Electron-Hole Correlation Effects in the Emission of Light from Quantum Wires

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We present a self-consistent treatment of the electron-hole correlations in optically excited quantum wires within the ladder approximation and using a contact potential interaction. The limitation of the ladder approximation to the excitonic low-density region is largely overcome by the introduction of higher-order correlations through self-consistency. We show the relevance of these correlations in the low-temperature emission, even for high density, when a large gain replaces the excitonic absorption. [S0031-9007(98)08199-X]

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Based on the singularity of the density of states (DOS) of massive particles at the band edge in one dimension (1D) and the single-particle picture, large gains and significant improvements in optical devices have been predicted for 1D systems [1]. However, inclusion of the Coulomb correlations among the photoexcited carriers results in a *finite* absorption at the band gap, i.e., in a vanishing Sommerfeld factor [2]. This effect is related to the large exciton binding energy and oscillator strength, which strongly reduce that available for free carriers: correlation between electrons and holes is therefore very important in 1D, and likely to persist up to large carrier densities. Time-dependent semiconductor Bloch equations (SBE) [3] and later developments successfully addressed the understanding of most optical properties of semiconductors under strong excitation. These equations dynamically include the Coulomb interaction among carriers in response to the external transverse electromagnetic fields, and thus also reproduce excitonic resonances at low density [3]. Extensions of the SBE include screening of the Coulomb potential and scattering terms between carriers, treated both at the semiclassical [4] and at the non-Markovian level [5]. Also, for the wires, a satisfactory description of absorption and emission at large density was obtained, even without including screening [6,7], which entails limited changes. In the SBE, the existence of bound excitons or the correlation between electrons and holes is not taken into account in the propagation of carriers. Bound pairs and correlations result from infinite scatterings between electrons and holes (and thus relate to a non-Markovian dynamics). We may expect that these effects are important in one dimension, at low density and/or temperatures. Electron-hole scattering to infinite order has been considered at the level of the optical response, using ladder-type diagrams [ladder approximation (LA)] in the calculation of the two-particle Green functions [8]. However, this is equivalent to the stationary (and quasiequilibrium) limit of the dynamical Hartree-Fock equations, or SBE, as bare carrier propagation only was again considered [8]. In this paper, we assess the importance of electron-hole correlation on the propagation of carriers. We thus dress their propagators with the scattering with other carriers to infinite order. The resulting ladder approximation is solved self-consistently (SCLA). It has been shown that multiple scattering between both bound and unbound pairs is thus effectively included [9]. These produce a frequency-dependent broadening that is not contained in the semiclassical treatment of scattering. We remark that the relevant energy scale for this correlation is the exciton binding energy, which can be significantly large in one-dimensional systems.

We consider one parabolic conduction band for electrons (creation operators $c_k^\dagger$) and one for holes ($d_k^\dagger$), with the same masses. Charges interact through a simple contact potential $V(x) = a\delta(x)$, where $a = 2E_b a_B$, with E_b and a_B being the binding energy and the Bohr radius of the exciton. Electrons and holes are independent fermions, and the corresponding operators commute. We introduce dimensionless units, where $E_b = 1/2$, and lengths are scaled with the 1D Bohr radius. Then, the Hamiltonian reads

$$\mathcal{H} = \sum_k \frac{k^2}{4}[c_k^\dagger c_k + d_k^\dagger d_k] - \sum_{k,k',q} c_{k-q}^\dagger d_{k'+q}^\dagger d_{k'} c_k \,.$$

The choice of the contact potential makes the T-matrix self-consistency manageable for both numerical and analytical treatments. Because of the Pauli exclusion principle, electron-electron and hole-hole interactions are absent for the same spin within this potential. If these terms were included, direct and exchange terms would cancel out exactly to all orders, whereas they do not for a long-range potential. We are thus missing long-range effects, but have better focusing on the electron-hole correlation alone. We also mention that screening cannot be consistently introduced within such a restricted form of the potential. This is presumably a minor drawback

in 1D, as remarked above. When the wires are optically excited with a short light pulse, the electron-hole pairs are coherent with the excitation fields, and the system is strongly out of equilibrium. At later times, energy is exchanged among the carriers and with the lattice, and coherences are washed out. Eventually, the electron-hole gas reaches quasithermal equilibrium, and the density slowly varies in time as electron and holes radiatively recombine. In the following, we consider only such a state of stationary quasithermal equilibrium, and we do not address the coherence and the nonequilibrium dynamics at short times. Standard finite temperature Green function techniques may thus be used to write the SCLA. However, the solution of the self-consistent problem is by necessity numerical, and the analytical extension to real frequencies is numerically difficult, due to the complex structure of the resulting Green functions [10]. For this reason we work directly with real frequencies and/or times, where the Green functions and self-energies are matrices defined by the Keldysh contour [11], and their matrix elements are related by the Kubo-Martin-Schwinger relations for thermal equilibrium. This also allows for extensions to nonequilibrium conditions. The full SCLA reads

