Journal of Luminescence 129 (2009) 1478–1482

Contents lists available at ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

Phase diagram of electron–hole systems: Interplay between exciton Mott transition and quantum pair condensation

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ARTICLE INFO

Available online 12 May 2009

PACS:
71.10.Fd
71.30.+h
71.35.Cc

Keywords:
Electron–hole systems
Exciton Mott transition
Quantum pair condensation
Electron–hole Hubbard model
Coulomb correlation
Dynamical mean-field theory

ABSTRACT

Quasi-thermal-equilibrium states of electron–hole (e–h) systems in photoexcited insulators are studied from a theoretical viewpoint, stressing the exciton Bose–Einstein condensation (BEC), the e–h BCS-type pair-condensed state, and the exciton Mott transition between an insulating exciton/biexciton gas phase and a metallic e–h plasma phase. We determine the quasi-equilibrium phase diagram of the e–h system at zero and finite temperatures with applying the dynamical mean-field theory (DMFT) to the e–h Hubbard model with both repulsive and attractive on-site interactions. Effects of inter-site interactions on the exciton Mott transition are also clarified with applying the extended DMFT to the extended e–h Hubbard model.

1. Introduction

The electron–hole (e–h) system is composed of two oppositely charged fermions, electrons in a conduction band and holes in a valence band, whose densities are assumed to be identical here. Such e–h systems realized in photoexcited insulators have been intensively studied not only because the e–h systems govern linear and nonlinear optical properties of matters [1] but also because various quantum phenomena are expected [2] to take place depending on particle density, interparticle interaction strength, temperature, and dimensionality. In usual materials, the intraband relaxation is much faster than the interband one. In a time scale after the intraband relaxation and before the interband relaxation time, therefore, the system gets settled in a quasi-thermal-equilibrium state. In this paper, we confine ourselves to such quasi-equilibrium situation. Dynamical features of carrier relaxation processes in e–h systems are not treated in this paper.

We focus on the quantum cooperative phenomena, e.g., the phase transitions and the quantum condensation in the e–h systems with varying interaction strength, temperature, and particle density. We shall guess intuitively what happens in a quasi-equilibrium e–h system as the particle density increases. In the low-density limit where only one electron and one hole are excited, an "exciton" may be formed as a bosonic bound state of an electron and a hole [3]. When two electrons and two holes are excited, a "biexciton" (an excitonic molecule) is a possible bound state (also bosonic). This biexcitonic state reflects characteristics of the exciton–exciton interaction [4]. In the case that many excitons are excited, dissociation of many excitons into a gas state of electrons and holes (called the "e–h plasma") may be possible. This is called the "exciton Mott transition." The main origins of this Mott transition are the Pauli blocking and enhancement of the Coulomb screening.

To describe the exciton Mott transition, many-body Coulomb correlation effects should be taken properly into account continuously from weak to strong coupling regimes: from the exciton gas (strong-coupling regime) to the e–h plasma (weak-coupling regime). The dynamical mean-field theory (DMFT) [5] is a powerful theoretical method to cover both regimes. In this paper, we introduce this method applied to the issues of the exciton Mott transition in three-dimensional e–h systems.

Assuming the quasi-thermal-equilibrium at very low temperature, macroscopic quantum phenomena, that is, e–h pair condensations are also expected, e.g., the Bose–Einstein condensation (BEC) of excitons and the e–h superconductor-like state (e–h BCS state). At relatively low e–h particle density (strong-coupling regime), strongly bound e–h pairs undergo the BEC as an exciton gas. On the other hand, at high e–h density (weak-coupling regime) where the mean interparticle distance is shorter than the

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doi:10.1016/j.jlumin.2009.04.079

exciton Bohr radius, weakly bound e–h pairs may behave like the Cooper pairs in conventional superconductors at sufficiently low temperatures, that is, the Bardeen–Cooper–Schrieffer (BCS) state of e–h pairs. We shall discuss these exciton BEC and e–h BCS states and its crossover with the use of the self-consistent t-matrix approximation in the DMFT framework.

