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Mott transition of excitons in GaAs-GaAlAs quantum wells

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Abstract. We investigate the breakup of bound electron–hole pairs, known as Mott transition of excitons, in GaAs-GaAlAs quantum wells with increasing excitation, comparing two different theoretical approaches. Firstly, a thermodynamic approach is used to investigate the ionization equilibrium between electrons, holes and excitons, where the abrupt jump of the degree of ionization from 0 to 1 indicates the Mott density. It is extended to a self-consistent quasi-particle approximation (QPA) for the carrier properties, including dynamical screening of the Coulomb interaction between carriers. Secondly, a spectral approach based on the semiconductor Bloch equations within linear optical response is used, considering the quasi-particle (QP) properties of carriers and the dynamical screening between electron–hole pairs. While the first is effectively a one-particle approach, in the second the whole two-particle spectrum is analyzed. Within the thermodynamic approach, a simple criterion for the Mott transition can be given: namely, if the sum of chemical potentials of carriers, reflecting the effective shrinkage of the band edge, crosses the exciton energy with increasing excitation. We demonstrate that this simple picture cannot be maintained in the two-particle approach. Here, a compact quantity, which describes the behavior of the band edge, does not exist. In fact, the behavior of the single states in the spectrum is generated by the interplay of dynamical screening in the interband self-energy and the effective interaction of the electron–hole pairs. Moreover, the band edge cannot be clearly resolved,

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since it is merged with excited exciton states (e.g. 2s state), which show up only for densities far below the Mott density. Instead of a Mott density, only a density range can be given, where the Mott transition appears. We demonstrate that a small damping as a prerequisite for the validation of the extended QPA in the thermodynamic approach breaks down, analyzing (i) the dephasing processes with increasing excitation, (ii) the strong increase of the excitonic linewidth and (iii) comparing with the lifetime of carriers in the QP description.

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1. Introduction

Description of the composition of a partially ionized electron–hole plasma (EHP), consisting of electrons, holes and excitons, in terms of the density of particles and the temperature, is an old problem in semiconductor physics. The breaking of the excitonic binding can be caused by either thermal ionization or, with increasing density, the screening of the Coulomb interaction between carriers. The latter phenomenon is usually referred to as the Mott transition.

Many experiments have shown that the exciton energy stays nearly unchanged with increasing excitation (for early experiments, see [1–4]), while the exciton is broadened and vanishes from the spectra due to the decreasing band edge. In [5], this behavior was founded theoretically, including the dynamic screening between excited carriers in a Bethe–Salpeter equation. It was shown that the nearly unchanged exciton energy with increasing excitation is caused by a wide cancelation of the dynamically screened two-particle self-energy with the dynamically screened effective two-particle interaction. The important point here is that both screening effects have to be taken into account on the same footing. Only in this case their cancelation is given.

Simplifying the dynamically screened self-energy in a quasi-static approximation by the Coulomb hole (CH) self-energy, as applied in [6, 7] for the calculation of gain spectra, the cancelation with the effective static screened interaction is strongly reduced and the exciton undergoes a strong shift to lower energies. In [5], the Lindhard dielectric function in the dynamically screened potential was reduced to a plasmon-pole approximation. The complete Lindhard dielectric function in the dynamically screened potential is included in [8–10].

Besides these more or less numerical extensive investigations, it is of interest to find simple analytical formulae for the Mott density \( n^M \), which can be easily applied in comparison with experiments. A rough estimation of the Mott density is \( n^M a_X^3 = 1 \) [12], where \( a_X \) is the Bohr radius of the exciton. A more physically founded condition \( \kappa a_X = 0.84 \) (\( \kappa \) is the inverse screening length) was found by the authors of [13], investigating the behavior of bound states.
of a statically screened Coulomb potential by the solution of Schrödinger’s equation. Other approaches, based on the renormalization of the energy of carriers due to static screening, were directed toward the investigation of the chemical equilibrium between electrons, holes and excitons in an excited semiconductor as a function of the temperature and the density of particles [14, 15]. This thermodynamic point of view allows the determination of the degree of ionization of the EHP via a mass-action law. This concept is also well known in plasma physics (see, e.g., [15, 16]); applications to the EHP are given in [17–23].

However, the question is whether these approaches applying static screening correspond to those with dynamical screening in the Bethe–Salpeter equation. In [24, 25], it was shown how the concept of dynamical screening with the Bethe–Salpeter equation can be extended to the semiconductor Bloch equations [7, 26]. In contrast to the approaches based on the Bethe–Salpeter equation [5, 8–10], problems arising due to the use of the Shindo approximation are omitted. These problems restrict the validation of these approaches at low temperatures to lower densities, well below the Mott transition. The results of our theoretical concept were confirmed in [11], where it was shown how to circumvent the Shindo approximation in the derivation of the Bethe–Salpeter equation. The theoretical approach was successfully applied to the description of subtleties of polariton propagation in bulk semiconductors [25, 27] and quantum wells [28]. However, the excitation in the experiments was rather low with densities of excited carriers well below the Mott transition. In [29, 30], we extended our theoretical concept to higher excitations and to the description of the Mott transition in bulk ZnSe. The main point here was to include the renormalization of the dynamically screened carrier self-energies and the chemical potentials within quasi-particle approximation (QPA) in a self-consistent way into the semiconductor Bloch equations, e.g. one-particle and two-particle properties are considered on the same footing.