$$T_k(\omega) = 1 + \int_C d\omega' \sum_{q'} [iG_{k-q'}(\omega')G_{q'}(\omega - \omega')]T_k(\omega),$$
$$G_k(\omega) = G_k^{(0)}(\omega) + G_k^{(0)}(\omega)\Sigma_k(\omega)G_k(\omega), \qquad (1)$$
$$\Sigma_k(\tau) = \sum_q iT_k(\tau)G_{q-k}(-\tau).$$

The SCLA is a *conserving approximation* [12]: particle number and energy are conserved, making the SCLA robust at any density, even when large scatterings produce significant renormalizations. A detailed study of SCLA for both the attractive and the repulsive (single-band) Hubbard model at half-filling shows that the SCLA is substantially close to the available exact solution in an extensive range of temperatures and/or magnetizations [13]. We expect this to be even more true for this case of negligible fillings (continuum model).

In the low-density limit, using the bare particle propagators, the T matrix shows a pole at the exciton energy, $T^+(k,\omega) \simeq [\omega - (-1/2 + k^2/8) + 2i\gamma]^{-1}$. At the next self-consistency step, the particle propagators show structures below the exciton energy through the self-energy Σ. Higher order terms in self-consistency then introduce exciton-exciton scattering effects [9]. These effects are actually overestimated for the contact potential with respect to a realistic Coulomb potential due to the lack of electron-electron (and hole-hole) interactions for equal spins [14]. We plot in Fig. 1 the spectral function at different steps of iteration for $T = 0.1$, $\mu = -0.3$. At step 0, the spectral function shows the usual single-particle single peak, broadened by $\gamma = 0.02$ for regularization. At step 1, excitonic correlations appear. Because of finite temperature, not only a single excitonic peak appears but contributions from larger exciton momenta are also evident. Further self-consistency steps significantly broaden and *shift* both the single-particle and two-particle peaks. Discretization of the k space to $N_k = 16$ points helps to illustrate these effects. We thus understand that interaction among carriers shifts both exciton energies and single-particle energies.

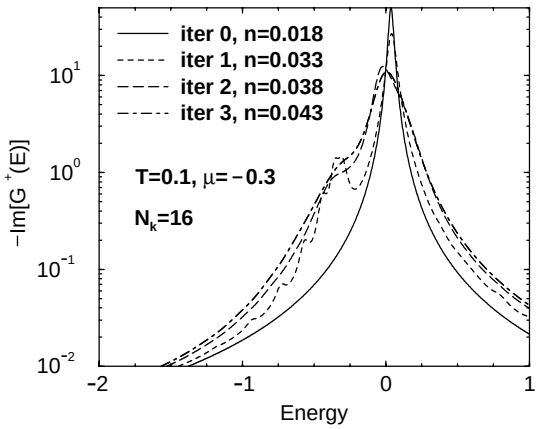

FIG. 1. The spectral function $\mathrm{Im}[G^+]$ at $k = 0$, first four steps of iteration. Corresponding densities are also shown.

Next, we introduce the optically measurable quantities, emission (or photoluminescence), and absorption. We calculate them in the dipole approximation, and neglect retardation in the electromagnetic interaction between carriers (polaritonic effects). The polarization P is the linear response to the classical external transverse field, calculated within the dipole approximation. It is thus related to the two-particle electron-hole Green function $G_2(\mathbf{k}_e, \mathbf{k}_h; \mathbf{k}_e - \mathbf{Q}, \mathbf{k}_h + \mathbf{Q})$, where $\mathbf{k} = (k, \omega)$, with the external electron and hole lines closed onto the interaction with the transverse photon:

$$P(\mathbf{k}) \propto \sum_{\mathbf{Q},\mathbf{k}_e+\mathbf{k}_h=\mathbf{k}} G_2(\mathbf{k}_e, \mathbf{k}_h; \mathbf{k}_e - \mathbf{Q}, \mathbf{k}_h + \mathbf{Q}) \qquad (2)$$
$$= H(\mathbf{k}) + H(\mathbf{k})T(\mathbf{k})H(\mathbf{k}), \qquad (3)$$

with $H(\mathbf{k}) = i\sum_{\mathbf{q}} G(\mathbf{k}/2 - \mathbf{q})G(\mathbf{k}/2 + \mathbf{q})$. Optical frequencies are measured from the gap. The first term of Eq. (3) describes the usual band-to-band recombination found in the single-particle picture, whereas the other term includes the correlation effects of bound electron-hole states in the optical properties. Here we notice that Eq. (2) gives the linear response of the system to the external field only when the exact G_2 is used [12]. However, we may expect that it is accurate also when G_2 is calculated within the SCLA [Eq. (3)] as this is quantitatively close to the exact solution. We further checked that deviations from current conservation are negligible.

