \frac{\hbar^2}{2m_{\boldsymbol{\xi}}} \boldsymbol{\nabla}_{\boldsymbol{\xi}} + V_\lambda(\boldsymbol{\xi})\right] \psi_{\lambda\mu}(\boldsymbol{\xi}) = E_{\lambda\mu}\, \psi_{\lambda\mu}(\boldsymbol{\xi})\ ;$$

$$\mu = 1,\ldots,\infty\ ;\ \ E_{\lambda\mu} < 0,$$

$$\left[-\boldsymbol{\nabla}_{\boldsymbol{\xi}} \cdot \frac{\hbar^2}{2m_{\boldsymbol{\xi}}} \boldsymbol{\nabla}_{\boldsymbol{\xi}} + V_\lambda(\boldsymbol{\xi})\right] \psi_{\lambda E}(\boldsymbol{\xi}) = E\, \psi_{\lambda E}(\boldsymbol{\xi})\ ;\ \ E \geq 0,$$

$$V_\lambda(\boldsymbol{\xi}) = \int d^{n-m}\boldsymbol{\eta}\, |\varphi_\lambda(\boldsymbol{\eta})|^2\, V(\boldsymbol{\xi},\boldsymbol{\eta})\,.$$

Choosing functions φ_λ, ψ_μ that are normalized and $\psi_{\mu E}$ δ normalized, we can use as a basis for the representation of the Hamiltonian of Eq. (2) the products $\varphi_\lambda(\boldsymbol{\eta})\,\psi_{\lambda\mu}(\boldsymbol{\xi})$ and $\varphi_\lambda(\boldsymbol{\eta})\,\psi_{\lambda E}(\boldsymbol{\xi})$. There exist an infinite number of pairs $|\lambda,\mu\rangle$ and $|\lambda',E\rangle$ $(\lambda > \lambda')$, so that $E_\lambda + E_{\lambda\mu} = E_{\lambda'} + E$. The matrix element of the Hamiltonian between these states is given by

$$H_{\lambda\mu\,\lambda' E} = \int d^{n-m}\boldsymbol{\eta} \int d^m\boldsymbol{\xi}\, \varphi_\lambda^*(\boldsymbol{\eta})\, \psi_{\lambda\mu}^*(\boldsymbol{\xi})$$
$$\times V(\boldsymbol{\xi},\boldsymbol{\eta})\, \psi_{\lambda' E}(\boldsymbol{\xi})\, \varphi_{\lambda'}(\boldsymbol{\eta})\,, \tag{4}$$

which is identical with the Fano coupling matrix element.[1] Apart from the trivial cases in which either V is independent of $\boldsymbol{\eta}$ or the integral (4) vanishes for symmetry reasons (geometrical or electron-hole symmetry), we expect Fano resonances for states with good quantum numbers λ,μ if there exists a λ', such that the relation $E_\lambda + E_{\lambda\mu} > E_{\lambda'}$ is fulfilled.

We describe the optical properties of a microstructure in the framework of the envelope function formalism. This approximation works well if the modulation depth of the bands is much smaller than the band gap for the optical transition and the lattice-periodic functions and the band structure are not changed qualitatively by the microstructure. For example, this is approximately true for the heavy-hole–electron transition in $Ga_{1-x}Al_xAs$, as long as the aluminum mole fraction x does not exceed the cross-over value between $x = 0.35$ and $x = 0.43$.[15] Without going into further detail, we suppose that this approximation holds for the system under consideration. Assuming, for simplicity, one conduction and one valence band, the optical susceptibility is given by[16]

$$\chi^{(nD)}(\omega) = \frac{|\mu|^2}{\varepsilon_0} \frac{1}{\Omega^{(nD)}} \int_\Omega d^3\mathbf{r} \int_\Omega d^3\mathbf{r}'$$
$$\times \sum_\Lambda \frac{\Phi_\Lambda(\mathbf{r},\mathbf{r})\, \Phi_\Lambda^*(\mathbf{r}',\mathbf{r}')}{E_g + E_\Lambda - \hbar(\omega + i\epsilon)}\,, \tag{5}$$

where $E_g = E_c(\mathbf{0}) - E_v(\mathbf{0})$ is the gap energy and $\Omega^{(nD)}$ is the n-dimensional normalization volume ($n = 1,2,3$). The pair energies and wave functions, E_Λ and Φ_Λ, responsible for the positions and oscillator strengths of the resonances, are determined by the two-particle Schrödinger equation,

$$\left[-\frac{\hbar^2}{2m_e}\Delta_e + W_e(\mathbf{r}_e) - \frac{\hbar^2}{2m_h}\Delta_h + W_h(\mathbf{r}_h) - \frac{e^2}{4\pi\varepsilon_0\varepsilon|\mathbf{r}_e - \mathbf{r}_h|}\right] \Phi_\Lambda(\mathbf{r}_e,\mathbf{r}_h) = E_\Lambda\, \Phi_\Lambda(\mathbf{r}_e,\mathbf{r}_h)\,. \tag{6}$$