The unbroken interest in the experimental verification of the Mott transition manifests itself in a series of papers, where the buildup of excitons was investigated in the time-resolved photoluminescence (PL) [32–38]. Here, the question of how the optically induced coherent polarization is transferred into an incoherent exciton population was considered. Furthermore, the conditions where the PL is dominated by plasma or by an incoherent exciton population were investigated. Otherwise, terahertz spectroscopy allows us to study the interplay of optically generated excitons and unbound electron–hole pairs (see [39]). In [40], a model for the dielectric function considering both the excitonic and EHP contributions was applied to describe the experimental data.

Recent experiments [41] have shown the transition from excitonic to plasma emission in the resonance fluorescence from localized excitons in quantum wells. Working with a tunable continuous wave laser with a spectral width much smaller than the exciton binding energy, one is able to detect the dielectric response over the whole spectral range between the 1s exciton and the band edge and open a direct comparison with results of the semiconductor Bloch equations. In a subsequent paper [42], we have extended the concept of self-consistent determination of the self-energies and chemical potentials for the carriers and its inclusion into the semiconductor Bloch equations presented for bulk ZnSe in [30] to quantum wells. There we could give a theoretical explanation of the weak exciton shift observed in the experiments, which changes from higher to lower energies when the temperatures increase from \( T = 5 \) K to \( T = 15 \) K. This corresponds to the results reported in [24] for bulk semiconductors. Moreover, our results agree with terahertz studies [40], where a lowering of the shift of the 1s–2p transition was found for \( T = 6 \) K.
One aim of this paper is to extend our theoretical investigations in [41, 42] to higher densities to describe the Mott transition as an interplay of the broadening and decreasing of the exciton peak and of band edge shrinkage. The second aim is a comparison with a more thermodynamic treatment of the Mott transition, consisting of the description of the chemical equilibrium between electrons, holes and excitons, and of the degree of ionization in a partially ionized EHP. An actual overview of this approach for bulk semiconductors can be found in [30], where the Mott transition was discussed with the possibility of Bose–Einstein condensation of excitons. In particular, results were presented for the carrier self-energies including dynamical screening and compared with the Hartree–Fock (HF) approximation and the results of the CH approximation [6, 7] based on static screening. In this paper, we apply this thermodynamic view of the Mott transition to quantum wells, including the dynamical screening and carrier self-energies within QPA, and discuss the validity of simple approximations based on static screening. Finally, we compare the results of the spectral approach with the semiconductor Bloch equations.

Our paper is organized as follows. In section 2, we extend the description of the Mott transition from the thermodynamic point of view presented in [30] to quantum wells. Investigating the one-particle properties of carriers (self-energies and chemical potentials) and the degree of ionization, one can obtain a simple criterion for the Mott transition. Section 3 starts with a description of the spectral approach based on the semiconductor Bloch equations. We analyze the linear susceptibility as a fingerprint of the composition of the EHP and show that the Mott transition has to be understood within an interplay of the decrease and broadening of the exciton peak and band gap shrinkage. Finally, in section 4 we discuss the connections between both approaches and analyze the limitations of the chemical picture the investigation of the ionization equilibrium is based on.

2. Ionization equilibrium and Mott transition—the thermodynamic approach

From a thermodynamic viewpoint, the partially ionized plasma in semiconductors can be described within a chemical picture as an ionization equilibrium between electrons (e), holes (h) and excitons (X) [43],

\[ e + h ⇄ X. \]  

Applying the concept of extended quasi-particle (QP) approximation [30, 43, 44], the density of free carriers is split into a QP contribution, considering many-body effects between carriers of one species, and a scattering part \( n_{a}^{\text{scatt}} = n_{a}^{\text{QP}} + n_{a}^{\text{scatt}} \) \((a = e, h, n_{h}^{*} = n_{e}^{*})\), which describes the scattering states of interacting electrons and holes. With the density of electron–hole bound states (excitons) \( n_{X} \), the total carrier density is given by \( n_{a} = n_{a}^{*} + n_{X} \), where due to electric neutrality \( n_{e} = n_{h} = n \). In the following, we will neglect the influence of scattering states [44]. Our QP picture is based on the QPA for the retarded one-particle Green’s function [45],

\[
G_{a,k}^{r}(\omega) = \frac{1}{[\omega - \varepsilon_{k}^{a} + i\Gamma_{k}^{a}/2]},
\]

\[
\varepsilon_{k}^{a} = \varepsilon_{k}^{a} + \Delta_{k}^{a,\text{HF}} + \Re \Sigma_{a,k}^{r}(\varepsilon_{k}^{a}),
\]

\[
\Gamma_{k}^{a} = -2 \Im \Sigma_{a,k}^{r}(\varepsilon_{k}^{a}),
\]

with the kinetic energies \( \varepsilon_{k}^{a} = \hbar^{2}k^{2}/2m_{a} \), the Coulomb HF self-energy \( \Delta_{k}^{a,\text{HF}} \) and \( a = e, h \) standing for electrons and holes. Effects of screening the Coulomb interaction are contained...
in the renormalized energies \( \varepsilon^a_k \) and the damping (inverse lifetime) \( \Gamma^a_k \), which are related to the retarded QP self-energy

\[
\Sigma^a_{\alpha,k}(\varepsilon^a_k) = \sum_q \int \frac{d\omega}{2\pi} \frac{[1 - f^a_q(V_{k-q}^{aa,\omega} + f^a_q V_{k-q}^{aa,\omega})]}{\varepsilon^a_k - \varepsilon^a_q - \omega + i\Gamma^a_q/2}.
\] (3)