The absorption α, related to $\mathrm{Im}[P^+]$ [the $(+)$ apex labeling the retarded function], can be written as

$$\alpha(\mathbf{k}) \propto -\mathrm{Im}[H^+(\mathbf{k})]\,|T^+(\mathbf{k})|^2.$$

Thus, absorption in the excitonic limit (low densities) shows a peak at the exciton energy. Moreover, by

using the small density analytic expression $H^+(k = 0, \omega) = -i/\sqrt{2\omega + i0^+}$, we find the absorption above the gap $\alpha(k = 0, \omega) = \sqrt{2\omega}/(1 + 2\omega)$, which clearly shows cancellation of the 1D DOS divergence at $\omega \to 0^+$, i.e., a Sommerfeld factor which vanishes linearly at $\omega = 0$. Emission is instead related to the correlation function of the dipole operator and therefore to $P^<(k, \omega)$. It can be found from absorption through the Kubo-Martin-Schwinger relations that

$$P^<(k, \omega) \propto [\exp \beta(\omega - 2\mu) - 1]^{-1} \operatorname{Im}[P^+(k, \omega)]. \tag{4}$$

It shows the typical *Bose* occupation factor for the emission. We conclude that at low densities and/or temperatures emission is excitonic, as also described in absorption by the poles of T^+.

In the following, we discuss emission and absorption at intermediate and high densities, when a relevant number of unbound pairs coexist with bound pairs. Stronger correlation effects are found at smaller temperatures, when most of the carriers are bound into excitons, but our aim is to show their relevance even in a less extreme situation. We solved Eq. (1) for $T = 0.25$ (corresponding to $E_b/2$) and densities up to $n = 0.8$. In Fig. 2 we plot the resulting absorption and emission. The low-density limit shows the excitonic absorption, and band-to-band absorption as a long tail at higher energy. We also notice small excitonic gain below the exciton peak, which is consistently predicted in the SCLA as are the broadenings [15]. For the $n = 0.06$ curve, the maximum gain is 0.1 at $E = -1$. At larger densities, excitonic absorption is bleached and substantial gain becomes evident. Even for $n = 0.38$, when large gain is present in the exciton spectral region, the emission peak stays fixed close to $E_b \sim -1/2$, the low-density exciton energy. For $n > 0.4$, the emission intensity saturates, and a peculiar structure of the emission peak develops from the significant depth of the Fermi sea of electrons and holes. Significant density-dependent broadenings, increasing with n, are predicted.

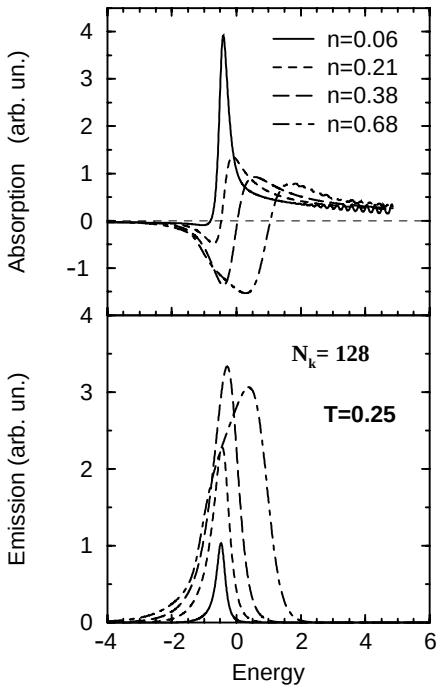

FIG. 2. The absorption and emission at various densities.

In Fig. 3 we compare the "exciton" energy [defined as the $|T^+|^2$ peak energy (TP)], the peak emission energy (EP), and the band gap renormalization [(BGR), twice the shift of the single-particle energy]. We notice large *negative* BGR even at relatively small density. For $n \sim 0.15$, the BGR is comparable to E_b, but a small change in TP and EP is observed. For $n > 0.15$ the BGR is larger than $E_B = -1/2$, and the exciton unbinds. The T matrix still shows a peak, originating from a two-particle scattering resonance in the two-particle continuum. However, the TP and EP show small blueshifts. The EP is shifting less than the TP, as a result of its skewness due to the Bose factor in Eq. (4) and the density dependence of the broadenings. Within the contact potential, we compared the above results with the dynamical Hartree-Fock (HF) results (Fig. 3). A fixed broadening had to be introduced by hand in the HF, whereas it is self-consistently calculated in our approach. (i) The BGR in this HF model stems from the Hartree term alone, and is simply $-2n$. In the SCLA it is smaller for $n < 0.1$, and then larger for $n > 0.1$. (ii) The HF model produces small EP blueshifts for $n < 0.15$: the vertex correction (reduction of exciton binding energy) in the emission of photons partially compensates the single-particle energy shift [8]. This was also remarked for the wires [7]. (iii) The blueshift is reduced in the SCLA. This is related to the density-dependent broadenings of the SCLA. Introducing a density-dependent broadening in the HF model could mimic this result, but is clearly beyond the HF approximation, and the scope of the present study. (iv) For $n \gg 0.8$, shifts from the HF and