Here, $m_{e/h}$ are the masses and $W_{e/h}$ the confinement potentials for electron and hole, respectively.

Equation (6) represents a six-dimensional Schrödinger equation. If there is no confinement in some directions, the corresponding center-of-mass motions can be separated. Only the zero-wave vector solutions appear in the optical susceptibility. For the remaining coordinates, we obtain an equation of the form (2) with a nontrivial potential W and a dimensionality $n \leq 6$. For quasi-zero-dimensional systems (quantum dots), the two-particle motion is bounded in all directions so that the conditions (3) cannot be fulfilled. The same holds for three-dimensional (bulk) semiconductors, since center-of-mass and relative motion can be separated and the latter is completely unbounded. Therefore, no Fano resonances can be expected in quasi-zero- and three-dimensional semiconductors, but they should appear in quasi-one- and quasi-two-dimensional ones.

The Coulomb potential is responsible for the Fano coupling and plays the role of the potential V in Eq. (3). The parameter q, which determines the line shape,[1] is proportional to the transition matrix element of the discrete state and inversely proportional to the transition matrix element of the degenerate continuum states and to the Fano coupling parameter (4). Of course, in real systems, the confinement potentials do not go to infinity. This does not change the results qualitatively, as long as the height is far above the frequency range of interest.

Interestingly, for semiconductors in a magnetic field an equation of the same type as Eq. (6) can be derived. The existence of Fano resonances in a bulk semiconductor in a magnetic field has been proven recently in theory and experiment.[4] For a quantum well in a magnetic field, however, the absorption spectrum is purely discrete because of the confinement in all directions.[17]

III. EXAMPLES

In the last section, we have shown that Fano resonances are a general feature of quasi-one- and quasi-two-dimensional systems. As examples, in this section we study the very simplest cases, the ideal quantum well and the parabolic quantum-well wire in the limit of strong QW confinement, i.e., infinitely high barriers. To simplify the numerical calculation, we assume a two-band model with electron-hole symmetry, i.e., equal confinement potentials and masses for electrons and holes. This, of course, does not change the physics that is contained in Eqs. (2–5). In all explicit results, Rydberg units are used. Energies are represented in units of the binding energy E_B of the three-dimensional exciton and lengths in units of the Bohr radius a_B.

A. Quantum well

In a quantum well, the optical transitions are characterized by pairs of single-particle quantum numbers (n_e, n_h) of electron and hole, where $n_{e/h} = 1, \ldots \infty$. We consider the case of a layer of thickness d, bound by infinite barriers. In this idealized model, it is strictly required that $n_e = n_h$ for optically allowed transitions.

Expanding the two-particle wave functions Φ_Λ (6) in terms of quantum-well subbands n_e and n_h, the optical susceptibility (5) can be written,

$$\chi^{(2D)}(\omega) = \frac{|\mu|^2}{\varepsilon_0} \sum_\lambda \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \frac{\varphi_{\lambda nn}(0)\, \varphi^*_{\lambda n'n'}(0)}{E_g + E_\lambda - \hbar(\omega + i\epsilon)},$$

where

$$\sum_{n'_e=1}^{\infty} \sum_{n'_h=1}^{\infty} \left\{ \left[-\frac{\hbar^2}{2m} \frac{1}{\rho} \frac{d}{d\rho} \left(\rho \frac{d}{d\rho}\right) + \frac{\hbar^2}{2m_e} \frac{n_e^2 \pi^2}{d^2} + \frac{\hbar^2}{2m_h} \frac{n_h^2 \pi^2}{d^2} \right] \delta_{n_e n'_e}\, \delta_{n_h n'_h} - V_{n_e n_h n'_e n'_h}(\rho) \right\}$$

