Dynamical mean-field theory for the exciton Mott transition in electron-hole systems

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Abstract. In electron-hole (e-h) systems in photoexcited insulators, the exciton Mott transition may take place, which is a typical photoinduced phase transition. To understand how the exciton Mott transition depends on the e-h carrier density and the Coulomb correlation, we study an e-h two-band Hubbard model by means of the dynamical mean-field theory assuming that electron-hole pairs do not condense. The phase diagram on the plane of interactions at zero temperature is obtained. When both electron and hole bands are half-filled, two types of insulating states appear: the Mott-Hubbard insulator and the biexciton-like insulator. Away from half-filling we find another insulating phase, the exciton-like insulator, which is followed by the formation of incoherent (not condensed) e-h pairs, while the Mott-Hubbard insulator phase disappears. The exciton Mott transition is found to be the first-order transition. Linear optical susceptibilities are also discussed.

1. Introduction

Systems that show drastic changes in chromic, magnetic, dielectric, and transport properties triggered by external light stimulation [1, 2] are an attractive target for fundamental physics and materials science [3, 4]. One important strategy for developing highly responsive photoexcited materials is to identify the photo-control of cooperative phenomena. This exotic photo-effect is called a photoinduced phase transition (PIPT). There are two types of PIPT processes: (i) photoinduced conversion from a “ground state” to new “ground state” via optically-excited states, and (ii) photoinduced creation of new material phases in an optically-excited state within a lifetime of decay. The former (i) has been extensively studied in terms of the photoinduced domino mechanism [5] and the photoinduced domain injection processes [6, 7]. We shall treat the latter (ii) in this paper.

Photoirradiation to an insulator induces the interband transition to form an electron-hole (e-h) system in a two-band system. The e-h system has the following characteristic features: (a) There are two types of fermions, which have opposite charge and different mass, (b) the Coulomb interactions of both repulsion and attraction among the fermions exist, and (c) depending on e-h densities, the range and energies of the interactions are changed by the screening. In such e-h systems, metal-insulator transitions take place and have attracted interest for many years: the exciton Mott transition (EMT) between an exciton/biexciton gas phase and an e-h plasma phase, and crossover between the Bose-Einstein condensation (BEC) of excitons at low density and the BCS-like condensation of e-h pairs at high density. We pay attention to the EMT as one of the most striking PIPT phenomena in an optically-excited state, which may depend on the excited particle density and (quasi)temperature. In this paper, we confine ourselves to the
case where the condensation of e-h pairs is not allowed, and thus the BCS-BEC crossover is left for other papers.

Since the screening mechanism plays an important role in emergence of the EMT, it may depends also on the dimensionality of the e-h system. To this end, we have investigated one-dimensional (1D) e-h systems in a high-density regime by means of bozonization techniques [8]. In the 1D case with the long-range Coulomb interactions, the system is always insulating even at the high density limit and that the EMT never occurs at zero temperature. A main aim of this paper is to clarify the EMT in higher-dimensional cases. We assume a quasiequilibrium neglecting the interband recombination processes (i.e., a radiative lifetime is much longer than an intraband thermalization time) to examine the EMT with the simplest model, the two-band Hubbard model at zero temperature, by using the dynamical mean-field theory (DMFT) [9]. The DMFT 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.

2. Dynamical mean-field theory

We consider an e-h system described by the two-band Hubbard model:

\[ H = - \sum_{<ij>,\sigma} t_{ij} d_{i\sigma}^\dagger d_{j\sigma} - \sum_{i,\sigma} \mu_{\sigma} d_{i\sigma}^\dagger d_{i\sigma} + U \sum_{i,\sigma} d_{i\sigma}^\dagger d_{i\sigma}^\dagger d_{i\sigma} d_{i\sigma} - U' \sum_{i\sigma\sigma'} d_{i\sigma}^\dagger d_{i\sigma'}^\dagger d_{i\sigma} d_{i\sigma'}, \]

where \( d_{i\sigma}^\dagger \) (\( d_{i\sigma}^\dagger \)) denotes a creation (annihilation) operator of an electron (a hole) with spin \( \sigma \) = \{↑, ↓\} at the \( i \)th site. The quantities \( t_{ij} \) (\( h_{ij} \)) and \( \mu_{\sigma} \) (\( \mu_{\sigma} \)) are the transfer integral of the electrons (holes) between the neighboring sites and the chemical potential measured from the center of the 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. The local Green function for electrons or holes of the model (1) is defined by

\[ G_\alpha^{\sigma}(\omega) = \int_{-\infty}^{\infty} \frac{d\varepsilon}{\omega + \mu_\alpha - \varepsilon - \Sigma_\alpha^{\sigma}(\omega)}, \]

where \( \Sigma_\alpha^{\sigma}(\omega) \) is the self-energy of electrons (\( \alpha = e \)) or holes (\( \alpha = h \)), which is local, i.e., does not depend on the wave number, in the limit of infinite-dimensions. We use the semicircular density of states (DOS), \( \rho_0^{\sigma}(\varepsilon) = \sqrt{\varepsilon^2 - \varepsilon_0^2}/(2\pi\varepsilon_0^2) \), which corresponds to the Bethe lattice in the limit of the infinite connectivity.