For the consideration of the confinement of carriers quantum wells by an expansion of Green’s functions with respect to their eigenfunctions in the growth direction, see [46–49]. It leads to an effective two-dimensional (2D) description of carrier motion in the layers. The distance of the lowest eigenenergy to higher sub-bands is, for the considered quantum wells, sufficiently large to restrict the analysis to the consideration of the lowest eigenstate. This simplifies the whole notation, since indices and sums over super-band levels can be omitted. Moreover, one arrives at a quasi-2D description, where the confinement is contained in the quasi-2D Coulomb interaction

\[
\nu_{q}^{ab} = \frac{4\pi}{q} \int dz_e \, dz_h |\Phi_e(z_e)|^2 |\Phi_h(z_h)|^2 e^{q(z_e - z_h)},
\] (4)

containing the eigenfunctions \( \Phi_{a}(z_{a}) \) of carriers in the wells.

A detailed description of the dynamically screened potentials \( V_{k-q}^{aa,\omega} \) is given in [50]. In that paper, the QP energies and damping have been determined with Fermi distributions \( f_{q}^{a,0} \) of ideal (non-interacting) carriers only including their kinetic energy \( \varepsilon^a_q \)

\[
f_k^{a,0} = f_k^{a,0}(\varepsilon_k^a) = \left[ e^{(\varepsilon_k^a - \mu^0_a)/kT} + 1 \right]^{-1}
\] (5)

and the chemical potentials \( \mu^0_a \). This approximate treatment is valid for low excitations, where the influence of many-body effects on the chemical potentials is small. In [30], we have demonstrated for a bulk semiconductor (ZnSe) that there is a pronounced influence of many-body effects on the chemical potentials even at higher excitation around the Mott transition. In this paper, we extend the earlier treatments for quantum wells [28, 50] and include the renormalized carrier energies in the distributions \( f_{q}^{a,0}(\varepsilon_{q}^{a}) \rightarrow f_{q}^{a}(\varepsilon_{q}^{a}) \). The change of chemical potentials \( \mu_{a} \) has to be determined for a given density \( n_{a}^{*} \) and temperature \( T \) of carriers from

\[
n_{a}^{*} = \sum_{k} f_{k}^{a}(\varepsilon_{k}^{a}), \quad f_{k}^{a}(\varepsilon_{k}^{a}) = \left[ e^{(\varepsilon_{k}^{a} - \mu_{a})/kT} + 1 \right]^{-1}
\] (6)

Now equation (6) has to be included in the self-consistent solution of equations (2)–(4). For a fixed density and temperature, we obtain both the QP quantities \( \varepsilon_{k}^{a}, \Gamma_{k}^{a} \) and the chemical potential \( \mu_{a} \).

A simple approximation for the QP energy is the so-called Debye shift [15] or CH self-energy [7] \( \Delta_{a}^{CH} \), which represents the renormalization of the kinetic energy of carriers by static screening of the Coulomb interaction with each other,

\[
\varepsilon_{k}^{a,CH} = \varepsilon_{k}^{a} - \Delta_{a}^{CH}, \quad \Delta_{a}^{CH} = \frac{2}{\alpha_{a}} \left[ 1 - e^{-2\pi\alpha_{a}n_{a}^{2}/kT} \right].
\] (7)

The mass ratio \( \alpha_{e/h} = m_{h/e}/(m_{e} + m_{h}) \) results from the transition to bulk excitonic units (exciton binding energy \( E_{X}^{b} = 4.2 \) meV; excitonic Bohr radius \( a_{X} = 12.4 \) nm). All the results presented in this paper are performed for a 19.8 nm quantum well with an exciton binding energy of \( E_{qw}^{b} = 7.21 \) meV = 1.716 \( E_{X}^{b} \). The mass ratios are \( \alpha_{e} = 0.87, \alpha_{h} = 0.13 \). Within excitonic units, the CH self-energy corresponds to the inverse static screening length \( \Delta_{a}^{CH} = \kappa_{a} \). Replacing (7)
in (6), the corresponding chemical potential is shifted by \( \mu_a^{\text{CH}} = \mu_a^0 - \Delta_a^{\text{CH}} \). A comparison of the CH self-energy with the HF energies \( \Delta_k^{\text{CH}} \) and with the QPA shift, determined by the self-consistent solution of equations (2)–(6), is presented in figure 1. Throughout this paper densities are given in units of cm\(^{-3}\). For that the 2D densities following from (6) are divided by the well width. Comparing first the QPA with the HF approximation, the role of dynamical screening is clearly visible, where the differences become weaker for lower temperatures. For higher temperatures, the CH shift (7) only weakly depends of the mass ratio \( \alpha \) and is of the order of the HF and QP shifts. For \( T = 5 \text{ K} \), the CH shift overestimates the screening by a factor of 3 (note that for \( T = 5 \text{ K} \), \( \Delta^{\text{CH}} / 3 \) is presented in order to make it comparable with the other approximations).