A main aim of this paper is to discuss the quasi-equilibrium phase diagram of the e–h systems as a function of interaction strengths and temperature. To this end, for simplicity, we employ the e–h Hubbard model,

$$\hat{H} = -\sum_{\langle ij\rangle,\sigma}\sum_{\nu=e,h} t_\nu \hat{a}^{\nu\dagger}_{i\sigma}\hat{a}^{\nu}_{j\sigma} - \sum_{j\sigma,\nu}\mu_\nu \hat{n}^{\nu}_{j\sigma} + U\sum_{j,\nu}\hat{n}^{\nu}_{j\uparrow}\hat{n}^{\nu}_{j\downarrow} - U'\sum_{j\sigma\sigma'}\hat{n}^{e}_{j\sigma}\hat{n}^{h}_{j\sigma'}, \quad (1)$$

where $\hat{a}^{e\dagger}_{j\sigma}$ ($\hat{a}^{h\dagger}_{j\sigma}$) denotes a creation operator of an electron (a hole) with spin $\sigma = \{\uparrow,\downarrow\}$ at the jth site and $\hat{n}^{\nu}_{j\sigma} = \hat{a}^{\nu\dagger}_{j\sigma}\hat{a}^{\nu}_{j\sigma}$ with $\nu = \{e,h\}$. The angle bracket $\langle ij\rangle$ stands for the sum of adjacent sites. The quantities t_e (t_h) and μ_e (μ_h) are the transfer integral of the electrons (holes) between the nearest-neighbor sites and the chemical potential measured from the center of the bare electron (hole) band, respectively. The on-site Coulomb interaction of the e–e (h–h) repulsion and that of the e–h attraction are expressed by U and $-U'$, respectively. Here we suppose that conduction electrons and valence holes, whose bands are isotropic, have infinite lifetime. Hereafter we use the bare DOS $\rho^0_\nu(\varepsilon) = \sqrt{4t_\nu^2 - \varepsilon^2}/(2\pi t_\nu^2)$. In this lattice model (1), the interaction strengths, U and U', are treated as parameters, which are independent of the particle density (the filling factor) $n \equiv n^e = n^h$. Effects of the inter-site interactions will be discussed later.

2. Exciton Mott transition

Here we first examine the exciton Mott transition. In this section, we focus on the normal phase where the quantum condensation of e–h pairs is not allowed. We shall employ DMFT, which requires only the locality of the self-energy, and can take full account of local correlations. This locality and the resulting DMFT become exact in the limit of infinite spatial dimensions and good approximation of the three-dimensional systems. Quantum orderings in one-dimensional e–h systems are investigated in detail in Refs. [6–9].

Within DMFT, the many-body problem of the lattice fermion model, i.e., the e–h Hubbard model (1) is mapped onto the problem of a single-site impurity embedded in an effective medium. The effective medium, which is dynamical and is represented by the noninteracting impurity Green function $\mathscr{G}^\nu_0(\omega)$ of an effective single-impurity Anderson model (SIAM), is determined from the self-consistency condition $\mathscr{G}^\nu_0(\omega)^{-1} = \omega + \mu_\nu - t_\nu^2 G_\nu(\omega)$, where $G_\nu(\omega)$ is the local Green function for electrons or holes of the model. The condition is read as $G^\nu_{\mathrm{imp}}(\omega) = G_\nu(\omega)$. The interacting impurity Green function of the effective SIAM, $G^\nu_{\mathrm{imp}}(\omega)$, should be calculated exactly such that effects of the interactions on the impurity site are fully included. Contrary to the ordinary mean-field approaches, thus, in the DMFT scheme the local correlations and dynamical quantum fluctuations are taken into full account [10].

