Effect of Particle-Hole Asymmetry on the Mott-Hubbard Metal-Insulator Transition

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The Mott-Hubbard metal-insulator transition is one of the most important problems in correlated-electron systems. In the past decade, much progress has been made in examining a particle-hole symmetric form of the transition in the Hubbard model with dynamical mean field theory, where it was found that the electronic self-energy develops a pole at the transition. We examine the particle-hole asymmetric metal-insulator transition in the Falicov-Kimball model and find that a number of features change when the noninteracting density of states has a finite bandwidth.

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The Mott-Hubbard metal-insulator transition [1,2] is a classic example of the physics of strongly correlated electrons. The physical mechanism of the transition arises from a local Coulomb repulsion U that forbids double occupancy of the electrons, creating an insulator when there is one particle per site (on average). Experimentally it is found in a variety of materials, including many transition metal compounds (MnO, NiO, NiS, $YBa_2Cu_3O_6$, etc.) for which band structure calculations severely underestimate the band gap or yield a metallic solution [3].

In the Hubbard model [2], a number of approximations have been employed that assume either the metallic state and head toward the insulator [2,4,5], or vice versa [2,6]. Often it is difficult to develop an approximate theory that is able to describe both the weakly correlated Fermi-liquid phase and the strongly correlated insulator; hence most approximate methods yield only limited information about the transition. Much progress has been made with dynamical mean field theory (DMFT) [7], where fundamental questions such as the following have been analyzed in great detail: Is the transition continuous or discontinuous? Does the Fermi-liquid metal survive up to the transition or do non-Fermi-liquid metallic phases intervene? Do metastable phases exist? However, the numerics are quite complicated and delicate because the different phases are separated by very small energies. Hence there has been much controversy about the answers to these questions and about the details of the Mott-Hubbard transition [8–14]; similar issues have been studied in the disordered case [15].

Some of these problems arise from the limit of infinite dimensions, where the noninteracting density of states (DOS) on a hypercubic lattice is a Gaussian, which has an infinite bandwidth. This means that the DOS can vanish (and thereby yield an insulator at $T = 0$) only when the self-energy diverges. Since this occurs only at the single point of an isolated pole, the DOS in the insulating phase is really a pseudogap and is nonzero (albeit exponentially small) in a region around the pseudogap. Hence, the metal-insulator transition (MIT) on a hypercubic lattice always occurs when the self-energy develops a pole (that lies infinitesimally below the real axis). It turns out that the same scenario occurs on the Bethe lattice at half filling, even though the DOS has a finite bandwidth; the MIT occurs when the self-energy develops a pole at the chemical potential (although here there is now a well-defined gap, where the DOS vanishes over a finite range of frequencies).

The question we wish to address is what happens when the MIT occurs for a system that does not possess particle-hole symmetry [16]. Following the argument given above, the pole formation must be the underlying cause of the MIT on the hypercubic lattice, but the situation on the Bethe lattice is unclear. The interest in examining particle-hole asymmetric cases lies in the fact that most real materials do not have particle-hole symmetry; so understanding consequences of breaking particle-hole symmetry is important for understanding experimental systems. There are two ways to break particle-hole symmetry: (i) one can modify the lattice, so it is not bipartite, and then there is no particle-hole symmetry at half filling, where the MIT occurs, or (ii) one can modify the model so that a MIT occurs at different fillings, away from the particle-hole symmetric limit. We choose to examine the latter here.

We consider the MIT in the spinless Falicov-Kimball (or simplified Hubbard) model [17], which is believed to describe correlated-electron behavior, and in particular, the MIT in materials that can be fit into a binary alloy picture. The canonical system that fits this picture is Ta_xN [18,19], which has its MIT occur at a particle-hole asymmetric value of $x = 0.6$. The Falicov-Kimball model has been previously applied to the MIT [20,21]. Its advantages over the Hubbard model are that it has a MIT for a wide range of fillings and the numerics are under much better control. The Falicov-Kimball Hamiltonian has the following form

$$H = -t\sum_{\langle i,j\rangle} c_i^\dagger c_j + U\sum_i w_i c_i^\dagger c_i, \tag{1}$$