The question now is: how do the chemical potentials change within the different approximations for the QP energies? In bulk semiconductors (3D case), the sum over the wave vector in (6) for ideal particles (5) cannot be performed analytically, and the chemical potentials as a function of carrier density and temperature have to be determined by numerical inversion. In contrast to that, for the quasi-2D description in quantum wells, this inversion can be performed analytically [6, 7]:

\[
n_a^{*,0} = \frac{kT}{2\pi\alpha_a} \ln \left| \frac{e^{\mu_a^0/kT} + 1}{\epsilon_a^0 - \epsilon_a^0} \right|.
\]  

In the left part of figure 2, we present the sum of chemical potentials of both carrier species \( \mu = \mu_e + \mu_h \) following from the inversion of equation (6). A comparison is given for different approximations as considered in figure 1. In the right part, the deviations from the chemical potentials \( \mu_0 \) of ideal carriers are shown. For all temperatures, the many-body effects give rise to a decrease of the chemical potentials. Effects of dynamical screening considered in QPA dominate for higher temperatures in comparison to the HF approximation, while the HF approximation gives the dominant contribution at low temperatures. The opposite holds for the CH approximation. For carrier densities up to \( n = 10^{16} \text{ cm}^{-3} \) which are relevant for the description of the Mott transition (see the next section), a strong decrease is found, in particular, for low temperatures. Additionally, in the right figure, the dashed horizontal line...
Figure 2. Sum of the chemical potentials of electrons and holes for different temperatures and different approximations in dependence of the carrier density (left). Deviation of the chemical potentials from those of non-interacting particles (right).

marks the exciton energy in the non-excited quantum well. As we have seen above, within the CH approximation, $\Delta \mu_{\text{CH}}$ is equal to the CH self-energy. Such a simple picture is not valid for the HF approximation and for the QPA, since the QP energies are not rigid shifts but still depend on the wave vector. Nevertheless, $\Delta \mu$ can be considered for these approximations as an effective shift, too. Having in mind the simple picture, in which the Mott transition is generated by the decrease of the band edge with increasing excitation, while the exciton energy stays nearly unchanged, the cross point of $\Delta \mu$ with the exciton energy can be considered as Mott density. This will be supported by the following investigations of the degree of ionization in the partially ionized plasma, consisting of non-bound carriers and excitons. The excitons are considered as ideal bosons in thermal equilibrium with the distribution

$$f^X(E_K) = \left[e^{(E_K-\mu_X)/kT} + 1\right]^{-1}$$

containing the kinetic energy of the center-of-mass motion $E_K = \alpha_e \alpha_h K^2$ (in excitonic units) and the chemical potential $\mu_X$ of the excitons. As for the ideal carriers (6) and (8), the density $n_X$ of excitons can be calculated analytically:

$$n_X = \sum_K f^X(E_K) = \frac{kT}{\pi \alpha_e \alpha_h} \ln \left| 1 - e^{\mu_X/kT} \right|.$$  

The chemical potentials of carriers and excitons are connected via the binding energy of excitons in the quantum wells $E_{\text{qw}}^X$ by the well-known thermodynamic condition for the chemical (ionization) equilibrium,

$$\mu_X = \mu_e + \mu_h - E_{\text{qw}}^X.$$  

Applying the relations (7) and (8), a Saha equation for the degree of ionization $\alpha$ [15],

$$n_\alpha^* = \alpha n, \quad n_X = (1 - \alpha) n,$$

can be derived, characterizing the composition of the partially ionized plasma:

$$e^{\pi \alpha_e \alpha_h n (1-\alpha)} = \left| 1 - z^\alpha_e z^\alpha_h \right| = 1 - e^{\Delta \mu_{\text{CH}}}$$.  

It is advantageous to introduce here the fugacities \( z_a^0 \)
\[
z_a^0 = e^{\mu_a^0/kT} = e^{2\pi a_n a/kT} - 1, \tag{14}
\]
and the total CH self-energy \( \Delta CH = \Delta e + \Delta h \). Equation (13) can be used to determine the degree of ionization in terms of density and temperature of carriers. For the bulk case, the sums over the wave vectors in (6)–(10) can be performed only numerically. However, an analytical form of the Saha equation can be given if Fermi/Bose distributions of carriers/excitons are replaced by Boltzmann distributions [44]. Doing this for the quasi-2D description in quantum wells, Saha’s equation (13) has the form
\[
1 - \alpha_B = \frac{n\lambda^2}{(\alpha_B)^2} e^{(\mu_X^0 - 2\lambda a_B)/kT}, \quad \lambda^2 = \frac{4\pi}{kT}, \tag{15}
\]
where \( \lambda \) is the thermal wavelength in units of the bulk excitonic Bohr radius. The CH self-energy in the case of Boltzmann distributions is simply \( E_{CH}^B = 2n\lambda^2\alpha_B \).

