Many-Body Renormalization of Semiconductor Quantum Wire Excitons: Absorption, Gain, Binding, Unbinding, and Mott Transition

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We consider theoretically the formation and stability of quasi-one dimensional many-body excitons in GaAs quantum wire structures under external photoexcitation conditions by solving the dynamically screened Bethe-Salpeter equation for realistic Coulomb interaction. In agreement with several recent experimental findings the calculated excitonic peak shows very weak carrier density dependence up to (and even above) the Mott transition density, \( n_c \sim 3 \times 10^5 \text{ cm}^{-1} \). Above \( n_c \) we find considerable optical gain demonstrating compellingly the possibility of one-dimensional quantum wire laser operation.

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An exciton, the bound Coulombic ("hydrogenic") state between an electron in the conduction band and a hole in the valence band, is an (extensively studied) central concept in semiconductor physics. Recent interest has focused on low dimensional excitons in artificially structured semiconductor quantum well or wire systems where carrier confinement may substantially enhance the excitonic binding energy leading to novel optical phenomena. In this Letter we consider the formation, stability, and optical properties of one dimensional (1D) excitons in semiconductor quantum wires, a problem which has attracted a great deal of recent experimental [1-3] and theoretical [4-6] attention. Our motivation has been a number of recent puzzling experimental observations [1,2], which find the photoluminescence emitted from an initially photoexcited semiconductor quantum wire plasma to be peaked essentially at a constant energy independent of the magnitude of the photoexcitation intensity. This is surprising because one expects a strongly density-dependent "red shift" in the peak due to the exchange-correlation induced band gap renormalization (BGR) (i.e. a density-dependent shrinkage of the fundamental band gap due to electron and hole self-energy corrections), which should vary strongly as a function of the photoexcited electron-hole density [7-9]. This striking lack of any dependence of the observed photoluminescence peak energy on the photoexcitation density has led to the suggestion [1,2] that the observed quantum wire photoluminescence may be arising entirely from an excitonic (as opposed to an electron-hole plasma (EHP)) recombination mechanism, and the effective excitonic energy is, for unknown reasons, a constant (as a function of carrier density) in 1D quantum wires. This, however, introduces a new puzzle because one expects the excitonic level to exhibit a "blue shift" (i.e. an increase) as a function of carrier density as the Coulomb interaction weakens due to screening by the finite density leading to a diminished excitonic binding energy. Thus the only way to understand the experimental observation is to invoke a near exact cancelation between the red-shift arising from the self-energy correction induced BGR and the blue-shift arising from screening induced excitonic binding weakening. In this Letter, focusing on the photoexcited quasi-equilibrium regime, we provide the first quantitative theory for this problem by solving the full many-body dynamical Bethe-Salpeter equation for 1D excitons. We include both self-energy renormalization and vertex correction (arising from the Coulomb interaction) on an equal footing under high photoexcitation conditions. We find that, in agreement with experimental observations, our calculated effective excitonic energy (indicating the luminescence peak frequency) remains essentially a constant (with an energy shift of less than 0.5 meV) as a function of 1D carrier density \( n \) for \( n < n_c \sim 3 \times 10^5 \text{ cm}^{-1} \) with the system making a Mott transition from an insulating exciton gas of bound electron-hole pairs \( n < n_c \) to an EHP \( n > n_c \) at \( n = n_c \). For \( n > n_c \) we find strong optical gain in the calculated absorption spectra.