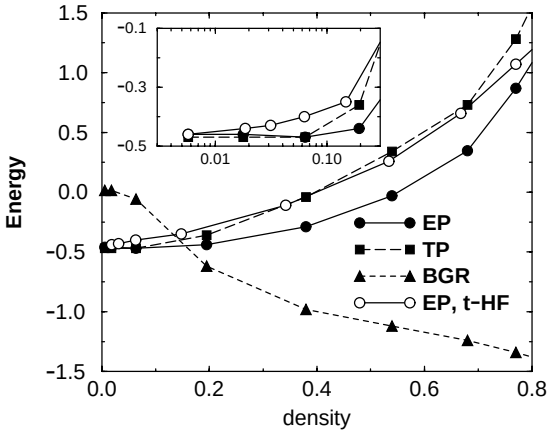

FIG. 3. Emission peak energy (EP), emission peak energy in HF (EP, t-HF), the exciton energy (TP), and BGR as functions of the density. Inset: An enlargement for the lower densities.

the SCLA coincide, given the larger weight of (high-energy) uncorrelated carriers in the system.

Direct measurement of the BGR in the wires is problematic, as absorption is prohibitively weak: emission only is usually measured. Moreover, the value of the BGR is also sensitive to the structure of the realistic Coulomb potential, and critically depends on the strength of lateral confinement [16–18]: in 1D, the exciton binding energy substantially increases with better lateral confinement [19]. In the first experiments of Cingolani *et al.* [20], the samples featured a relatively weak lateral confinement, and exciton binding comparable to 2D [19]. Thus, vertex corrections in the emission were presumably weak, and the fit of emission with simpler free-carrier models substantially reproduced accurate calculations of the BGR [16]. Even in this case, the band gap was located well below the EP. In later samples, with stronger lateral confinement, the role of correlations and vertex corrections was enhanced. Greus *et al.* [7], using the SBE, noticed that it was difficult to extract the BGR from emission, as the position of the peak is not directly related to it and stays rather fixed [7]. They also remarked that large inhomogeneous broadenings, comparable to the subband separation, do not allow one to discriminate fine features such as small blueshifts in the EP; n is known with poor accuracy; T depends on time, and has to be fitted from the emission, entailing additional uncertainties in the comparison of theory with experiment. We reiterate that our results stem from a short-range potential, and important effects of long-range interactions and dependences on lateral confinement are missed. They have to be considered, with reserve for real systems, as giving interesting indications only for the strongly 1D systems, featuring large E_B and correlations. We argue that they clearly reinforce the conclusions of Ref. [7], that extracting the BGR from emission in the current samples is not trivial. Moreover, deducing the existence of (bound) excitons and the absence of BGR even at large densities from the observation of a peaked emission, as reported in Ref. [21], is also misleading, as we also predict a peaked emission well after excitons disappear, and BGR is significant. Finally, we wish to mention that the large blueshifts predicted by both SCLA and HF within the contact potential at $n > 0.5$ are related to a sizeable depth of the Fermi sea, when the emission height saturates, and peaks are considerably broadened and skewed to higher energy (Fig. 2, $n = 0.68$). Although a saturation of the emission height as a function of density was observed, large blueshifts and skewness were not [7,20,21]. The large blueshift could be thus an artifact of the model potential we use, showing that quantitative comparison with experiment is beyond its purpose. Moreover, even for the latest measurements on samples with a subband separation of about 20 meV [21], the higher subbands become populated well before $n > 0.4$ in our units, and a multisubband analysis should be carried out anyway for $n > 0.4$.

In conclusion, we have introduced the self-consistent ladder approximation, simplified for a short-range potential. For temperatures smaller or comparable to the exciton binding energy, it includes important electron-hole correlation effects on the propagation of carriers: These result in the formation and scattering of bound pairs at low density, but they are also present at higher density when pairs unbind. These correlation effects show in the optical emission as frequency- and density-dependent broadenings, relevant corrections to the band gap renormalization calculated in the Hartree-Fock approximation, and a reduction of the overestimated vertex correction in the emission of light.

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