Mott transition versus Bose-Einstein condensation of excitons

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Abstract. We investigate the influence of many-body effects on both the chemical potentials of carriers, and the Mott transition of para-excitons in Cu$_2$O over a wide range of temperatures and carrier densities, in order to determine the region where an excitonic fraction can exist. In contrast to simplified approximations used in the literature we consider full dynamical screening between carriers and find out that (i) the chemical potentials are much less decreased by the many-body effects, and (ii) for low temperatures the density, where the Mott transition appears, is one order of magnitude higher. This leads to an extension of the region of existence of excitons and, therefore, of a possible BEC, to higher temperatures.

1. Introduction
Collective quantum phenomena have been of great interest in semiconductor physics for many years. Following the theory of the ideal Bose gas, the Bose-Einstein condensation (BEC) of excitons should occur, when their chemical potential reaches the exciton resonance by reduction of the temperature or increase of their density (for an overview see [1, 2, 3]). On the other hand, the existence of an excitonic fraction in a semiconductor is limited due to the Mott transition of excitons, induced by many-body effects between excitons and their constituents (electrons and holes). In particular, the weakening of the Coulomb interaction due to screening leads to a breakup of the excitonic binding with increasing carrier density (density ionization). Considering the chemical equilibrium of unbound and bound electron-hole pairs (excitons), the degree of ionization can be used to describe the composition of the partially ionized electron-hole plasma (EHP) in an excited semiconductor [4, 5].

The aim of our paper is the investigation of the influence of many-body effects on the chemical potentials of electrons, holes, and para-excitons in Cu$_2$O, which have a long lifetime [6]. We determine the composition of the partially ionized electron-hole plasma in dependence on the temperature and the density of carriers. In comparison to recent treatments [7, 8] we go beyond simple approximations for the many-body effects (static screening) or interpolation formulas for the chemical potentials valid for a wide range of temperatures and densities (for a review see [4, 5]). Our approach is based on a self-consistent calculation of one-particle carrier selfenergies in quasi-particle approximation including the dynamical screening of the Coulomb interaction. In a series of papers we have demonstrated that this approach is convenient to describe the details of the excitonic absorption at lower excitation, showing up in phase-resolved interferometric measurements [10, 11]. In this paper we extend our treatment to higher densities and lower
temperatures in order to describe the Mott transition and to reach the region where BEC can be expected.

2. Quasi-particle approximation and chemical potentials of carriers

The influence of many-body effects between carriers in the EHP of an excited semiconductor can be described by their retarded selfenergies \( \Sigma_a \), with \( a = e, h \) denoting electrons, holes. Within quasi-particle approximation their real part describes the renormalization of the energy dispersion \( \varepsilon \) and the imaginary part the damping \( \Gamma \) (inverse life time) of carrier states

\[
\varepsilon_k^a = \frac{h^2 k^2}{2m_a} - \Sigma_{a,k}^{HF} - \text{Re} \Sigma_{a,k}^{sc}(\varepsilon_k^a), \quad \Gamma_k^a = -2 \text{Im} \Sigma_{a,k}^{sc}(\varepsilon_k^a). \tag{1}
\]

We use the selfenergy in random phase approximation (RPA), which can be split into a Hartree-Fock contribution \( \Sigma_{a,k}^{HF} \) and a second term \( \Sigma_{a,k}^{sc} \) which considers the dynamical screening between carriers

\[
\Sigma_{a,k}^{HF} = \sum_q f^a(\varepsilon_q) v_{k-q}, \quad \Sigma_{a,k}^{sc}(\omega) = \sum_q \int \frac{d\omega'}{2\pi} \frac{1 - f^a(\varepsilon_q') + n^B(\omega')}{\omega - \varepsilon_q - \omega' + i\Gamma_k^a/2} \tilde{V}_{k-q}(\omega'). \tag{2}
\]

The Fermi distribution \( f^a(\varepsilon_q) \) contains the renormalized dispersion and the chemical potential \( \mu_a \) of carriers being related to the carrier density \( n_k^a \) by

\[
n_k^a = \sum_q f^a(\varepsilon_q), \quad f^a(\varepsilon_q) = \left[ e^{(\varepsilon_q - \mu_a)/kT} + 1 \right]^{-1}. \tag{3}
\]

\( n^B(\omega') \) is a Bose distribution of the elementary excitations in the EHP (plasmons), which are determined by the dynamical screened potential \( \tilde{V} \). For further details we refer to [10, 11]. In order to determine the quasi-particle energy and damping for a given carrier density and temperature, the Eqns. (1)-(3) have to be solved self-consistently, including the chemical potential. We start our numerical procedure with the calculation of the chemical potential \( \mu_0 \)

