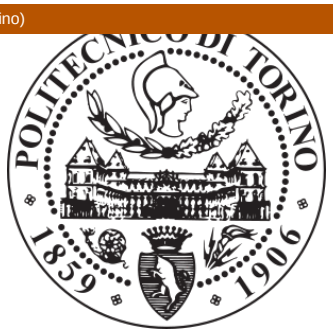

Coulomb-Induced Suppression of Band-Edge Singularities in the Optical Spectra of Realistic Quantum-Wire Structures

Coulomb-Induced Suppression of Band-Edge Singularities in the Optical Spectra of Realistic Quantum-Wire Structures

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The linear and nonlinear optical properties of realistic quantum wires are studied through a theoretical approach based on a set of generalized semiconductor Bloch equations. Our scheme allows a full three-dimensional multisubband description of electron-hole correlation for any confinement profile, thus permitting a direct comparison with experiments for available quantum-wire structures. Our results show that electron-hole Coulomb correlation removes the one-dimensional band-edge singularities from the absorption spectra, whose shape is heavily modified with respect to the ideal free-carrier single-subband case over the whole density range. [S0031-9007(96)00125-1]

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The importance of excitonic and correlation effects in optical spectra of semiconductors and their dependence on dimensionality has now been long recognized [1,2]. More recently, increasing interest has been devoted to one-dimensional (1D) systems [3], prompted by promising advances in quantum-wire fabrication and application, e.g., quantum-wire lasers. The main goal of such an effort in basic research as well as technological applications is to achieve structures with improved optical efficiency as compared to two-dimensional (2D) and three-dimensional (3D) ones.

A common argument in favor of this effort is based on the well known Van Hove divergence in the 1D joint density of states (DOS), which is expected to give rise to very sharp peaks in the optical spectra of 1D structures. Such a prediction is, however, based on free-carrier properties of ideal 1D systems, and it ignores any disorder-induced and Coulomb-correlation effects.

As pointed out in the pioneering papers by Ogawa and Takagahara [4] and later by Benner and Haug [5], electron-hole correlation is expected to strongly influence the optical spectra of 1D systems. Their theoretical investigation, based on a single-subband-model solution of the 1D Schrödinger equation in terms of a modified 1D Coulomb potential [4], shows that the inverse-square-root singularity in the 1D DOS at the band edge is smoothed when excitonic effects are taken into account. The question is still open whether one can expect that the above theoretical predictions, obtained for model 1D systems, also apply to the real quantum wires made available by the present technology. Indeed, wires with the best optical quality presently include structures obtained by epitaxial growth on nonplanar substrates (V-shaped wires) [3,6–10], or by cleaved-edge quantum well overgrowth (T-shaped wires) [11]. Owing to the shape of the confinement potential, these systems are far from an ideal 1D character. While quasi-1D confinement has been demonstrated for the lowest level [3,6–9,11], excited states gradually approach a 2D-like behavior. Moreover, in the available samples the subband separation is still relatively small, so that the coupling between different subbands may be important.

From an experimental point of view, it is a matter of fact that while 2D features are clearly observed in photoluminescence excitation spectra of quantum wells, so far no "sharp" 1D features have been detected in the corresponding spectra of quantum wires. This is true despite the high quality of some of these structures, whose 1D character has been independently established by other methods [3,6–9,11]. However, the measured spectra are expected to be also strongly influenced by disorder-induced inhomogeneous broadening [12], and, therefore, it has so far been difficult to identify the role played by electron-hole correlation.

From all these considerations, the following questions still need to be answered: Are electron-hole correlation effects playing a dominant role also in realistic quantum-wire structures? If so, are these expected to hinder the possible advantages of the reduced dimensionality for relevant values of temperature and carrier density?

To answer these questions, we have undertaken a full calculation of the optical properties of realistic quantum wires. The proposed theoretical approach is based on a generalization of the well known semiconductor Bloch equations (SBE) to the case of a multisubband wire. More specifically, by denoting with $\{k_z \nu^{e/h}\}$ the free electron and hole states (k_z and $\nu^{e/h}$ being, respectively, the wave vector along the wire direction, z, and the subband index corresponding to the confinement potential in the x-y plane), we consider as kinetic variables the various distribution functions of electrons and holes $f^{e/h}_{k_z\nu}$ as well as the corresponding diagonal ($\nu^e = \nu^h = \nu$) interband polarizations $p_{k_z\nu}$ [13]. This kinetic description is a generalization to 1D systems of a standard approach for the study of bulk semiconductors [2,14] recently applied also to superlattice structures [15]. Within our $k_z\nu$ representation, the SBE, describing the time evolution

