Absorption spectrum of a weakly n-doped semiconductor quantum well

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We calculate, as a function of temperature and conduction band electron density, the optical absorption of a weakly n-doped, idealized semiconductor quantum well. In particular, we focus on the absorption band due to the formation of a charged exciton. We conceptualize the charged exciton as an itinerant excitation intimately linked to the dynamical response of itinerant conduction band electrons to the appearance of the photo-generated valence band hole. Numerical results for the absorption in the vicinity of the exciton line are presented and the spectral weights associated with, respectively, the charged exciton band and the exciton line are analyzed in detail. We find, in qualitative agreement with experimental data, that the spectral weight of the charged exciton grows with increasing conduction band electron density and/or decreasing temperature at the expense of the exciton.

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Following Kheng et al.’s seminal experiment [1], which showed that in a weakly n-doped semiconductor quantum well (QW) photo-generated excitons are accompanied by negatively charged excitons [2], i.e. by bound states comprising two conduction band (CB) electrons and one valence band (VB) hole, numerous theoretical [3–5] and experimental [6–8] studies have been devoted to investigate optical properties of weakly n-doped QWs. There is also growing experimental interest in weakly p-doped QWs where positively charged excitons have been identified [9]. Among the most prominent open questions is the question about the microscopic structure of the trion. (We focus here on n-doped QWs and use the term ‘trion’ to refer to a negatively charged exciton.) In particular, it is still an unresolved issue, whether the trion is a localized, defect-like excitation or an itinerant excitation, freely propagating in the QW plane. There is experimental evidence that in modulation doped QWs the trion is indeed trapped by the potential fluctuations of the remote donors [10]. On the other hand, in optically doped, high-quality QWs, where trions have been also unambiguously observed (see, for example, Ref. [11]), remote donors are absent. Moreover, potential fluctuations caused by interface roughness, which could also act as a source for localization, are presumably very small. Therefore, the trion in these samples is most likely freely propagating. Evidently, the broad theoretical task is to develop a trion theory for realistic QWs treating both possibilities on an equal footing, which is however rather formidable. As a first step, we focus therefore on an idealized QW, neglecting localization processes altogether. In particular, we demonstrate, that in such an idealized, high-quality sample, it is the dynamical response of itinerant CB electrons to the appearance of the photo-excited VB hole, which, at low densities, inevitably gives rise to an itinerant trion and to the experimentally observed absorption band below the exciton line. In contrast to previous few-body theoretical studies of the trion ground-state properties [12,13] and the trion absorption band [14], our genuine many-body theoretical approach accounts for both trion and exciton. Moreover, because the dynamical response of CB electrons is also of utmost importance at high density [15], our approach indicates how a complete lineshape theory for n-doped QWs, covering the whole density range, could be constructed.

Since the details of the electronic band structure of a finite width QW unnecessarily masks the many-body theoretical concepts we are envoicing, we introduce our approach for a strictly two-dimensional (2D) idealized QW with two (isotropic) parabolic bands. Of course, with an appropriate choice of single particle states our formalism can be also applied to a more realistic QW model. Our calculation is based on linear response theory which relates the (interband) optical susceptibility $\chi(\omega)$ to the eh-pair propagator $P(12;\omega)$ [12]. Using techniques developed in nuclear many-body physics [13], $P(12;\omega)$ satisfies a Dyson equation (here ‘1’ etc. stands for 2D momentum and spin variables, see below however, and summation convention is implied),

$$P(12;\omega) = P^{(0)}(12;\omega) + P^{(0)}(13;\omega) \times [M^{st}(34) + \delta M(34;\omega)]P(42;\omega), \quad (1)$$

with an eh-pair self-energy containing a static part, $M^{st}(34)$, and a dynamic part $\delta M(34;\omega)$. For a momentum independent transition matrix element $r_{ec}$, the optical susceptibility $\chi(\omega) = |r_{ec}|^2 \sum_{12} P(12;\omega)$, the imaginary part of which determines the optical absorption, i.e. apart from a constant factor $I_{abs}(\omega) \sim -Im\chi(\omega)$.