For our results to be presented here we have considered quantum wire parameters [1] corresponding to the T-junction structure of width 70 Å in both transverse directions with only the lowest 1D subband occupied by the carriers. But our results and conclusions should be generically valid for arbitrary 1D quantum wire confinement (e.g. the V-groove wire of Ref. 2). The many-body exciton is given by the so-called Bethe-Salpeter equation [10] for the 2-particle Green’s function which is shown diagrammatically in Fig. 1. The many-body diagrams shown in Fig. 1 correspond to a rather complex set of coupled non-linear integral equations which must be solved self-consistently with the bare interaction being the Coulomb interaction. These equations are notoriously difficult [10] to solve without making drastic approximations. We use the parabolic band effective mass approximation considering the highest valence and the lowest conduction band only. The simplest approximation is to neglect all many-body effects and consider the one-electron problem when self-energy (Fig. 1(b)) and screening (Fig. 1(c)) effects disappear leaving the standard [4] excitonic binding problem (Fig. 1(a)) for a conduction band electron and a valence band hole in-
teracting via the effective "one dimensional" Coulomb interaction. Even this zeroth order exciton problem for quantum wires is far from trivial, however, because one must include proper quantum confinement effects in the Coulomb interaction matrix elements appropriate for the specific quantum wire geometry of interest. Not surprisingly a rather large theoretical literature \cite{4,5} exists in treating this zeroth order one-electron quantum wire exciton problem, which is the effective dilute or zero density ($n \to 0$) limit of the many-body problem of interest to us. We include quantum wire confinement effects appropriate for a T-junction system in all the results presented in this paper.

In carrying out the full many-body dynamical calculation for the Bethe-Salpeter equation we are forced to make some approximations. Our most sophisticated approximation uses the fully frequency dependent dynamically screened electron-hole Coulomb interaction in the single plasmon-pole random phase approximation (Fig. 1(c)), which has been shown to be an excellent approximation \cite{11} for 1D quantum wire dynamical screening. It is essential to use the actual Coulomb interaction in solving this problem and the simplistic model interactions (such as the delta function zero range interaction used recently in Ref. 6) are not particularly meaningful from either a theoretical perspective or in understanding experimental data. In addition to our full dynamical screening theory (which is computationally extremely difficult) we have also carried out a number of simpler approximations (to be described below) in order to assess the quantitative contributions of various physical mechanisms to the 1D many-body exciton formation. For the self-energy correction we use the single-loop GW diagram shown in Fig. 1(b). Ward Identities then fix the vertex correction, entering Fig. 1(a), to be the appropriate ladder integral equation.

Before solving the full Bethe-Salpeter equation, it is instructive to study the excitonic and EHP effects separately by treating the influence of the plasma on the excitonic states as a perturbation \cite{10}. Using an effective Hamiltonian derived from the Bethe-Salpeter equation, we can obtain the exciton energy by minimizing the energy expectation value variationally through an 1s excitonic trial wave function. The BGR is calculated by the GW approximation (Fig. 1(b)). Note that the variational calculation is quantitatively valid only when the exciton-plasma hybridization is not particular important. In Fig. 2 we show our calculated zero-temperature (variational) excitonic energy and BGR separately as a function of 1D electron-hole density. The full dynamical screening solution is shown as the solid line and the quasi-static screening approximations (described below) is shown as the dashed line. For the purpose of comparison we also show as an inset in Fig. 2 the purely one-electron static screening result where the electron-hole interaction is modeled by the density dependent statically screened 1D Coulomb interaction, and all many-body effects (e.g. BGR) are ignored. The exciton binding energy shows a monotonic decrease ("blue-shift") in the inset.
with the experimental finding of an approximately constant excitonic peak independent (at least in some finite range) of the free carrier density. We find [12] that this large blue shift is not cancelled by the many-body self-energy effects within the same static screening approximation. Therefore, it is essential to consider the dynamical effects when one calculates the excitonic effects in quasi-1D quantum wire systems. Inclusion of dynamical many-body effects, shown in the results in the main part of Fig. 2, qualitatively modifies the situation: (1) the effective many-body excitonic energy is almost the same in the low density limit ($\sim 13$ meV for $n < 10^4$ cm$^{-1}$) in all the approximations; (2) for density between $10^4$ and $10^5$ cm$^{-1}$ the exciton energy has a few meV red-shift in the quasi-static approximation and almost no shift (less than 0.5 meV blue-shift) in the dynamical screening approximation; (3) the Mott transition density for the quasi-static approximation is about $10^5$ cm$^{-1}$, while it is about $3 \times 10^5$ cm$^{-1}$ for the dynamical theory; (4) below $n_c$ our variational solution corresponds to an excitonic wavefunction which is that of a bound electron-hole pair in the 1s hydrogenic state with a radius of about 100-500 Å [12], and this description is approximately valid with a constant (variational) ground state energy upto $n_c$ [12]; (5) above $n_c$ the calculated effective excitonic wave function is completely delocalized (with a very large radius) and the EHP becomes the dominant state of the system; (5) the quasi-static approximation, while being qualitatively valid, is quite poor quantitatively compared with our dynamical screening approximation.