For simplicity, first results for the degree of ionization by the solution of the Saha equation (13) with the CH self-energy are presented in the left part of figure 3. Regions where most carriers are bound in excitons (\( \alpha \rightarrow 0 \)) are given in blue and regions with high degree of ionization (\( \alpha \rightarrow 1 \)) in red. With increasing temperature, thermal ionization occurs. As a Mott transition the strong increase of \( \alpha \) with increasing carrier density is understood. Additionally, with a blue line the condition \( \Delta \mu_{CH} = \mu_{CH}^0 = \mu_{CH}^0 - \Delta CH = E_{qW}^B \) is plotted which describes those values of temperature and density where the total CH self-energy (corresponding to the sum of the changes of chemical potentials with respect to the chemical potentials of ideal carriers) crosses the exciton binding energy. In particular, at low temperatures, the condition exactly corresponds to those densities where the excitons are ionized and can be used as a criterion for the Mott transition. Within excitonic units, the criterion can be rewritten as \( \kappa a_X = 1 \), and corresponds to the condition \( \kappa a_X = 0.84 \) found in [13] for the vanishing of the bound states in a statically screened potential.

Figure 3. Degree of ionization for the CH approximation according to equation (13) as a function of density and temperature, where the blue line corresponds to \( \Delta \mu_{CH} = E_{qW}^B \) (left). Conditions for the Mott transition with QPA, CH and HF approximation (right).
The abrupt increase of $\alpha$ in the left part of figure 3 for low temperatures can be explained as follows. For low temperatures, the Saha equation (13) has three solutions for $\alpha$ as a function of $n$, corresponding to a classical hysteresis curve with two stable (upper and lower) and one unstable solution [20]. The question arises as to whether this may give rise to a first-order phase transition from an exciton gas to a fully ionized EHP (Mott phase transition) or to the possible existence of an excitonic condensate still at rather high densities. In [30] we have shown that for the bulk case the hysteresis is strongly reduced if the chemical potentials are used within QPA instead of CH approximation. Moreover, it is known for hydrogen plasmas that the atom–atom interaction completely destroys the bistability [31]. The same can be expected by inclusion of the exciton–exciton interaction. This also holds for quantum wells. The reduction of the hysteresis can be seen in the right part of figure 3, where we have used the upper values for $\alpha$ to compare the condition for the Mott transition following from the CH self-energy (blue line, as shown in the left part) with the corresponding conditions for the QPA (black) and for the HF approximation (red). Here $\Delta \mu$ describes the deviations of the respective chemical potentials from those of ideal carriers.

The CH self-energy (static screening) strongly overestimates the screening effects for low temperatures; here the HF approximation is very close to the full QPA. However, for higher temperatures we find better agreement of the CH approximation with the QPA than in the bulk case (see [30]). Actually, the comparison of the CH approximation representing only a rigid shift with both the other approximations is questionable, because the latter ones describeQP energies depending on the wave vector. However, $\Delta \mu$ represents a kind of effective rigid energy shift due to the many-body effects. Finally, the condition $\Delta \mu = -E_{qw}^b$ is a convenient Mott criterion from the thermodynamic point of view. Applying the simple criterion $n^M = 3.3 \times 10^{17} \text{cm}^{-3}$. The thermal ionization corresponding to $kT = E_{qw}^b$ would appear above $T = 20 \text{K}$. Both conditions are only rough estimations.

3. Semiconductor Bloch equations—the spectral approach

Another approach to the description of the Mott transition of excitons in semiconductors more related to experiments is the investigation of the optical response. The dielectric response of a semiconductor to an external light field can be described with the semiconductor Bloch equations. In the linear case, the following equation for the susceptibility in semiconductor quantum wells can be derived [25, 50]:

$$\{\omega - \epsilon_k^{\text{HF}} - \Sigma_k^\text{r}(\omega) + i \gamma_0\} \chi_k(\omega) + \sum_q \left\{N_k v_{k-q}^{\text{eh}} + \Theta_{k,q}(\omega)\right\} \chi_q(\omega) = N_k. \quad (16)$$

Equation (16) represents a generalization of an effective two-particle Schrödinger equation of the electron–hole motion derived first in [5]. It corresponds to the treatment in [47, 51], where the equation was solved in the time domain. We have considered an additional broadening $\gamma_0$ of the excitons which is caused by the roughness of the quantum wells. It was determined by the fit of the excitonic spectrum at low excitation $\gamma_0 = 0.16 \text{meV} = 0.038 E_X^b$. The terms with the carrier distributions in $N_k = 1 - f_k^e - f_k^h$ (Pauli blocking) partially compensate with the sum over the HF contribution of carriers $\epsilon_k^{\text{HF}} = \sum_a (\epsilon_k^a + \Delta_k^{a,\text{HF}})$. Screening effects are contained in the effective interaction $\Theta_{k,q}(\omega)$ and in the retarded interband self-energy $\Sigma_k^r(\omega)$. Both are
Figure 4. Real (left) and imaginary parts of the interband self-energy in excitonic units (17) as a function of the energy $E$ relatively to the band edge $E_g$ for $T = 30 \, \text{K}$ and $n = 10^{15} \, \text{cm}^{-3}$.