2.1. Phase diagram at zero temperature

We shall discuss the case of arbitrary filling (not half filling). Results in the case of half filling are given in Ref. [10]. For the e–h particle density $n \neq 1$, the process for determining μ_ν is added to the self-consistency cycle. We carried out the exact diagonalization calculation to solve the SIAM. Here $t_h/t_e = 1$ is fixed. Fig. 1 shows the phase diagram for $n = 0.25$ ($\frac{1}{8}$-filling) at zero temperature. We find that there are mainly three typical regions in the phase diagram. In the case of weak attractive interaction, the system is metallic, corresponding to the e–h plasma phase. When U and U' increase, the system becomes insulating. A remarkable feature is that there are two types of insulating states: the "exciton-like insulator phase" (exciton phase) for strong repulsion (large U and $U \geq U'$) and the "biexciton-like insulator phase" (biexciton phase) for strong attraction (large U' and $U' \geq U$). The former exciton phase appears only in the not-half-filling case.

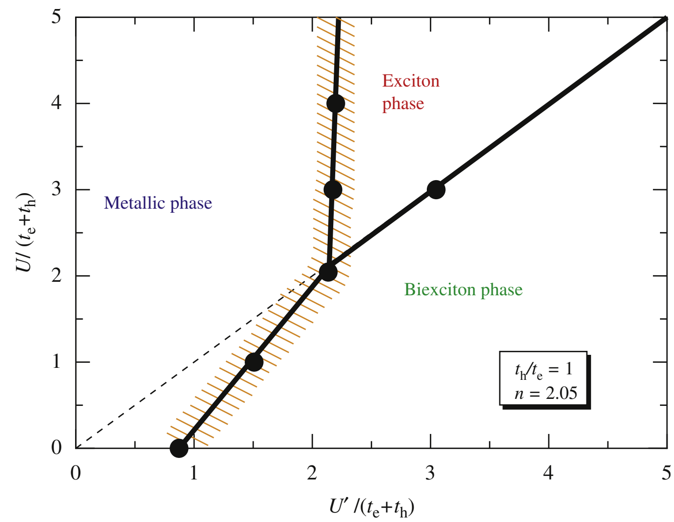

Fig. 1. Phase diagram for the exciton Mott transition in the (U', U) plane at $n = 0.25$ at zero temperature for $t_h/t_e = 1$. The shaded area is the coexistent region of the metallic and insulating phases reflecting the first-order transition.

In the metallic phase, the quasi-particle weight Z_ν has a finite value and there is finite DOS at the Fermi level (the quasi-particle coherent peak), i.e., the interacting DOS $\rho_\nu(0) \neq 0$. On the other hand, in the biexciton-like insulator phase, $Z_\nu = 0$ and the coherent peak of the DOS disappears. The exciton-like insulator is characterized by that $Z_\nu \neq 0$ but $\rho_\nu(0) = 0$. In addition, this metal–insulator phase transitions are the first-order; coexisting regions of several phases exist along the phase boundaries.

Appearance of the "exciton-like insulator phase" is understood by considering the limit of $U \to \infty$. In this limit, the model (1) can be mapped onto a single-band attractive Hubbard model with the attraction $-U'$:

$$\hat{H} = -t\sum_{\langle ij\rangle}\sum_{\nu=e,h}\hat{a}^{\nu\dagger}_i\hat{a}^{\nu}_j - U'\sum_j \hat{n}^e_j\hat{n}^h_j. \quad (2)$$

According to the results of DMFT study of this model [11,12], a paring state appears in addition to the metallic state. This paring state corresponds to the exciton-like insulator phase in our model, in which incoherent local e–h pairs (do not condense) are formed. Optical absorption spectra also show characteristic features of the exciton-like insulator phase [13].

To confirm the phase diagram (Fig. 1), we apply also the slave-boson mean-field approximation to the e–h Hubbard Hamiltonian (1). Details are given in Ref. [14]. We here show only results. The ground state of the e–h Hubbard model is characterized by three different phases: (i) The "e–h plasma" phase (metallic); all of the probability amplitudes of the slave bosons are finite. This phase is a normal Fermi liquid. (ii) The "exciton gas (X gas)" phase (exciton-like insulator); only the amplitudes for the empty and excitonic configurations are finite. (iii) The "biexciton gas