The Dyson equation is valid for arbitrary CB electron densities. We are concerned, however, with low densities, where, in leading order, CB electrons do neither modify the wavefunction nor the energy of bound states (i.e. exciton and trion). Accordingly, we can ignore Pauli blocking as well as screening and treat CB electrons solely as a reservoir for bound state formation. We can, furthermore, neglect (first order) electron and hole self-energy corrections. Then,

$$P^{(0)}(12;\omega) = \frac{\delta_{12}}{\omega + \eta - \epsilon_c(1) - \epsilon_e(-1)}, \quad (2)$$

$$M^{st}(34) = -\nu(34), \quad (3)$$
with $e_{\nu/c}(1)$ and $v(34)$ denoting, respectively, the bare 2D conduction and valence band dispersion relations and the unscreened 2D Coulomb interaction. The dynamical response of CB electrons, i.e., the creation of virtual $ee$ pairs, is contained in the dynamical part $\delta M(34;\omega)$ which is related to a eight-point ($eeeh$) function $R$. In the dilute limit we can show, however, using again techniques developed in Ref. [13], together with a low-density (or $\epsilon$-line) expansion for $\delta M(34;\omega)$, that the eight-point function $R$ featuring in $\delta M(34;\omega)$ factorizes, in leading order of the CB electron density, into a two-point function describing the (free) propagation of a CB hole and a six-point function $G$ describing the (correlated) propagation of an $eeh$ cluster (see Fig. 1). Hence, at low densities, due to the creation of a “single” virtual $ee$ pair, optically generated $eeh$ pairs are intrinsically coupled to $eeh$ clusters. If we then employ the spectral representation for $G$, keeping only bound $eeh$ clusters, which we expect to influence the optical response most dramatically, and anticipating that the trion, i.e. the groundstate of the bound $eeh$ cluster, has anti-parallel electron spins, we find (now a bold ‘1’ etc. denotes only 2D momentum variables)

$$\delta M(34;\omega) = \sum_{5n} f_e(5) \frac{\Xi_{5n}(3)\Xi_{5n}^*(4)}{\omega + i\bar{\eta} + e_\nu(5) - \Omega_{5n}(5)},$$  \hspace{1cm} (4)

where all multiple $eeh$ scattering processes are encoded in a vertex function $\Xi_{5n}(3) = \sum_{7\delta} v(57)[\Psi_{5n}(37) - \Psi_{5n}(73 + 57)]$, given in terms of a momentum-space $eeh$ wavefunction $\Psi_{5n}(37)$, whose Fourier transform to real space satisfies a 2D $eeh$ Schrödinger equation with eigenvalue $\Omega_{5n}(5)$. Here, ‘$n$’ and ‘5’ depict, respectively, internal quantum numbers and the (center of mass) momentum of the (propagating) bound $eeh$ cluster. The Fermi function, $f_e(5)$, coming form the free propagation of the CB hole, accounts for the reservoir of CB electrons. Note, for vanishing CB electron density $\delta M(34;\omega) \rightarrow 0$ and Eqs. (3) – (5) reduce to the ladder approximation for the exciton.