connected to each other,

$$
\Sigma^r_k(\omega) = \sum_q \Theta_{q,k}(\omega), \quad \Theta_{k,q}(\omega) = \sum_{a \neq b} \int \frac{d\bar{\omega}}{2\pi} \frac{[1-f^e_k(V^{\text{ab,}+}_{k-q} (\bar{\omega})) + f^h_q V^{\text{ab,-}}_{k-q}(\bar{\omega})]}{\omega - \epsilon^a_k - \epsilon^b_q - \bar{\omega} + i[\Gamma^a_k + \Gamma^b_q]/2}
$$

and compensate likewise. In contrast to the one-particle self-energy (2), the interband self-energy (17) does not describe transitions within the bands, but an interband transition which is initiated by a photon with the energy $\omega$ of the external probe field. The photon generates an electron–hole pair with the energies $\epsilon^a_k + \epsilon^b_q$ accompanied by the absorption/emission of a quantum $\bar{\omega}$ of the elementary excitations in the EHP [25]. Moreover, in contrast to the one-particle description, where we have applied the QPA (2), the interband self-energy is a dynamical quantity, depending on the energy $\omega$. Neglecting the QP damping in the denominator of $\Sigma^r_k(\omega)$ (17) and replacing in the sense of a QP description $\omega \rightarrow \epsilon^a_k + \epsilon^b_q$, the interband self-energy corresponds to the sum of the carrier self-energies $\Sigma^r_{e,k}(\epsilon^e_k) + \Sigma^r_{h,k}(\epsilon^h_k)$ in QPA. Earlier, this treatment was used as dephasing rate approximation for the imaginary part of the self-energy. In [25, 27], we have demonstrated that only the dynamical treatment of the interband quantities provides the correct description of polariton propagation in bulk semiconductors for excitations well below the Mott transition. This will be examined in the following for higher excitations up to the Mott transition for quantum wells. The results of the previous section for the QPA are incorporated here into the QP energy and damping and also into the chemical potentials of the carrier distributions.

In figure 4, we show the real and the imaginary part of the interband self-energy according to (17). The imaginary part, representing the so-called diagonal dephasing $\Gamma_k(\omega) = \text{Im} \, \Sigma^r_k(\omega)$, is peaked at the two-particle dispersion and decreases to lower and higher energies. For $k = 0$, the maximum is at the band edge which is renormalized by the many-body effects (interband self-energy and HF energy). Within the dephasing rate approximation, the imaginary part would be constant over the whole energy range and correspond roughly to the peak value. The energy range of interest for the description of the Mott transition is between the exciton energy and the band edge.

In figure 5, the diagonal dephasing is presented within this energy region for two temperatures and different carrier densities. The vertical dashed lines mark the exciton position $-E^b_{qw}$, and the horizontal line the inhomogeneous (background) damping $\gamma_0$. While the diagonal dephasing is more or less localized around the non-renormalized band edge $E - E_g = 0$ for
Figure 5. Diagonal dephasing at \( k = 0 \) at \( T = 10 \text{K} \) (left) and \( T = 50 \text{K} \) (right) with increasing carrier densities (in \( \text{cm}^{-3} \)) from bottom to top: \( n = 10^{13}, 10^{14}, 2 \times 10^{14}, 5 \times 10^{14}, 10^{15}, 2 \times 10^{15} \) and \( 5 \times 10^{15} \text{cm}^{-3} \). Markers highlight the dephasing at the exciton (▲) and the maximum dephasing (□).