$$\times \varphi_{\lambda n'_e n'_h}(\rho) = E_\lambda\, \varphi_{\lambda n_e n_h}(\rho)$$

$$V_{n_e n_h n'_e n'_h}(\rho) = \frac{e^2}{4\pi\varepsilon\varepsilon_0} \frac{4}{d^2} \int_0^d dz_e \int_0^d dz_h \frac{\sin\left(\frac{n_e \pi z_e}{d}\right) \sin\left(\frac{n_h \pi z_h}{d}\right) \sin\left(\frac{n'_e \pi z_e}{d}\right) \sin\left(\frac{n'_h \pi z_h}{d}\right)}{\sqrt{(z_e - z_h)^2 + \rho^2}}.$$

Here, $m = m_e\, m_h/(m_e + m_h)$ is the reduced exciton mass. Furthermore, in the case of electron-hole symmetry, it is $m = {}^1\!/_2\, m_e = {}^1\!/_2\, m_h$. Using Rydberg units, the only parameters are the quantum-well thickness d and the homogeneous line broadening $\hbar\epsilon$.

The nondiagonal Coulomb matrix elements, $V_{nnn'n'}$ $(n \neq n')$, perform the coupling of different allowed optical transitions (n, n) and (n', n') and lead to Fano resonances when a discrete state of a higher subband pair is degenerate with a continuum state of a lower subband pair. In the ideal case, all Fano parameters are negative because all $V_{nnn'n'}(\rho) > 0$, the transition matrix elements for the subband pairs (n, n) and (n', n') are positive, and the minus sign of the Coulomb potential has to be taken into account. However, for arbitrary confinement potentials or complicated valence band structure the wave functions cannot be deduced from the confinement potentials in a simple manner, and no general rule can be given for the sign of the Fano parameter q.

In Fig. 1, the imaginary part of the two-dimensional optical susceptibility for a quantum well is shown for $d = 2.4$ (lower curve), 2.0 (intermediate curve), and $1.6 a_B$ (upper curve) versus $\hbar\omega - E_g$. A homogeneous linewidth of $\hbar\epsilon = 0.2\, E_B$ is introduced. As expected, the distances between consecutive peaks increases as the thickness decreases. The positions of the excitons of the higher subbands overlap energetically with the continua

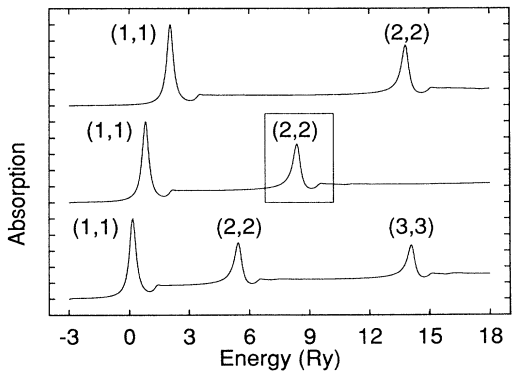

FIG. 1. Imaginary part of the optical susceptibility vs energy $\hbar\omega - E_g$ for quantum wells with $d = 2.4$, 2.0, and $1.6 a_B$. The homogeneous linewidth is taken as $\hbar\epsilon = 0.2 E_B$. The allowed optical transitions are characterized by the subband pairs (n_e, n_h).

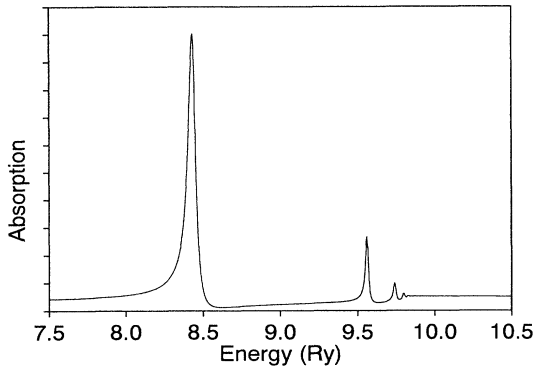

FIG. 2. The transition (2,2) from the framebox in Fig. 1, with a higher resolution $\hbar\epsilon = 0.01\,E_B$.

of the lower-lying subbands.