In Fig. 3, we show our calculated absorption and gain spectra by solving the full Bethe-Salpeter equation in the quasi-static and the dynamical screening approximations. The integral equation for the two-particle Green’s function (Fig. 1(a)) is solved by the matrix inversion method with a singular kernel [10] which arises from the singularity of the Coulomb interaction. The full dynamical screening approximation (which has never been solved in the literature before) has a multi-singular kernel with multiple momentum-dependent singularities (poles of the integrand) which arise from the many-body hybridization of photons, single particle excitations, and plasmons. This makes the usual singularity-removal method ineffective. This fact forces us to use a rather large matrix (about $1500 \times 1500$ in a Gaussian quadrature) in the matrix inversion [10] method in order to get good overall accuracy. We now discuss the important features of Fig. 3: (1) There are generally two absorption peaks in the low density ($n < 10^4$ cm$^{-1}$) spectra, one is the exciton peak at 1537 meV and the other one is the band edge peak at, for example, 1547.5 meV for $n = 10^2$ cm$^{-1}$ in Fig. 3(b). The exciton peak has much larger oscillator strength which occurs as the exciton gas becomes an EHP at some high density ($n_c$). The statically screened single exciton behavior shown in the inset of Fig. 2 disagrees completely (induced by static screening) as the exciton eventually merges with the band continuum with a Mott transition density $n_c \sim 10^5$ cm$^{-1}$. The quasi-static approximation [10], shown as dashed lines in Fig. 2, involves making the screened exchange plus Coulomb hole approximation in the self-energy diagrams neglecting the correlation hole effect. The simpler approximations (static screening and quasi-static) are done in order to assess the importance of various terms in the full dynamical Bethe-Salpeter equation which is extremely difficult and computationally time-consuming to solve in the RPA dynamical screening approximation.

The Mott transition may be thought of as the unbinding of the bound electron-hole pair in the exciton to a free electron and a free hole — it is therefore effectively an interaction-induced insulator to metal transition which occurs as the exciton gas becomes an EHP at some high density ($n_c$). The statically screened single exciton behavior shown in the inset of Fig. 2 disagrees completely with the experimental finding of an approximately constant excitonic peak independent (at least in some finite range) of the free carrier density. We find [12] that this large blue shift is not cancelled by the many-body self-energy effects within the same static screening approximation. Therefore, it is essential to consider the dynamical effects when one calculates the excitonic effects in quasi-1D quantum wire systems. Inclusion of dynamical many-body effects, shown in the results in the main part of Fig. 2, qualitatively modifies the situation: (1) the effective many-body excitonic energy is almost the same in the low density limit ($\sim 13$ meV for $n < 10^4$ cm$^{-1}$) in all the approximations; (2) for density between $10^4$ and $10^5$ cm$^{-1}$ the exciton energy has a few meV red-shift in the quasi-static approximation and almost no shift (less than 0.5 meV blue-shift) in the dynamical screening approximation; (3) the Mott transition density for the quasi-static approximation is about $10^5$ cm$^{-1}$, while it is about $3 \times 10^5$ cm$^{-1}$ for the dynamical theory; (4) below $n_c$ our variational solution corresponds to an excitonic wavefunction which is that of a bound electron-hole pair in the 1s hydrogenic state with a radius of about 100-500 Å [12], and this description is approximately valid with a constant (variational) ground state energy upto $n_c$ [12]; (5) above $n_c$ the calculated effective excitonic wave function is completely delocalized (with a very large radius) and the EHP becomes the dominant state of the system; (5) the quasi-static approximation, while being qualitatively valid, is quite poor quantitatively compared with our dynamical screening approximation.