low excitation, it grows upward with increasing excitation due to the many-body effects. This becomes clear considering the increase at the exciton energy \( E_{\text{qw}}^b \) of the non-excited quantum well (triangles in the right part of figure 5 and at the maximum (squares)). For low excitation the maximum is direct at the non-renormalized band edge, while it is shifted to lower energies with increasing excitation. The behavior of the diagonal dephasing and of the renormalization of the interband energy will be compared later with the QP properties (2) and the shift of the exciton which is generated by an interplay of the retarded interband self-energy and the effective interaction (17). For lower temperatures, there is a stronger decrease of the diagonal dephasing towards the exciton. Whereas, at \( T = 50 \text{K} \), the carrier-induced diagonal dephasing for \( n = 5 \times 10^{14} \text{cm}^{-3} \) grows up to the inhomogeneous broadening \( \gamma_0 \), it becomes a multiple of \( \gamma_0 \) for higher densities. This is in contrast to lower temperatures. For \( T = 5 \text{K} \), the carrier-induced diagonal dephasing stays below the inhomogeneous broadening at the exciton. However, we see here the limitation of validity of the QP treatment within the one-particle description for low temperatures and high densities (close to the Mott transition). As shown in figure 2, the total chemical potential of electrons and holes strongly increases for lower temperatures and comes into the order of the exciton binding energy. The imaginary part of the susceptibility should have a transition from gain to absorption at the chemical potentials according to the Kubo–Martin–Schwinger (KMS) relation [52]. This property is not fulfilled within the QP description for the carriers (2) in the interband self-energy and the effective interaction (17). On the other hand, we want to omit a more intuitive manipulation to solve this problem as shown in [7] for the free carrier absorption. The complete dynamical treatment of the one-particle retarded Green’s function and its inclusion into interband self-energy in the semiconductor Bloch equations is a very extensive numerical problem. A principal way to solve the problem was demonstrated in [53] for static screening with a Bethe–Salpeter equation, completely neglecting the dynamical screening. A dynamical treatment for the one-particle properties was presented in [54, 55] and included in the calculation of the retarded interband self-energy \( \Sigma^R_k(\omega) \). However, it is an open question of how to include it in the effective interaction \( \Theta_{q,k}(\omega) \) on the same level in order to get the correct description of the excitonic features below the Mott transition. These features arise with strong compensation of both terms in the two-particle equation (16). For this reason, we have to restrict ourselves to the description
Results for the susceptibility calculated by the solution of the semiconductor Bloch equation (16) including the many-body effects within QPA are given in figure 6 for the imaginary part of the susceptibility $\chi(E) = \sum_k \chi_k(E)$, describing the absorption in the quantum well. With increasing excitation, (i) a decrease of the excitonic absorption peak, (ii) an increase of the linewidth and (iii) a weak shift of the exciton peak to lower energies appear. In [42], we have reported that this shift turns to higher energies, if the temperature is decreased down to $T = 5$ K. It is an advantage of our spectral investigation of the Mott transition in comparison with investigations of the dynamics of the PL in [32–38] that we are able to address single electron–hole pair states in the optical response over the whole spectrum. In order to analyze the spectra in more detail, we have marked the maximum and the half-maximum absorption at the exciton left part, which are considered in figure 7. In the right part of figure 6, the whole spectral range between exciton and the band edge is shown on the level of the band edge absorption. For the lowest density $n = 10^{13}$ cm$^{-3}$, the 2s state of the exciton is still resolved, which disappears with increasing excitation. In any case the band edge is merged with the higher excited exciton states up to the 2s state and cannot be identified separately. For a quantitative description of the behavior of these states near the band edge, additional markers are set in figure 6. The blue crosses in the left part mark the minimum absorption between the exciton and the band edge, and the blue triangles those points where the band edge absorption is decreased to one half of the absorption minimum. Already for densities above $n = 4 \times 10^{15}$ cm$^{-3}$, the band edge and the higher excited exciton states merge with the exciton, giving rise to an increasingly asymmetric look of the exciton line at the bottom. The single markers are summarized in figure 7. In the left part, the normalized peak (left scale) and the FWHM (right) of the excitonic absorption are
With increasing carrier density, the maximum absorption strongly decreases, while the excitonic broadening (FWHM) is increased due to the many-body effects. For low densities, the influence of screening vanishes and the broadening changes into the inhomogeneous broadening \((2\gamma_0 = 0.076E_b^X)\). The vertical dashed lines mark the region where the Mott transition occurs. This is explained in more detail in the discussion of the right part of the figure. The position and the increase of the linewidth of the exciton resonance are marked by the shaded area in the right part. The strong increase of the upper half value of the maximum absorption above \(n = 2 \times 10^{16} \text{ cm}^{-3}\) indicates that the band edge absorption is higher than one half of the peak absorption. The question is now whether this density corresponds to the Mott density where the exciton line is bleached out. To answer this question, one should first have a look at the other curves in the right figure, which compare the band edge with excitonic absorption. The red triangles mark those energies where the level of band edge absorption (including the absorption of the excited exciton states) crosses the excitonic absorption (see the horizontal dashed line in figure 6). Additionally, the curves with blue markers according to figure 6 are given, representing the minimum and one half of minimum and band edge absorption. The latter three curves end in one point at a density of about \(n = 5 \times 10^{15} \text{ cm}^{-3}\), which means that band edge and 1s-exciton merge into each other. Should this density be considered as the Mott density? As seen in the left part of the figure, the peak absorption is reduced nearly to \(1/4\) of the peak absorption at low excitation, and the HWHM is increased to more than sixfold of the inhomogeneous broadening. Finally, from the spectral point of view, the Mott density cannot clearly be determined, but it is for the considered temperature of \(T = 30\) K in a density region between \(n = 6 \times 10^{15}\) and \(2 \times 10^{16} \text{ cm}^{-3}\), marked by both dashed lines in figure 7, the Mott range.

4. Comparison between the two approaches

Finally, we want to compare our results above for the spectral description of the Mott transition with the change of the chemical potentials within QPA (see section 2 and the results in figure 2),
representing a comparison of a two-particle with a one-particle description. In the right part of figure 7, we have added the shift of the chemical potentials in QPA by a dashed line. From the thermodynamic point of view, the Mott transition appears when this line crosses the exciton energy. This is the case for $T = 30\,\text{K}$ above the highest considered density $n = 5 \times 10^{16}\,\text{cm}^{-3}$ in figures 6 and 7 and that at a much higher density than the density range found by the investigation of spectral properties. This is to be expected, since the QPA (2) and the thermodynamic concept of the description of the chemical equilibrium (1) described in section 2 are valid for small damping $\Gamma$ or long life times of the carrier states, which is not valid in the Mott region. This is demonstrated in figure 8.

In our thermodynamic approach presented in the last section, the shift of the chemical potential $\Delta\mu^{\text{QPA}}$ corresponds to an effective shift of the sum of one-particle energies of electrons and holes in QPA (see (2) and figure 2). Within the two-particle (spectral) approach based on the semiconductor Bloch equations (16), the shift of the interband energy $\Delta E_{\text{inter}}(E) = \text{Re}[\Sigma_0(E)] + \Delta_{\text{HF}}$ is a comparable quantity. Since the retarded interband self-energy $\Sigma_k^r(\omega)$ depends on the energy $\omega$ (see the discussion after figure 5), we compare $\Delta E_{\text{inter}}(E)$ at the energy where the diagonal dephasing (see right part of figure 5) becomes a maximum and that at the exciton energy. While the QP-shift $\Delta_0^{\text{QPA}}$ corresponds in a wide density range to the interband shift at maximum dephasing, the shift at the exciton is much lower. However, the interband shift at the exciton is still much larger than the shift of the exciton. This is due to the compensation of the interband energy with the effective interaction and the Pauli blocking in the semiconductor Bloch equations. The problem here is to find a convenient quantity for an effective description of the band edge shrinkage which can explain the vanishing of the exciton line at lower densities than those expected by the discussed interband and QP shifts (see figure 7). Finally, the shrinking gap is described by an interplay of the interband self-energy with the effective interaction and of the HF renormalization with the Pauli blocking which cannot be expressed by one compact quantity.