In order to study the line shape, we have reduced the value of the homogeneous broadening. In Fig. 2, the optical absorption of the transition (2,2) is shown for a well thickness of $2.0a_B$ and a homogeneous linewidth of $\hbar\epsilon = 0.01E_B$ (framebox in Fig. 1). The line shape clearly exhibits features of a Fano resonance, which can be characterized by the Fano parameters q and Γ.[1] The line is asymmetric, and the high-energy tail is smaller than the low-energy one, indicating a negative coupling parameter q. The total linewidth Γ is considerably larger than the introduced homogeneous linewidth $\hbar\epsilon$ and is, therefore, a result of the mixture of discrete and continuum states. The Fano parameter of the main peak is about $q \approx -6$. This resonance is followed by a whole series of lines stemming form higher order excitons. As predicted in the Fano model, the total linewidth is decreasing with increasing order.[1]

B. Quantum-well wire

In the case of a quantum-well wire with only the first well subband in the energy range of interest, the influence of the higher well subbands on the first one can be neglected. The gap energy then has to be replaced by the effective two-dimensional gap, $E^{(2D)}$, which takes into account the energy of the first quantum-well subband. We use for the relevant Coulomb matrix element $V_{1111}(\rho) = e^2/(4\pi\varepsilon_0\varepsilon\rho)$, which is obtained in the limit $d = 0$. This restriction is not necessary. Zimmermann[18] has shown that for a large range of thicknesses, the correct binding energy can be obtained if the Coulomb potential is replaced by an effective one.

For quasi-one-dimensional structures, the optical spectra are dominated by the center-of-mass motion.[12,13,16] Therefore, the eigenstates of the two-particle Schrödinger equation are characterized by quantum numbers $[N,n]$ of center-of-mass and the relative motion, where $N,n = 0,\ldots,\infty$.

For parabolic confinement and electron-hole symmetry, center-of-mass and relative motion can be separated, and the optical susceptibility becomes

$$\chi^{(1D)}(\omega) = \frac{|\mu|^2}{\varepsilon_0} \sum_{\substack{N=0 \\ (N\ \text{even})}}^{\infty} 2\sqrt{2\pi\Sigma^2}\,\frac{(N-1)!!}{N!!} \times \sum_n \frac{|\varphi_n(0,0)|^2}{E_g^{(2D)} + \hbar\bar{\omega}(N+\frac{1}{2}) + E_n - \hbar(\omega+i\epsilon)}\,.$$

Here, $\hbar\bar{\omega}$ is the subband spacing which is identical for electrons and holes, and $\Sigma = [\hbar/(2M\bar{\omega})]^{1/2}$ with $M = m_e + m_h$ can be interpreted as an effective wire width.

The remaining Schrödinger equation for the eigenfunctions φ_n and eigenvalues E_n of the internal motion is

$$\left[-\frac{\hbar^2}{2m}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right) + \frac{1}{2}\,m\,\bar{\omega}^2 y^2 - \frac{e^2}{4\pi\varepsilon_0\varepsilon\sqrt{x^2+y^2}}\right]\varphi_n(x,y) = E_n\,\varphi_n(x,y)\,.$$

This time, the Coulomb potential couples different subbands n,n' of the relative motion. Considering the matrix elements of the Coulomb potential with respect to the quantum numbers of the relative motion, we find that the Fano parameter q is negative for the allowed even-parity states. In quantum-well wires, the Coulomb interaction is generally stronger than in quantum wells, no matter which confinement is assumed or how large the distance of consecutive wire subbands is. The reason for this is that for a dimensionality lower than or equal to one the binding energy of the exciton is not limited.[19] Hence, we have to expect a more pronounced Fano effect.

In Fig. 3, optical spectra of quantum-well wires are plotted versus the energy $\hbar\omega - E_g^{(2D)}$ for a subband distance of $\hbar\bar{\omega} = 2.0$ (lower curve), 4.0 (intermediate curve), and $6.0E_B$ (upper curve). The corresponding wire widths are in the range of one exciton Bohr radius. A homogeneous broadening $\hbar\epsilon = 0.2\,E_B$ is introduced. The main peaks are due to the center-of-mass quantization, yet there are smaller ones which result from the relative motion of the exciton. For small values of $\hbar\bar{\omega}$, the