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of the above kinetic variables, are written as

$$\frac{\partial}{\partial t} f^{e/h}_{\pm k_z \nu} = \frac{1}{i\hbar}(\mathcal{U}_{k_z \nu} p^*_{k_z \nu} - \mathcal{U}^*_{k_z \nu} p_{k_z \nu}) + \frac{\partial}{\partial t} f^{e/h}_{\pm k_z \nu}|_{\text{inco}} ,$$

$$\frac{\partial}{\partial t} p_{k_z \nu} = \frac{1}{i\hbar}(\mathcal{E}^e_{k_z \nu} + \mathcal{E}^h_{-k_z \nu}) p_{k_z \nu} + \frac{1}{i\hbar}\mathcal{U}_{k_z \nu}(1 - f^e_{k_z \nu} - f^h_{-k_z \nu}) + \frac{\partial}{\partial t} p_{k_z \nu}|_{\text{inco}} , \quad (1)$$

where $\mathcal{U}_{k_z \nu}$ and $\mathcal{E}^{e/h}_{k_z \nu}$ are, respectively, the renormalized fields and subbands [2] whose explicit form involves the full 3D Coulomb potential [15]. The $\pm$ sign in Eq. (1) refers to electrons (e) and holes (h), respectively, while the last terms on the right hand side of Eq. (1) denote the contributions due to incoherent processes, e.g., carrier-carrier and carrier-phonon scattering.

In this Letter we focus on the quasiequilibrium regime. Therefore, Fermi-Dirac $f^{e/h}_{k_z \nu}$ are assumed, and the solution of the set of SBE (1) simply reduces to the solution of the polarization equation. This is performed by means of a direct numerical evaluation of the stationary solutions, i.e., polarization eigenvalues and eigenvectors. These two ingredients fully determine the absorption spectrum as well as the exciton wave function in 3D real space. In particular, the electron-hole correlation function vs the relative free coordinate $z = z^e - z^h$ is given by $g(z) = \sum_{k_z k'_z \nu} p^*_{k_z \nu} p_{k'_z \nu} e^{i(k'_z - k_z)z}$.

The above theoretical approach has been applied to the GaAs/AlGaAs V-shaped wires of Ref. [8]. The main ingredients entering our SBE are the single-particle energies and wave functions, obtained for the 2D confinement potential deduced from transmission electron microscopy [8]. To illustrate the role played by electron-hole correlation, we start by showing in Fig. 1 the linear-absorption spectra obtained when taking into account an increasing number of wire subbands. Results of our Coulomb-correlated (CC) approach are compared with those of the free-carrier (FC) model [16] (solid and dashed lines, respectively). In Fig. 1(a) only the lowest wire subband is considered. Electron-hole correlation gives rise to two important effects: First, the excitonic peak arises below the onset of the continuum, with a binding energy E_b of about 12 meV. Second, the shape of the CC spectrum in the continuum region is drastically modified with respect to the FC one. In agreement with previous investigations based on simplified 1D models [4,5], we find a strong suppression of the 1D DOS singularity. The same behavior characterizes the spectrum obtained considering the second subband only: The main difference is a reduction of E_b to about 9 meV, which, for the particular V-like structure considered [8], reflects the increased wave-function delocalization along the wire sidewalls. These excitonic splittings are in excellent agreement with recent magneto-luminescence experiments [8].

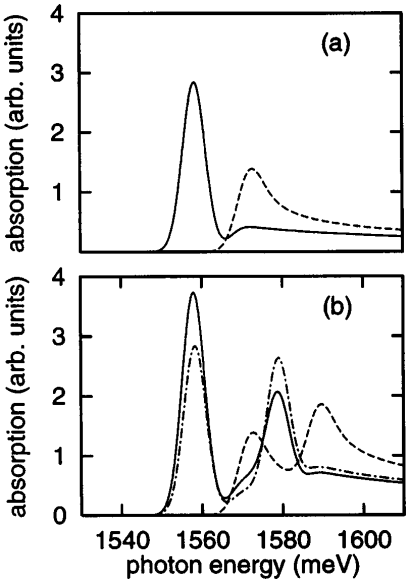

FIG. 1. Comparison between the absorption spectra of the V-shaped wire obtained by including electron-hole Coulomb correlation (CC model, solid line) or by assuming free carriers (FC model, dashed line). In (a) only the first subband is considered, while in (b) the two lowest subbands are taken into account. The dash-dotted line in (b) illustrates the effect of neglecting intersubband coupling in the CC spectrum (see text). All curves were computed assuming a Gaussian energy broadening of 2 meV.