where $c_i^\dagger$ (c_i) denotes the creation (annihilation) operator

for a spinless electron on site i, and the summation is restricted to nearest neighbors. The classical variable w_i equals one or zero, corresponding to the presence of an A or a B ion at the given lattice site, and U is the diagonal site-energy difference between the two ionic configurations. The hopping integral t is appropriately scaled in order to be finite in the limit of large dimensions (d) or coordination number (Z) [7] and to result in the same effective bandwidths $W = \sqrt{\int \epsilon^2 \rho(\epsilon) d\epsilon}$ (with $\rho(\epsilon)$ the noninteracting DOS) for both lattices [14]. Therefore, $t = t^*/\sqrt{Z}$ on the Bethe lattice and $t = t^*/\sqrt{2d}$ on the hypercubic lattice. The conduction electrons interact with the localized particles (which have an average filling of $w_1 = \langle w_i \rangle$) with an interaction strength U; this forms the canonical binary alloy picture. Such a picture is particularly useful because the model exhibits a MIT when the particle-hole symmetry is broken ($w_1 \neq 0.5$), as long as the total particle density equals one. Hence, we constrain our calculations to fix the total number of particles, i.e., the number of conduction electrons satisfies $\rho_e = 1 - w_1$. If we choose to measure all energies in the units of t^*, then the noninteracting DOS on the Bethe and hypercubic lattices are a semicircle and a Gaussian, respectively, $\rho_{\text{Bethe}}(\epsilon) = \sqrt{4 - \epsilon^2}/2\pi$ and $\rho_{\text{HC}}(\epsilon) = \exp(-\epsilon^2/2)/\sqrt{2\pi}$.

In the limit of infinite dimensions the Falicov-Kimball model can be solved exactly using DMFT [22,23]. Because the self-energy $\Sigma(\omega)$ has no momentum dependence, one can employ an iterative scheme using the following relationships between the self-energy $\Sigma(\omega)$, the retarded Green's function $G(\omega)$, and the effective medium $G_0(\omega)$:

$$G(\omega) = \int d\epsilon \rho(\epsilon) \frac{1}{\omega + \mu - \Sigma(\omega) - \epsilon + i\delta} \quad (2)$$

$$G_0(\omega) = [G(\omega)^{-1} + \Sigma(\omega)^{-1}]^{-1} \quad (3)$$

$$G(\omega) = (1 - w_1) G_0(\omega) + w_1 \frac{1}{G_0(\omega)^{-1} - U} \quad (4)$$

closed in the iterative loop until the self-energy is converged to a desired accuracy [24]. The algorithm normally converges to about 13 digits when iterated. The calculations are particularly easy in the case of the Bethe lattice since the integral in Eq. (2) can be determined analytically (for the hypercubic lattice it is a complex error function). Once the algorithm is converged, the interacting DOS is defined to be $\rho_{\text{int}}(\omega) = -\text{Im}[G(\omega)]/\pi$. The essential difference in the physics on these two lattices arises from the behavior of the interacting DOS. While the Bethe lattice exhibits a well-defined gap where the interacting DOS is exactly zero for a finite range of frequencies when $U > U_{cg}$ (the critical value of the interaction strength for opening a gap in the interacting DOS), on the hypercubic lattice the DOS splits into two subbands divided by a "pseudogap," where the interacting DOS is exponentially small and exactly zero only at one point. The interacting DOS away from half filling has appeared elsewhere [20,25]. We should note that in the Falicov-Kimball model there is no quasiparticle peak at the chemical potential, which normally develops for U less than U_{cg} in the Hubbard model in infinite dimensions. This is because the Falicov-Kimball model lacks quasiparticles in the strict definition of the term, since the lifetime of Fermionic excitations never becomes infinite as $T \to 0$.

The metal-insulator transition at half filling is closely related to the development of a pole in the self-energy, where the DOS is suppressed to zero (U_{cg}) at the same interaction strength for which the pole forms (U_{cp}); i.e., $U_{cg} = U_{cp}$. Therefore one could suggest to use the residue of the pole as an order parameter for the MIT. In fact, a plot of the residue versus $(U - U_{cp})/U_{cp}$ is universal, for all fillings on both the hypercubic and Bethe lattices, indicating that a scaling theory holds for the residue of the pole (see inset of Fig. 1). Away from half filling, these two processes (pole formation and the MIT) are decoupled on the Bethe lattice, with the pole formation occurring after the MIT (in particular, the real part of the Green's function does not cross the horizontal axis [within the band gap] until the pole forms).

We begin with the cubic equation that is satisfied by the Green's function on the Bethe lattice [20]:

$$G^3 - 2xG^2 + \left(1 + x^2 - \frac{U^2}{4}\right) G - (x + \alpha) = 0, \quad (5)$$

where $x \equiv \omega + \mu - U/2$ and $\alpha \equiv U(w_1 - \frac{1}{2})$. The pole in the self-energy develops when $G = 0$, requiring $x + \alpha = 0$, or $\omega_{\text{pole}} = U(1 - w_1) - \mu$. Next, one notes that