Figure 8. Comparison of energy shifts $\Delta\mu^{\text{QPA}}$ and damping $\Gamma^{\text{QPA}}$ within QPA (red curves) with corresponding spectral properties (black curves) for $T = 50\,\text{K}$. Left: shift of the exciton energy $\Delta E_X$, energy $E@\text{Max}[\Gamma]$, where the diagonal dephasing becomes a maximum (compare with figure 5), and shift of interband energy at the exciton $\Delta E_{\text{inter}}(E_X)$. Right: carrier-induced broadening (HWHM) of the exciton, maximum diagonal dephasing $\text{Max}[\Gamma(E)]$ (see figure 5) and diagonal dephasing at the exciton $\Gamma(E_X)$.
A similar behavior is found for the broadening (HWHM) of the exciton presented in the right part of figure 8. Due to the compensation of the diagonal dephasing with the imaginary part of the effective interaction, the half-width of the exciton (HWHM) is much smaller than the dephasing at the exciton ($\Gamma(E_X)$). Only for higher densities, this compensation is weakened and the excitonic linewidth strongly increases as we have already discussed in the description of the Mott transition. Furthermore, we find that the QP damping $\Gamma_{\text{QPA}} = \Gamma_0' + \Gamma_0^b$ (see (2)) is much larger than the maximum diagonal dephasing $\text{Max}[\Gamma_0(E)]$ at the two-particle dispersion. Considering that the QP damping corresponds to the inverse life time of carrier states $T_1^a$ and the inverse diagonal dephasing to the dephasing time $T_2$, this contradicts the simple relation $1/T_1^e + 1/T_1^b = 1/T_2$ used in the description of two-level systems (see [7]).

5. Summary

We have studied the Mott transition of excitons in GaAs–GaAlAs quantum wells. Many-body effects were taken into account including the dynamical screening of the Coulomb interaction both in the one-particle properties of the carriers and in the two-particle properties of electron–hole pairs. The Mott transition is described within two approaches. The first one is a thermodynamic approach, based on the self-consistent calculation of self-energies and chemical potentials of carriers in the framework of the chemical equilibrium between unbound carriers and excitons. The second one is a spectral approach focused on the electron–hole pair properties which can be seen experimentally in the dielectric response and which is calculated here with the semiconductor Bloch equations. A comparison of the dynamically screened self-energy in QPA with simple approximations shows that the HF approximation is convenient for low temperatures ($T < 10$ K), where the CH approximation (static screening) drastically overestimates the effect of screening. This is contrary to the case of higher temperatures ($T > 50$ K), where the HF approximation fails and static screening becomes a more convenient approximation. Studying the degree of ionization, we find that the Mott density $n_{\text{Mott}}$ is reached when the change of the chemical potential due to many-body effects crosses the exciton binding energy $\Delta\mu = -E_{b\text{qw}}$. This corresponds to the general view of the Mott transition, to be generated by the decreasing band edge with increasing density, while the exciton energy stays nearly unchanged. The changes of the chemical potential play here the role of an effective rigid shift of the band edge.

In contrast, it is difficult to identify the band edge in the region of the Mott transition within the spectral approach. For low excitation, the 1s-exciton can be clearly resolved. However, the band edge absorption cannot be separated from the absorption of the higher excited excitonic bound states. With increasing excitation, the band edge first bleaches the 2s-state and then immerses into the upper exciton tail. Further increase of the excitation leads to an increasing broadening and a decreasing peak of the 1s-exciton absorption, while the band edge cannot be identified any more. Thus, the Mott transition does not occur as an abrupt jump at a certain density, but is rather a smooth transition in a broader density region. This density region lies considerably below the predicted Mott density in the thermodynamic approach.

Our analysis shows that the band edge shift cannot be adequately described within the QPA for the one-particle self-energies. Rather the interband self-energy and the diagonal dephasing (i) strongly change over the spectral range between the exciton and the band edge and (ii) cannot be described as the sum of the one-particle self-energies calculated with QPA.
Moreover, the electron–hole pair spectrum is generated by the interplay of the interband self-energy and the effective interaction which widely cancel each other. The cancelation of the real part of both effects results in a nearly unchanged exciton position which undergoes only a weak shift depending on the temperature. For the imaginary parts it is reflected by the much lower excitonic broadening in comparison to the diagonal dephasing (imaginary part of the interband self-energy).

Finally, the large increase of the one-particle damping in the vicinity of the Mott transition shows the limit of validity of the QP picture both in the calculation of the chemical potentials and in its use for the derivation of the mass-action law within the thermodynamic approach. Therefore, for the considered system of quantum wells the spectral approach should be preferred for a consistent picture of the Mott transition.

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