To demonstrate that $\delta M(34;\omega)$ gives rise to the absorption band below the exciton line, we approximately solve Eqs. (3) – (5) for energies $\omega$ close to the exciton groundstate energy $E_X$. We adopt the following strategy: i) We keep in Eq. (5) only the groundstate of the bound $eeh$ cluster, i.e. the trion denoted by $n = T$, whose wavefunction and energy we determine with a variational technique developed by Stébé and coworkers [2]. ii) We project Eq. (4) to the exciton groundstate. Ignoring the coupling between the exciton groundstate and excited exciton states, the projected $eh$-propagator becomes $P_X(\omega) = [\omega + i\bar{\eta} - E_X - \delta M_X(\omega)]^{-1}$, with an exciton self-energy $\delta M_X(\omega) = \sum_{34} \phi_X(3)\delta M(34;\omega)\phi_X(4)$ (where $\phi_X(3)$ is the momentum-space wavefunction for the exciton groundstate). Evidently, this is only meaningful for large enough exciton binding energies and, of course, only valid for $\omega$ in the vicinity of $E_X$. iii) Using the variational trion wavefunction, we work out an analytical expression for $\delta M_X(\omega)$ for non-degenerate CB electrons, i.e. we assume low enough CB electron densities such that the Fermi energy is sufficiently inside the energy gap and that the Fermi function in Eq. (5) can be replaced by a Boltzmann function parametrized by the CB electron density $\Xi_{5n}(3)$. For simplicity we also neglect the weak dependence of $\Xi_{5n}(3)$ on the trion momentum $5$.

In terms of $P_X(\omega)$, the optical absorption in the vicinity of the exciton resonance reads $I_{abs}(\omega) \sim 2|\epsilon_{\nu/c}|^2|S_X|^2|\delta M_X(\omega)|$, with $S_X = \sum_1 \delta_X(1)$. We now introduce dimensionless quantities, measuring energy from the energy gap $E_g$ in units of the 2D exciton binding energy $4R$ (R being the 3D exciton Rydberg), and length in units of the 3D Bohr radius $a_B$. Specifically, we define $\omega = [\omega - E_g]/4R$ and combine all numerical factors into a constant $I_0$. Then, $I_{abs}(\omega) = I_0 \cdot D_X(\omega)$, with a dimensionless and, due to the spectral weight sum rule [13], normalized spectral (or lineshape) function

$$D_X(\omega) = \frac{1}{\pi} \frac{\bar{\eta} + \Gamma_X(\omega)}{|\omega + 1 - \Delta_X(\omega)|^2 + [\bar{\eta} + \Gamma_X(\omega)]^2},$$  \hspace{1cm} (5)

Here $\bar{\eta} = \eta/4R$ and the dimensionless functions $\Delta_X(\omega)$ and $\Gamma_X(\omega)$ are related to $\delta M_X(\omega) \equiv \delta M_X(4R\omega + E_g)/4R$. Specifically,

$$\delta M_X(\omega) = \bar{\beta} \frac{M_T}{M_X}(na_B^2)|u_T^X|^2I(\omega) - \Delta_X(\omega) - i\Gamma_X(\omega),$$  \hspace{1cm} (6)

where $\bar{\beta} = 4\beta R$, $M_X$, $M_T$, and $n$ are, respectively, the thermal energy measured in units of $E_R$, the exciton mass, the trion mass, and the CB electron density. $u_T^X$ is the dimensionless exciton-trion coupling constant, given by a real space integral involving the variational trion wavefunction, the Coulomb interaction, and the exciton wavefunction, and

$$I(\omega) = \int_0^\infty dy \frac{e^{-\frac{\beta M_T}{4R y}}}{\omega + i\bar{\eta} - \bar{\epsilon}_T + y},$$  \hspace{1cm} (8)

with $\bar{\epsilon}_T = [\Omega_T(0) - 2E_g]/4R$ denoting the energy of a trion at rest, i.e. the internal trion energy, which is the binding energy of two CB electrons to a VB hole. The assumption of non-degenerate CB electrons constrains the temperature and density range in which our theory is valid. Specifically, using the well-known formula, $\beta\mu = \log(\exp(\pi n\beta/m^*_e) - 1)$, which relates the chemical potential $\mu$ of a 2D gas of free electrons (with mass $m^*_e$) to its density $n$, we estimate $\bar{\beta}(na_B^2) < 1$, i.e. depending on the density the temperature has to be sufficiently high.