![Figure 1. Sum of the chemical potentials of electrons and holes in dependence of the density of carriers within QPA as solution of Eqns. (1)-(3), left: comparison of QPA for different temperatures, right: comparison of the QPA (solid,bold) with that of free carriers (solid,thin), and with different approximations for the selfenergy: only Hartree-Fock selfenergy (dash-dotted), Debye shift (dotted), and with the Padé formula of [13] (dashed).](image)

of free (non-correlated) carriers, considering only the kinetic energy part in (1) for the iteration of (3). This is used to iterate the Eqns. (1),(2) self-consistently. As a result we get the quasi-particle energy and damping which than are employed in the calculation of the carrier density of the correlated EHP with (3). In earlier papers the quasi-particle energy was approximated by the so-called Debye shift [4] or Coulomb-hole selfenergy [12], given in excitonic units by \( \Sigma^{CH} = -\kappa a_{ex} E_{ex}^0 (\kappa - \text{inverse screening length}) \). We use the following parameters for Cu$_2$O [6]:
binding energy $E^b_{ex} = 150 \text{meV}$, Bohr radius $a_{ex} = 0.65 \text{nm}$. In [13] a Padé formula for the sum of the chemical potentials was presented for a wide range of temperatures and densities. The formula was constructed with some data points $(n, T)$ of the numerical iteration of Equs. (1)-(3) and known analytical formulas for limiting cases. However, the damping was neglected $(\Gamma \rightarrow 0)$ in the numerical iteration. Our calculations show that this leads to exaggerated quasi-particle energies. In [10] we have shown, that the consideration of the quasi-particle damping is necessary for a correct description of the excitonic lineshape, too.

Results of our numerical iteration, as described above, for different temperatures and a comparison of our results with simplified earlier treatments are given in Fig. 1 for the sum $\mu = \mu_e + \mu_h$ of the chemical potentials of electrons and holes in units of the thermal energy $kT$. The strong influence of many-body effects becomes obvious. Comparing the behavior of the chemical potential in QPA (left part), the largest deviation from that for ideal particles is found for low temperatures. A comparison of our approach with different approximations is presented in the right part for a temperature of $T = 33K$. As one can see, the CH shift (dashed line) overestimates the influence of screening considerably. The deviation becomes larger for lower temperatures. Only for temperatures where the thermal energy $kT$ is of the order of the excitonic binding energy $(kT = E^b_{ex} = 150 \text{meV} \text{ for } T = 1740K)$ the Debye approximation (static screening) comes closer to the QPA (dynamic screening) presented above. The deviation of the Padé formula from [13], which has been used in a previous paper [8] is still remarkable. As we have pointed out, this is due to the neglected quasi-particle damping in (2). In contrast we find that the Hartree-Fock approximation $(\Sigma^{sc} = 0)$ comes closer to the full calculations at higher carrier densities, even for higher temperatures.

3. Composition of the partially ionized electron-hole plasma

Depending on the carrier density and temperature, electrons and holes in an excited semiconductor can bind to excitons, and the total density of carriers $n_a$ at quasi-equilibrium is split into a part of free (correlated, but non-bound) carriers $n_a^*$ and a part bound to excitons $n_X$, where due to charge neutrality $n_e = n_h$ is valid. The EHP can be considered to be in a chemical (ionization) equilibrium $e + h \rightleftharpoons X$, and the composition can be characterized by the degree of ionization

$$\alpha = \frac{n_a^*}{n_a}, \quad n_a = n_a^* + n_X. \quad (4)$$

The chemical potential of excitons follows from the well-known thermodynamic condition for the chemical equilibrium [4]

$$\mu_X = \mu_e + \mu_h - E^b_{ex}, \quad n_X(\mu_X, T) = \sum_k \left[ \frac{e^{(\frac{K^2}{2M_k} - \mu_X)/kT} - 1}{e^{(\frac{K^2}{2M_k})/kT} - 1} \right]^{-1} \quad (5)$$

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2}
\caption{Degree of ionization $\alpha$ vs. density, Eq. (4). Left: for different temperatures $T = 10K$ (solid), $T = 33K$ (dashed), and $T = 100K$ (dotted), Right: comparing different approximations for the chemical potential: our treatment (solid), CH shift (dotted), Padé formula [13] (dashed).}
\end{figure}
Here the excitons are considered as ideal bosons with the density-dependent binding energy $E_b^x$ and the scattering states in the excitonic spectrum are neglected. For further details we refer to [8, 9].

The degree of ionization, calculated with our chemical potentials (last section) is presented in the left part of Fig. 2. At lower densities most e-h pairs are bound to excitons ($\alpha \ll 1$), while a breaking of the excitonic binding appears very abruptly at the Mott density. A comparison with the CH shift (static screening) in the right part of the figure shows, that (i) the Mott density is increased by nearly one order of magnitude, and (ii) the region of multivaluedness (shaded area) where a phase transition may be expected \[4\], is reduced considerably. An overview for the degree of ionization in a wide range of temperatures and densities is presented in Fig. 3. Due to the increased Mott density at low temperatures ($n^{\text{Mott}} = 3 \times 10^{18} \text{ cm}^{-3}$) the region where BEC of ideal bosons can be expected is shifted to higher temperatures (see peak of the dashed triangle in Fig. 3).

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**Figure 3.** Contour plot of the degree of ionization in the density-temperature plane for Cu$_2$O. Excitons are dominant in the low density and low temperature region, where $\alpha \ll 1$. The dashed triangle at the bottom borders the region where BEC of ideal excitons is possible: $(1 - \alpha)n_e A_X^3 \geq 2.612$ \[3\].