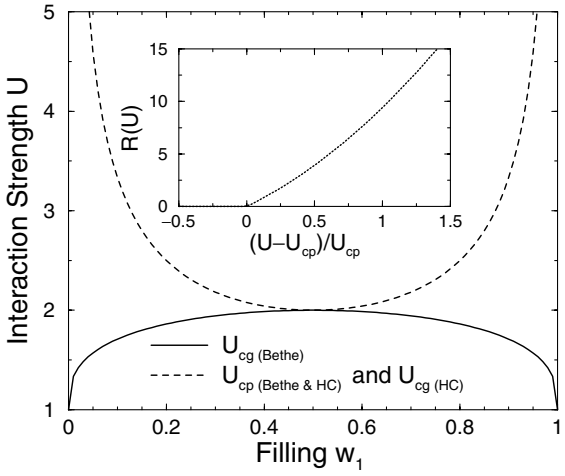

FIG. 1. Phase diagram for the MIT in the Falicov-Kimball model on the Bethe and hypercubic lattices. The solid line denotes U_{cg} for the gap formation on the Bethe lattice, the dashed line denotes U_{cp} for the pole formation on the Bethe lattice, and $U_{cg} = U_{cp}$ on the hypercubic lattice. The inset shows the universal curve of the residue of the pole $R(U)$ as a function of the interaction strength for both lattices and all fillings.

$G = 0$ is a physical root [26] when $1 + x^2 - U^2/4 \leq 0$, which leads to the simple formula for U_{cp}, the critical value of U at which the self-energy develops a pole

$$U_{cp} = \frac{1}{\sqrt{w_1(1 - w_1)}}. \quad (6)$$

Away from half filling, the critical values of U for the MIT and the pole formation are not necessarily the same on the Bethe lattice, and the former is found from the following. Let us use standard notation for the cubic equation's coefficients (see [27]) $q \equiv (3 - x - 3U^2/4)/9$ and $r \equiv x(1 + U^2/2)/6 + \alpha/2 - x^3/27$. The condition for the location of the band edges ($q^3 + r^2 = 0$) is a fourth order equation in ω and must have exactly three distinct roots at U_{cg}, the critical interaction strength for the MIT (four distinct roots for $U > U_{cg}$). As shown in Ref. [20], this condition leads to an equation for the critical interaction:

$$1 - 4\frac{\alpha^2}{U^2} = \frac{4(U^2 - 1)}{27U^2}. \quad (7)$$

Solving Eq. (7) for U yields the critical interaction strength for the MIT (gap opening on a Bethe lattice) U_{cg}:

$$U_{cg} = \sqrt{1 + 3w_1^{1/3}(1 - w_1)^{1/3}[(1 - w_1)^{1/3} + w_1^{1/3}]}. \quad (8)$$

Both expressions (6) and (8) yield two at half filling ($w_1 = 0.5$). The behavior of U_{cp} and U_{cg} for arbitrary fillings is shown in Fig. 1. As the system moves away from half filling, the particle-hole asymmetry allows for the formation of a third "phase," in which the interacting DOS has a gap, but there is no pole in the self-energy (region between the solid and dashed lines in Fig. 1 on the Bethe lattice). This dramatic difference in the MIT as we move away from half filling is most likely due to the difference between an infinite bandwidth and a finite bandwidth [28]. On the hypercubic lattice, the pole formation and MIT always occur at the same value of U ($U_{cg} = U_{cp}$), and it is possible to obtain an exact expression for the pole formation, which turns out to coincide with Eq. (6). Numerical calculations show that the interacting DOS for the hypercubic lattice has an exponentially small region over a similar range in frequency as the gap region on the Bethe lattice (for the same value of U).

Figure 2 shows the relative interaction strength as a function of the relative location of the pole within the gap, on the Bethe lattice. The pole is located in the middle of the gap at half filling for any value of $U > U_{cg}$. However, as the particle-hole symmetry is broken, the pole first appears at the lower or upper band edge (for $U = U_{cp}$), depending on whether w_1 is larger or smaller than 0.5, and as U increases the pole drifts closer to the center of the band gap. Note that there is no smooth transition between the half-filled case and the particle-hole asymmetric case.

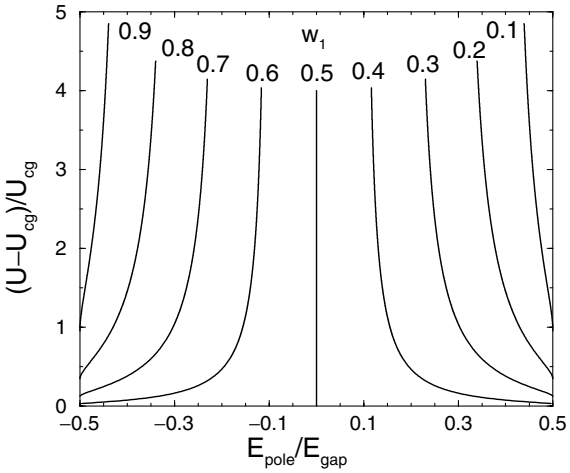

FIG. 2. The relative interaction strength for the opening of a gap versus the relative location of the pole (E_{pole}) in the gap (of width E_{gap}) on the Bethe lattice; the origin on the horizontal axis lies at the center of the gap. The lines from left to right correspond to different fillings w_1 ranging from 0.9 to 0.1, in steps of 0.1.