We now discuss representative numerical results for an effective mass ratio $\sigma = m^*_e/m^*_h = 0.146$ corresponding to GaAs. To obtain the exciton-trion coupling constant and the internal trion energy, we specifically used, following Stébé and coworker [2], a 2D (22-term) Hylleraas
corresponding to previously obtained results [2,5]. The spectral function with experimental results. Below a narrow peak in the small frequency range, we observe a broad absorption band characterized by a sharp high energy edge at \( \tilde{\omega} = -1.1151 \) and a low-energy tail. To facilitate a microscopic understanding of the absorption features, we plot in the right inset of Fig. 2 real and imaginary part of the exciton self-energy. The narrow peak is obviously a consequence of the pole of the spectral function \( D_X(\tilde{\omega}) \) at \( \tilde{\omega} = -0.94 \equiv \tilde{\epsilon}_X \), where \( \tilde{\epsilon}_X \) is the energy which simultaneously satisfies \( \Delta_X(\tilde{\epsilon}_X) = \tilde{\epsilon}_X + 1 \) and \( \Gamma_X(\tilde{\epsilon}_X) = 0 \). A pole in a spectral function is usually interpreted as a quasi-particle. Accordingly, we attribute the narrow peak in Fig. 2 to a quasi-exciton. We also see, the peak has actually no width, because, due to the assumed non-degeneracy of CB electrons, \( \Gamma_X(\tilde{\epsilon}_X) = 0 \). The origin of the broad absorption band, in contrast, is the itinerant trion. The sharp high energy edge, for example, comes from the pole of \( \Delta_X(\tilde{\omega}) \) at the trion formation energy \( \tilde{\epsilon}_T = -1.1151 \), i.e. the high energy edge of the band corresponds to a trion with momentum zero. The low energy tail, on the other hand, is due to trion states with finite momenta. Note, due to the large value of \( \Gamma_X(\tilde{\omega}) \) in the vicinity of the edge, trion states with small momenta are significantly damped. Accordingly, the maximum of the absorption due to the trion is inside the band, corresponding to the creation of a trion with finite momentum. The maximum coincides with the high energy edge only for very low CB electron density, in agreement with the calculations of Ref. [1]. The spectral weight of the band, however, is then also very small (see Figs. 3 and 4 below). To analyze the spectral weights associated with, respectively, the trion absorption band and the quasi-exciton line, we show in the left inset of Fig. 2 the integrated spectral weight up to an energy \( \tilde{\omega} \) defined in terms of the cumulant \( C_X(\tilde{\omega}) = \int_{-\infty}^{\tilde{\omega}} d\tilde{E} D_X(\tilde{E}) \). As can be seen, the integrated spectral weight of the trion absorption band corresponds to the plateau value just below \( \tilde{\omega} = -1 \), i.e. \( f_T = C_X(-1) \). The inset also demonstrates that the total spectral weight adds up to one, as it should, due to the spectral weight sum rule. Accordingly, the spectral weight of the quasi-exciton is given by \( f_X = 1 - f_T \).

We now present a quantitative investigation of the integrated spectral weights as a function of CB electron density and temperature. To that end, we calculated for various trion absorption bands the integrated spectral weights \( f_T = C_X(-1) \) and deduced from the sum rule, \( f_X = 1 - f_T \), the corresponding spectral weights for the quasi-exciton. Fig. 3 depicts typical trion absorption bands used to obtain the data shown in Fig. 4. As expected, for fixed temperature, \( f_T/f_X \) grows with CB electron density because the number of CB electrons available for trion formation increases. The density dependence is not linear, however. Instead, we observe an initial fast increase and then a pronounced slowing down indicating the subtle interplay between trion and quasi-exciton. At high CB electron densities, beyond the validity range of our approach, we anticipate an even more complicated behavior. \( f_T/f_X \) should show non-trivial features especially for densities close to a critical density, where the trion is expected to break up into an exciton and a free CB electron. For fixed CB electron density, \( f_T/f_X \) increases with decreasing temperature. The increase also tends to saturate. We emphasize, the numerical results for \( f_T/f_X \) are consistent with experimental data, despite the simplicity of the QW model, which neglects, e.g., the finite width of the QW. Unfortunately, experimentally achieved CB electron densities are above the density range in which our theory is directly applicable. A quantitative comparison of, for example, line-shapes and oscillator strengths, requires, besides a more realistic QW model, to take modifications of the exciton and trion wavefunctions and energies due to CB electrons explicitly into account. Our approach is feasible as long as bound states are stable. We only have to decorate various expressions with Pauli blocking factors, \( f_X = 1 - f_T \), and use a screened Coulomb interaction. The numerical implementation is rather involved, however, and is beyond the scope of this paper [10].