Because of the relatively small intersubband splitting compared to the exciton binding energy, a significant intersubband coupling is expected. Figure 1(b) shows spectra obtained by including both (first and second) subbands. To better identify the role played by intersubband coupling, in addition to the CC and FC results we also report the coupling-free CC result (dash-dotted line). The main effect is an oscillator-strength transfer toward the low-energy region which results in a significant increase of the first exciton peak and a corresponding decrease of the second one.

Let us now discuss the physical origin of the dramatic suppression of the band-edge singularity in the CC absorption spectrum [Fig. 1(a)]. Since the absorption spectrum is proportional to the product of the electron-hole DOS and the oscillator strength (OS), we have studied these two quantities separately. In Fig. 2 we compare the DOS obtained within the CC and FC models (solid and dashed lines, respectively). The difference is hardly visible on this scale, and the pronounced 1D singularity in the DOS is not reduced by electron-hole correlation. Figure 3(a) shows that the quantity which is mainly modified by CC is the OS. Here, the ratio between the CC and FC OS is plotted as a function of excess energy with respect to the band edge (solid line). This ratio is always less than 1, and, in agreement with the results of 1D models [4], it goes to 0 at the band edge.

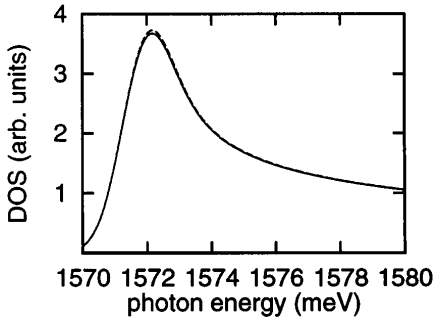

FIG. 2. Electron-hole DOS of the V-shaped wire close to the band edge, obtained within the CC (solid line) and FC (dashed line) model. Both curves were computed assuming a Gaussian energy broadening of 2 meV. The difference is hardly visible on this scale (see text).

Such vanishing behavior is found to dominate the 1D DOS singularity, and, as a result, the absorption spectrum exhibits a regular behavior at the band edge [Fig. 1(a), solid line].

Since the OS reflects the value of the correlation function $g(z)$ for $z = 0$ [4], i.e., the probability of finding electron and hole at the same place, the vanishing behavior of the OS in Fig. 3(a) seems to indicate a sort of electron-hole "effective repulsion." This is confirmed by a detailed analysis of the electron-hole correlation function, $g(z)$, reported in Fig. 3(b). Here, $g(z)$ (corresponding to the square of the exciton wave function in a 1D model) is plotted for three different values of the excess energy. We clearly see that the values of g for $z = 0$ correspond to the values of the OS ratio at the same energies [Fig. 3(a), solid line]. Moreover, we notice the presence of a true "electron-hole correlation hole," whose spatial extension strongly increases when approaching the band edge.

The above analysis provides a positive answer to our first question. Also, for realistic quantum-wire structures, electron-hole correlation leads to a strong suppression of the 1D band-edge singularity in the linear-absorption spectrum. Contrary to the 2D and 3D case, the Sommerfeld factor, i.e., the ratio between the CC and FC absorption, is less than unity [dashed line in Fig. 3(a)].

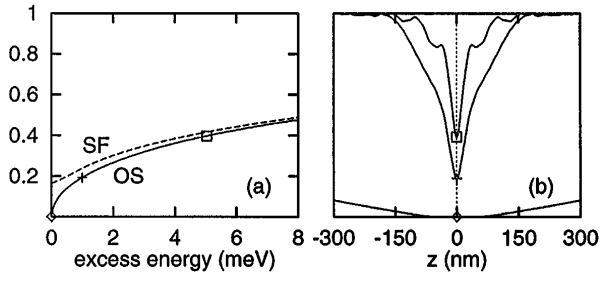

FIG. 3. (a) Oscillator-strength ratio between CC and FC spectra (solid line) and Sommerfeld factor (dashed line) as a function of the excess energy. (b) Electron-hole correlation function $g(z)$ vs relative distance z for three different values of the excess energy, identified by the corresponding symbols in (a). Note that $g(z = 0)$ gives directly the oscillator strength for the corresponding excess energy.