With the use of the DMFT, the many-body problem of the lattice 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 \( G_0^{\alpha}(\omega) \) of an effective single-impurity Anderson model (SIAM), is determined from the self-consistency condition \( G_0^{\alpha}(\omega)^{-1} = \omega + \mu_\alpha - t_0^2 G^{\alpha}(\omega) \). The condition is also read as \( G_{\text{imp}}^{\alpha}(\omega) = G^{\alpha}(\omega) \). The interacting impurity Green function of the effective SIAM, \( G_{\text{imp}}^{\alpha}(\omega) \), should be calculated exactly such that effects of the interactions on the impurity site are fully included.

First we employ the two-site DMFT, which is a simplified version of the DMFT. In the two-site DMFT, the effective medium \( G_0^{\alpha}(\omega) \) is represented simply by a few parameters, i.e., the effective SIAM consists of a single impurity and only a single bath sites. For the model (1), the corresponding effective two-site SIAM is

\[ H_{\text{imp}} = \sum_{\sigma,\alpha} \left[ \varepsilon_c^\sigma \epsilon_c^\alpha \epsilon_c^\alpha + V_\alpha \left( d_{\sigma}^\dagger e^\dagger_{\sigma'} + \text{h.c.} \right) - \mu_\alpha d_{\sigma}^\dagger d_{\sigma} \right] + U \sum_{\alpha} d_{\sigma}^\dagger d_{\sigma}^\dagger d_{\sigma} d_{\sigma} - U' \sum_{\sigma\sigma'} d_{\sigma}^\dagger d_{\sigma'}^\dagger d_{\sigma} d_{\sigma'}, \]

where the bath parameters \( V_\alpha \) and \( \varepsilon_c^\sigma \) denote the hybridization between the impurity (\( d \)) and bath (\( c \)) sites, and the energy level of the bath site, respectively. The Green function of the effective
medium becomes $G^0_\alpha(\omega)^{-1} = \omega + \mu_\alpha - V^2_\alpha/(\omega - \varepsilon^0_\alpha)$, which is the noninteracting impurity Green function. The self-energy is expanded in the low-energy region, $\Sigma(\omega) \sim a_\alpha + b_\alpha\omega$, and then the resulting local Green function (2) and impurity Green function $G^\alpha_{\text{imp}}(\omega)^{-1} = G^0_\alpha(\omega)^{-1} - \Sigma(\omega)$ should coincide with each other at high-energy region. Thereby, the self-consistency equation for $V_\alpha$ is $V^2_\alpha = t^2_\alpha Z_\alpha$, where $Z_\alpha = (1 - b_\alpha)^{-1} = [1 - d\Sigma(\omega)/d\omega|_{\omega=0}]^{-1}$, is the quasiparticle weight. The particle densities of the original and impurity models should be equal, i.e., $n^\alpha = n^\alpha_{\text{imp}}$, leading to the self-consistency condition for $\varepsilon^\alpha_c$.

$$
\int_{-\infty}^{0} d\omega \operatorname{Im} G^\alpha(\omega + i0^+) = \int_{-\infty}^{0} d\omega \operatorname{Im} G^\alpha_{\text{imp}}(\omega + i0^+).
$$

Consequently, the model (1) can be solved within the two-site DMFT by the following self-consistency cycle [10]; (i) $G^\alpha_{\text{imp}}(\omega)$ is directly calculated by the exact diagonalization of the two-site SIAM (3) with $\varepsilon^\alpha_c$ and $V_\alpha$. (ii) By using $\Sigma(\omega) = G^0_\alpha(\omega)^{-1} - G^\alpha_{\text{imp}}(\omega)^{-1}$, a new value of $V_\alpha$ is determined from the conditions $V^2_\alpha = t^2_\alpha Z_\alpha$ and $Z_\alpha = (1 - b_\alpha)^{-1}$. (iii) By substituting $\Sigma(\omega)$ for (2), a new value of $\varepsilon^\alpha_c$ is chosen so as to satisfy the condition (4). This process (i)–(iii) is iterated until $\varepsilon^\alpha_c$ and $V_\alpha$ converge.