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meV, consistent with the result shown in Fig. 2 which was obtained variationally. (4) Consistent with the variational energy shown in Fig. 2, the excitonic peak of the full dynamical screening approximation is almost a constant (with only a 0.5 meV blue-shift) up to \( n_c \). (5) Below the Mott density (\( n_c \sim 3 \times 10^{5} \) cm\(^{-1} \)) the oscillator strength of the excitons decreases rapidly as the carrier density increases in the quasi-static approximation; however, in the full dynamical theory the strength of the exciton peak remains almost a constant with increasing carrier density, indicating the interesting prospect of excitonic lasing in 1D quantum wires. (6) In the dynamical screening approximation, considerable excitonic gain is seen for \( n \sim n_c \) without any observable energy shift in the spectrum. We find that at very high densities (\( n > n_c \sim 3 \times 10^{5} \) cm\(^{-1} \)) the excitonic features in the absorption spectra are smeared out by the EHP continuum, and the BGR induced red-shift is observed. These very high density results will be presented elsewhere [12].

We note that our dynamical screening Bethe-Salpeter equation results are in excellent qualitative and quantitative agreement with the recent experimental findings [1,2]. In particular, the effective constancy of the exciton peak as a function of the photoexcited carrier density as well as the possibility of excitonic absorption and lasing well into the high density regime (even for \( n > n_c \sim 3 \times 10^{5} \) cm\(^{-1} \)) turn out to be characteristic features of the full dynamical theory (but not of the static and the quasi-static approximation). A full dynamical self-consistent theory as developed in this Letter is thus needed for an understanding of the recent experimental observations. We also note that in the recent literature the Mott density for 1D GaAs quantum wire systems has often been quoted as \( n_c \sim 8 \times 10^{5} \) cm\(^{-1} \) which is substantially higher than our full dynamical theory result, \( n_c \sim 3 \times 10^{5} \) cm\(^{-1} \). The higher value of the Mott density (\( n_c \sim 8 \times 10^{5} \) cm\(^{-1} \)) follows from a simple estimate based on ground state energy comparison where one equates the calculated density-dependent BGR (the light solid line in Fig. 2) with the zero-density exciton energy (\( \sim 13 \) meV in Fig. 2) — as one can see from Fig. 2, the calculated BGR (the light solid line in Fig. 2) equals 13 meV, the zero-density exciton energy, around \( n \sim 8 \times 10^{5} \) cm\(^{-1} \). In the full interacting theory the Mott transition (the intersection of the light and the heavy solid lines in Fig. 2) moves to a lower density, \( n_c \sim 3 \times 10^{5} \) cm\(^{-1} \), which is also consistent with our full dynamical Bethe-Salpeter equation based calculation of the absorption/gain spectra shown in Fig. 3.

In summary, our main accomplishments reported in this Letter are the following: (1) The first fully dynamical theory of a photoexcited electron-hole system in semiconductors which treats self-energy, vertex corrections, and dynamical screening in a self-consistent scheme based on the GW self-energy and ladder-bubble vertex-polarization diagrams within a realistic Coulomb interaction-based Bethe-Salpeter theory; (2) a reasonable qualitative and quantitative agreement with the recent experimental observations of an effectively (photoexcitation density-independent) constant exciton peak, which in our fully dynamical theory arises from an approximate cancelation of self-energy and vertex corrections in the Bethe-Salpeter equation; (3) an effective 1D quantum wire Mott transition density of \( n_c \sim 3 \times 10^{5} \) cm\(^{-1} \) which is below earlier estimates based on less sophisticated approximations; (4) the concrete theoretical demonstration of the possibility of excitonic gain and lasing in 1D quantum wire structures in the density range of \( n > 3 \times 10^{5} \) cm\(^{-1} \) where considerable optical gain is achieved in our calculated absorption spectra.

In conclusion, we have carried out the first fully dynamical many-body theory for the photoexcited electron-hole plasma in 1D semiconductor quantum wires by solving the Bethe-Salpeter equation treating self-energy ("band gap renormalization") and vertex ("excitonic shift") corrections on an equal footing within the ladder-bubble-GW self-consistent conserving scheme. We find, consistent with a number of hitherto unexplained experimental observations [1-3], that the self-energy and the vertex corrections tend to cancel each other leading to an almost constant (in density) absorption/gain peak all the way to (and considerably above) the Mott transition which occurs around a density of \( n_c \sim 3 \times 10^{5} \) cm\(^{-1} \) for 70 Å wide T-quantum wires.

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