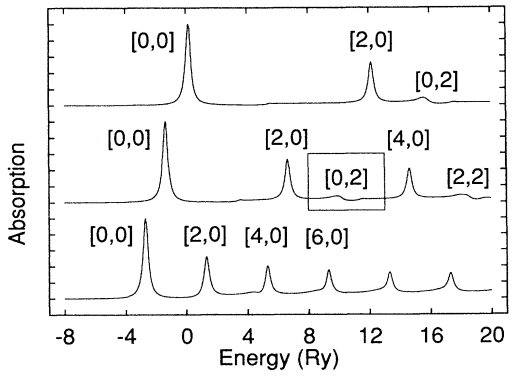

FIG. 3. Imaginary part of the optical susceptibility vs energy $\hbar\omega - E_g^{(2D)}$ for wires with different confinement $\hbar\bar{\omega} = 2.0$, 4.0, and $6.0\,E_B$ and a homogeneous broadening $\hbar\epsilon = 0.2\,E_B$. The pairs $[N,n]$ denote the quantum numbers for center-of-mass and relative motion.

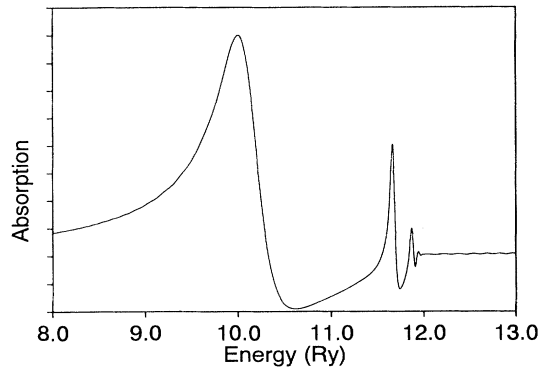

FIG. 4. Line shape of the transition [0, 2], from the framebox in Fig. 3, from the framebox with a higher resolution $\hbar\epsilon = 0.01E_B$.

fine structure is hardly visible. The side peaks become more pronounced as the quantization energy increases. Obviously, there is a difference between the first series of side peaks and the higher ones. The reason for this is that the excitons of the first wire subband remain discrete states, whereas the higher ones are energetically degenerate with continuum states of lower-lying subband, leading to Fano resonances. They are smaller but considerably broader, and their shape departs strongly from the Lorentzian form.

In order to study the line shape in detail, the optical absorption is calculated for $\hbar\bar{\omega} = 4.0E_B$, for a much smaller broadening of $0.01E_B$. The transition [0, 2] is shown in Fig. 4, which corresponds to the marked sector in Fig. 3. Again, features of a Fano resonance are visible. The Fano parameter is $q \approx -2$, which indicates a strong coupling.

IV. CONCLUSIONS

The analytical treatment in Sec. II has shown that the occurrence of Fano resonances is a typical feature of quasi-one- and quasi-two-dimensional semiconductor structures, independent of the details of the confinement. The Coulomb interaction between electrons and holes, which can never be avoided in semiconductors, gives rise to the necessary coupling.

Two simplified examples have been treated numerically in order to illustrate the theoretical predictions. In quantum wells, the Fano parameter q of the optically allowed transitions $n_e = n_h$ is rather large in magnitude and a Fano line shape is visible only for extremely small homogeneous broadening. In quantum-well wires, the Fano parameters are much smaller in magnitude because of the larger Coulomb coupling of subbands in quasi-one-dimensional systems. Therefore, the effect should be observable in quasi-one-dimensional systems with small homogeneous broadening and a wire confinement of the order of the exciton Bohr radius.

The examples, in the paper, were not chosen to give a quantitative description of the absorption spectra of a large variety of microstructures studied experimentally. Nevertheless, we will briefly discuss the changes that occur for real structures.

In a quantum well with electron-hole symmetry, optically allowed continuum states are Coulomb coupled to forbidden discrete lines leading to Fano resonances with a parameter $q = 0$, i.e., a Lorentzian dip in the continuum. If the assumption of electron-hole symmetry is lifted, those forbidden states will lead to Fano resonances with small values of q. Those lines are more likely to be observed experimentally and have, in fact, been found in asymmetric double quantum wells.[3]

In quantum-well wires the electron-hole-asymmetry-induced coupling of center-of-mass and relative coordinate is proportional to Yy, where Y and y are the center-of-mass and relative coordinate, respectively. Since all allowed transitions have even parity with respect to N and λ, the single lines in the spectrum are not coupled to any visible continuum states. Therefore, the Lorentzian lines are not accidental and should still appear if the condition of electron-hole symmetry is not fulfilled. On the other hand, visible continuum states can be coupled to invisible discrete states leading to Fano resonances with $q = 0$.

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