Finally, in order to answer our second and more crucial question, we must consider that most of the potential quantum-wire applications, i.e., 1D lasers and modulators, operate in strongly nonlinear-response regimes [3]. In such conditions, the above linear-response analysis has to be generalized, taking into account additional factors as (i) screening effects, (ii) band renormalization, and (iii) space-phase filling. We want to stress that all these effects are already accounted for in our SBE (1) [2]. Figure 4 shows the first quantitative analysis of nonlinear absorption spectra of realistic V-shaped wire structures for different carrier densities at room temperature. The screening model used in the calculation is described in [17]. In Fig. 4(a) we show as a reference the results obtained by including only the lowest subband. In the low-density limit (case A: $n = 10^4$ cm^{-1}) we clearly recognize the exciton peak. With increasing carrier density, the strength of the excitonic absorption decreases due to phase-space filling and screening of the attractive electron-hole interaction, and, moreover, the band renormalization leads to a redshift of the continuum. Above the Mott density (here about 8×10^5 cm^{-1}), the exciton completely disappears. At a density of 4×10^6 cm^{-1} (case D) the spectrum already exhibits

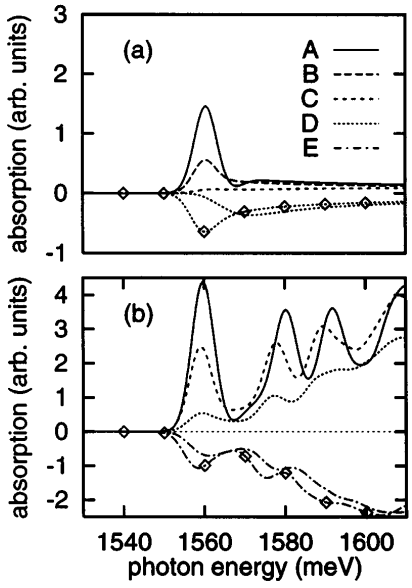

FIG. 4. Nonlinear absorption spectra of the V-shaped wire at room temperature for different carrier densities: A = 10^4 cm^{-1}; B = 5×10^5 cm^{-1}; C = 10^6 cm^{-1}; D = 4×10^6 cm^{-1}; E = 2×10^7 cm^{-1}. (a) Single-subband case, (b) realistic twelve-subband spectrum. For the highest densities [case D in (a) and E in (b)], the corresponding FC result is also shown (curve marked with diamonds). For a better comparison of the line shapes, the FC band edge has been redshifted to align with the corresponding CC one.

a negative region corresponding to stimulated emission, i.e., gain regime. As desired, the well pronounced gain spectrum extends over a limited energy region (smaller than the thermal energy). However, its shape differs considerably from the ideal FC one. The FC curve is plotted in the same figure and marked with diamonds; note that it has been shifted in energy to align the onset of the absorption to allow a better comparison of the line shapes [18]. The typical shape of the band-edge singularity in the ideal FC gain spectrum is strongly modified by electron-hole correlation. Also, at this relatively high carrier density, the OS corresponding to the CC model goes to zero at the band edge as for the low-density limit [Fig. 3(a)]. As a consequence, the FC peak is strongly suppressed, and only its high-energy part survives. The overall effect is a broader and less pronounced gain region.

Finally, Fig. 4(b) shows the nonlinear spectra corresponding to the realistic case of a twelve-subband V-shaped wire. In comparison with the single-subband case [Fig. 4(a)], the multisubband nature is found to play an important role in modifying the typical shape of the gain spectra, which, for both CC and FC models, extends over a range much larger than that of the single-subband case for the present wire geometry [Fig. 4(a)]. In addition, the Coulomb-induced suppression of the single-subband singularities, here also due to intersubband-coupling effects, tends to reduce the residual structures in the gain profile. Therefore, even in the ideal case of a quantum wire with negligible disorder and scattering-induced broadening, our analysis indicates that, for the typical structure considered, the shape of the absorption spectra over the whole density range differs considerably from the sharp FC spectrum of Fig. 1.

These conclusions allow us to reconsider the perspectives of quantum-wire physics and technology. In particular, comparing the nonlinear absorption spectra of Figs. 4(a) and 4(b), we see that the broad gain region in Fig. 4(b) is mainly ascribed to the multisubband nature or, more precisely, to the small intersubband splitting compared to the single-subband gain range in Fig. 4(a). This confirms that, in order to obtain sharp gain profiles, one of the basic steps in quantum-wire technology is to produce structures with increased subband splitting. However, the disorder-induced inhomogeneous broadening, not considered here, is known to increase significantly the spectral broadening [12], and this effect is expected to increase with increasing subband splitting. Therefore, extremely high-quality structures (e.g., single-monolayer control) seem to be the only possible candidates for successful quantum-wire applications.

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[16] All the spectra were computed assuming a Gaussian broadening of 2 meV. This small broadening (as compared to that of realistic wire structures [12]) allows us to better identify the role played by electron-hole correlation.
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