Occurrence of the metal-insulator transition for the normal phase of the model (1) is judged by behaviors of both the quasiparticle weight $Z_\alpha$ and the interacting DOS $\rho^\alpha(\omega) = -\operatorname{Im} G^\alpha(\omega + i0^+)/\pi$, with varying $U$, $U'$, $t_h/t_e$ and also the particle density $n (\equiv n^e = n^h)$.

3. Phase diagram at half filling

For $t_h/t_e = 1$ (an effective mass of the hole is the same as that of the electron), the phase diagram on the plane of $U'$ and $U$ is shown in figure 1 [10]. There are three phases: (I) metallic state, (II) Mott-Hubbard insulating state, and (III) biexciton-like insulating state. The second-order phase transitions between these phases occur on the solid curves. The metallic and insulating states can be distinguished by whether the quasiparticle weight $Z_\alpha$ is nonzero or zero. In the metallic state (I), $Z_\alpha$ has a finite value and there is finite DOS at the Fermi level (the quasiparticle coherent peak), i.e., $\rho^\alpha(0) \neq 0$. On the other hand, in the both insulating states (II) and (III), $Z_\alpha = 0$ and the coherent peak of the DOS disappears. However, the physical pictures of the two insulating states (II) and (III) are quite different, as drawn schematically in insets of figure 1. This is confirmed also by evaluating and comparing the spin susceptibility and the double-occupancy rate. The state (II) is induced by the e-e (h-h) repulsion $U$ on each electron and hole band, while the state (III) results from the e-h attraction $U'$ on each site. The competition of these two states stabilizes the metallic state for $U \sim U'$. We note that these results are equivalent to those obtained for the two-orbital repulsive Hubbard model [11].

We have also examined effects of the difference between the electron and hole masses [10]. In figure 2, the phase diagram on the plane of $U'$ and $U$ is shown for $t_h/t_e = 0.5$. A new state (IV) appears between states (I) and (II), in which $Z_e \neq 0$ but $Z_h = 0$, i.e., the electron (hole) band is metallic (insulating). In other words, the Mott-Hubbard transition of holes does not coincide with that of electrons when $t_e \neq t_h$. This “band-selective” Mott-Hubbard transition corresponds to the “orbital-selective” Mott transition in the two-orbital repulsive model [12].

In the case of half filling, the following universal features are found; (a) The metal-insulator transition between states (I) and (III) is by no means “band-selective” for any ratio $t_h/t_e$, and (b) the position of that phase boundary on the plane of interactions scaled by $t_e + t_h$ is universal with regard to the ratio $t_h/t_e$. These facts indicate that the transition between the metallic state (I) and the biexciton-like insulator (III) occurs as a result of the competition between the interactions and the relative motion of electron and hole. Note that the quantity $t_e + t_h$ is proportional to the energy of the relative motion.
4. Optical absorption spectra at half filling

To distinguish several metallic and insulating phases, we have calculated optical responses of the system. In particular, optical absorption spectrum is one of important quantities, which can be observed in experiments. The response function $\chi(q, i\nu)$ is then need to be evaluated:

$$
\chi(q, i\nu) = \frac{1}{\beta} \sum_{\omega} \bar{\chi}_q(i\omega, i\omega'; i\nu)
= - \frac{1}{N} \int_0^\beta d\tau e^{i\nu\tau} \sum_{kk'} \sum_{\sigma\sigma'} \langle T_\tau d^h_{-k+q,-\sigma}(\tau) d^{\dagger}_k\sigma(\tau)d^{\dagger}_{k'\sigma'}(0)d^h_{-k'+q,-\sigma'}(0) \rangle,
$$

where $d^\dagger_{k\sigma}$ is an annihilation operator in the momentum space. The local vertex function $\Gamma^{\text{eh}}$, which represents the e-h correlation or the excitonic correlation, is defined through

$$
\bar{\chi}_q(i\omega, i\omega'; i\nu) = \bar{\chi}^{(0)}_q(i\omega; i\nu) \delta_{\omega', \omega} + \frac{1}{\beta} \sum_{\omega''} \bar{\chi}^{(0)}_q(i\omega; i\nu) \Gamma^{\text{eh}}(i\omega, i\omega''; i\nu) \bar{\chi}_q(i\omega'', i\omega; i\nu),
$$