Numerical calculations show the evolution of the real part of self-energy on the Bethe lattice compared to that on the hypercubic lattice in Fig. 3, when approaching the MIT from the metallic side. Aside from obvious similarities between the two lattices, this graph demonstrates the existence of the third phase in the middle panel, where $U = 2$. The half-filled curves ($w_1 = 0.5$) always show poles in the insulator, and therefore the DMFT scenario discussed in Refs. [5,11,14] holds. However, the curves corresponding to $w_1 = 0.25$ exhibit large (negative) but finite values of $\text{Re}[\Sigma(\omega)]$ (the pole has not yet developed), but the system is an insulator on the Bethe lattice with a well-developed gap at this value of U. (Both $\text{Re}[\Sigma(\omega)]$ and $\text{Re}[G(\omega)]$ develop kinks at the new band gap edges, but there is no obvious way to use the presence of a kink

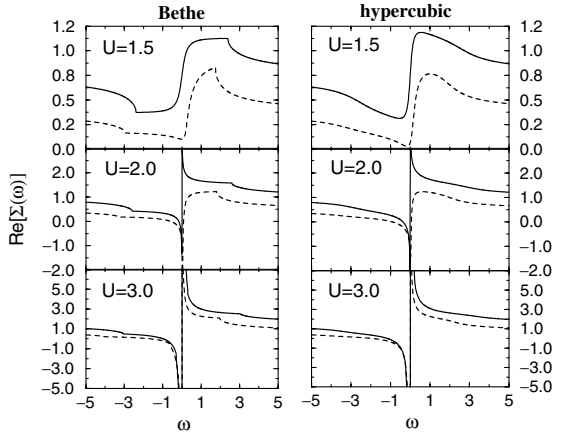

FIG. 3. Evolution of the real part of the self-energy for different values of U, for both the Bethe (left panel) and hypercubic (right panel) lattices, at half filling $w_1 = 0.5$ (solid line) and at $w_1 = 0.25$ (dashed line). The origin of the frequency axis is set to $U(1 - w_1) - \mu$.

as an order parameter for the MIT.) Hence, we conclude that the development of the pole and the MIT are decoupled away from half filling and, even if it might be tempting to use the residue of the pole as an "order parameter" for the MIT, it fails to describe the situation off of half filling on lattices with a finite bandwidth. So what significance can be made of the pole formation? In order to investigate this, we calculate the dc conductivity in the relaxation time formalism (see [29] for details). These calculations (not shown here) indicate that the MIT is always a continuous transition at $T = 0$, with the conductivity being suppressed continuously to zero as $U \rightarrow U_{cg}$. Also, at finite temperature we see no evidence for the influence of the pole on the transport. The conductivity curves are smooth functions of T with no unusual features occurring when $U > U_{cp}$.

On the hypercubic lattice, one finds the imaginary parts of $G(\omega)$ and $\Sigma(\omega)$ exponentially approaching zero as $\omega \rightarrow U(1 - w_1) - \mu$ [25], while they are exactly zero on the Bethe lattice. These, at first sight small differences, in fact turn out to cause dramatic changes in the low temperature transport properties [25,30] when going from the Bethe lattice (where the relaxation time vanishes in the gap) to the hypercubic lattice (where the relaxation time has a power-law dependence around the pseudogap). The most significant departure is in thermal transport properties, where the thermopower diverges on the Bethe lattice, but vanishes on the hypercubic lattice in the insulating phase as $T \rightarrow 0$ (when particle-hole symmetry is broken).

In conclusion, we have analyzed the effect of particle-hole asymmetry on the Mott transition in the infinite dimensional Falicov-Kimball model (on both the Bethe and the hypercubic lattices). Hitherto, it was believed that the scenario for the MIT on both lattices was the same, as indeed is the case at half filling. We find that this is not true when the particle-hole symmetry is removed, as is often the case in real materials. We show that in the absence of particle-hole symmetry the pole formation and the MIT are two unrelated processes on the Bethe lattice. So even though the residue of the pole satisfies many of the properties expected of an order parameter, it cannot be employed to describe the MIT in all cases (although one could use it on the hypercubic lattice). Furthermore, there seems to be little difference between the properties of a correlated insulator with or without a pole in the self-energy. We conjecture that all of the conclusions about the character of the MIT on the Bethe lattice will hold for other systems with a finite bandwidth, and hence much of these results will play a role in realistic models of the MIT for real materials in finite dimensions.

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