To summarize, we investigated the optical absorption due to exciton and trion formation in a weakly n-doped idealized QW, conceptualizing the trion as an itinerant excitation formed in the course of the dynamical response of itinerant CB electrons to the photo-generated VB hole. In particular, we focused, in an exploratory calculation, on low CB electron densities, treating CB electrons as a reservoir for bound state formation. The overall structure of the calculated absorption is consistent with experimental data indicating that, at least in high-quality samples, the dynamical response of CB electrons might play a key role in the formation of trions.

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pressed by phase space factors. The hatched box depicts the
ing events involving a conduction band hole are strongly sup-
approximation \([13]\). The remaining three diagrams have the
same structure except that the first (last) scattering events
are \( eeh \) (\( e'eh \)), \( ee eh \) (\( e'eh \)), and \( ee ee \) (\( e'eh \)), respectively. Since we focus
on the groundstate of the \( \bar{\delta} M \) cluster, we restrict the (inter-
mediate) spin summations in these diagrams to anti-parallel
electron spins. All spin summations can then be done explicit-
yielding an overall factor of 2 in the final expression for \( \chi(\omega) \).

FIG. 1. Diagrammatic representation of one term (out of four) contributing to \( \delta M(34; \omega) \). Solid lines in forward (backward) direction stand for \( e (e') \) propagation, wavy lines depict Coulomb interactions and dashed lines denote \( h \) propagation. In the low density limit the \( e' \) line is free because scattering events involving a conduction band hole are strongly sup-
pressed by phase space factors. The hatched box depicts the
\( eeh \) propagator \( G \) to be determined in the Faddeev-Watson approxima-
tion \([13]\). The remaining three diagrams have the
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FIG. 2. The spectral function \( D_X(\tilde{\omega}) \) for \( na_\beta^2 = 0.008 \) and
\( 4R\beta = 10 \). The effective mass ratio \( \sigma = 0.146 \), corresponding
to GaAs, and \( \tilde{\eta} = 0.001 \). The left inset depicts the integrated spectral weight \( C_X(\tilde{\omega}) \), whereas the right inset dis-
plays \( \tilde{\Gamma}_X(\tilde{\omega}) \) (dot-dashed line) and \( \Delta_X(\tilde{\omega}) \) (solid line). The energy for which \( \Delta_X(\tilde{\omega}) \) crosses \( \tilde{\omega} + 1 \) (dotted line) defines the quasi-exciton line at \( \tilde{\omega} = -0.94 \).

FIG. 3. The left and right panels show, respectively, trion
absorption bands as a function of CB electron density for a
fixed temperature \( 4R\beta = 10 \) and as a function of temperature
for a fixed CB electron density \( na_\beta^2 = 0.01 \). The quasi-exciton
line located at \( \tilde{\omega} > -1 \) is not shown. The effective mass ratio
\( \sigma = 0.146 \), corresponding to GaAs, and \( \tilde{\eta} = 0 \). For clarity
absorption bands corresponding to different parameters are
artificially shifted along the vertical axis.

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FIG. 4. The ratio of the integrated spectral weights $f_T/f_X$ as a function of CB electron density and temperature. The effective mass ratio $\sigma = 0.146$, corresponding to GaAs, and $\tilde{\eta} = 0$. 