where $\bar{\chi}^{(0)}_q(i\omega; i\nu) = -N^{-1} \sum_{k\sigma} G_{k\sigma}(i\omega) G_{-k+q,-\sigma}^{\text{eh}}(-i\omega + i\nu)$ is the response function without the vertex part. Optical absorption spectrum is proportional to $-|1 - \exp(-\beta\omega)|^{-1}\text{Im} \chi_{\text{ret}}(\omega)$ with the retarded response function $\chi_{\text{ret}}(\omega) = \chi(q = 0, i\nu \rightarrow \omega + i\delta)$ given by the analytic continuation $i\nu \rightarrow \omega + i\delta$. We employ the T-matrix approximation for the vertex function $\Gamma^{\text{eh}}$ to lead $\chi(q = 0, \omega)$ as

$$
\chi(q = 0, \omega) \simeq \frac{\chi^{(0)}(q = 0, \omega)}{1 + \frac{\beta}{2} \chi^{(0)}(q = 0, \omega)},
$$

where $\chi^{(0)}(q = 0, \omega) = \beta^{-1} \sum_{\omega''} \bar{\chi}^{(0)}_{q=0}(i\omega; i\nu \rightarrow \omega + i\delta)$.

We can show only preliminary results. Figure 3 (4) is the absorption spectrum in a metallic (insulating) phase at half-filling ($n = 1$) for $t_h/t_e = 1$ with the excitonic correlation through the vertex function $\Gamma^{\text{eh}}$ (solid line) and without it (broken line). In the metallic phase, “excitonic enhancement” of oscillator strength is observed near the absorption edge, while a distinguished peak structure appears as an “exciton in many-body Fermi seas” in an insulating phase. Details will be reported elsewhere.

**Figure 1.** Phase diagram in the $U'$-$U$ plane at half-filling ($n = 1$) for $t_h/t_e = 1$ [10].

**Figure 2.** Phase diagram in the $U'$-$U$ plane at half-filling ($n = 1$) for $t_h/t_e = 0.5$ [10].
5. Phase diagram at arbitrary filling

We discuss the case of arbitrary filling. For $n \neq 1$, the process for determining of the chemical potential $\mu_\alpha$ is added to the self-consistency cycle for $\varepsilon_\alpha^c$ and $V_\alpha$. We carried out also the exact diagonalization calculation to solve the SIAM in addition to the two-site DMFT. Hereafter $t_h/t_e = 1$ is fixed.

Figure 5 shows a sketch of the phase diagram on the plane of $U'$ and $U$ for $n = 0.25$. The Mott-Hubbard insulator disappears, while the metallic state and the biexciton-like insulator remain. A remarkable feature is appearance of new insulating phase, called the “exciton-like insulator phase” in rather strong repulsion (large $U$ and $U \geq U'$). This exciton-like insulator is characterized by that $Z_\alpha \neq 0$ but $\rho^s(0) = 0$ (i.e., a gap opens at Fermi level). In addition, phase transitions among these states 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 $-U'$. According to the results of the DMFT study of this model [13, 14], a paring state appears in addition to the metallic state. This paring state corresponds to the exciton-like insulating state in our model, in which incoherent local e-h pairs (do not condense) are formed.
6. Summary
In this paper, we obtained the phase diagram of the EMT for the two-band Hubbard model by using the DMFT with varying the parameters $U$, $U'$, $t_{h}/t_{e}$, and $n$. We found the phase transitions among the exciton-like insulator, biexciton-like insulator, the Mott-Hubbard insulator, and the metallic state. The EMT is found to be described essentially by the simple lattice model with only short-range (on-site) interactions.

However, the range of interactions is important for understanding the e-h systems in more detail. This requires advanced studies for not only static but also dynamical screening mechanisms in these systems. From this viewpoint, how the dimensionality affects the EMT is a crucial question. We have clarified only a part of this problem: (i) at the high-density limit in one dimension [8] and (ii) at an arbitrary density in large ($\infty$) dimensions. Several numerical techniques will be employed to attack this problem.

We shall discuss briefly the relevance of our assumption that the e-h pairs do not condense. Although the calculation was performed at zero temperature, we believe that our present results will be valid for the intermediate temperatures, above a critical temperature ($T_C$) of exciton BEC, but below temperature corresponding to the e-h binding energy ($E_B$). Such a temperature region actually exists. In the strong limit of $U'$, $T_C$ can be estimated as of order $(t_e + t_h)^2/U'$. On the other hand, in the low-density limit $n \to 0$, $E_B$ is of order $U'$. Comparing these, an intermediate temperature region exists actually even for not so large $U'$ ($\sim t_e + t_h$). Finite temperature effects and the e-h condensation processes are in investigation and will be reported elsewhere.

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