Impurity induced insulator-to-metal transitions in half-filled Mott insulators

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Abstract. In view of cuprate superconductors, effects of point-type impurity potential \( V \) on half-filled antiferromagnetic and paramagnetic states are studied for a strongly correlated square-lattice Hubbard model \( (U/t = 12) \) with a diagonal transfer \( (t') \), using a variational Monte Carlo method. In the trial states, we introduce a Rice-Brinkman-type one-body projector for \( V \), which enables us to treat the whole range of \( V (\pm \infty < V/t < \infty) \) with small statistical errors. Filling-control-type Mott (insulator-to-metal) transitions are found to occur at \( V \approx \pm U \). In the metallic states \( |V| > |V^{(\pm)}| \), charge carriers are holes (electrons) in the case of attractive (repulsive) potential. The mechanism of the transitions is intuitively understood.

1. Introduction

Cuprate superconductors (SCs) are in the regime of Mott physics, and have inherent disorder originating in the carrier dopants. Furthermore, the impurity effect of apical oxygen in the T'-structure cuprates (typical for electron-doped systems) has been widely noticed [1] in connection with the so-called non-carrier-doped SCs [2]. Thus, it is important to clarify the impurity effects on strongly correlated systems.

In the previous reports, we studied the effects of point-type impurity potentials \( V \) on antiferromagnetic (AF) and paramagnetic (PM) states using a variational Monte Carlo (VMC) method. We mainly discussed \( U/t \) dependence of the impurity effect and also \( V/t \) dependence for large values of \( U/t \) in attractive potential \( (V < 0) \) [3], and metal-to-insulator (Mott) transitions occurring in partially-filled systems for \( \delta_{\text{imp}} \approx \delta \) \( (\delta_{\text{imp}}: \text{impurity density}, \delta: \text{doping rate}) \) in weakly repulsive potential \( (V = V_M = 0 - 2t) \) [4]. However, the VMC optimization with the trial states used in these studies, \( \Psi(#x) \) \( (x = 1 - 6) \), often brought about uncontrollable large statistical fluctuations for \( V > V_M \), which prevented us from making a systematic study in the repulsive potential. From the VMC data, we presume the cause of this shortcoming of \( \Psi(#x) \) to be the fact that many-body correlation factors, which distinguish the impurity and host (non-impurity) sites, are used for controlling the one-body impurity potential prior to the original role.

To remedy such shortcomings, we consider a new trial state \( \Psi(\theta) \), in which a one-body projector for \( V (P_\theta) \) [5] is introduced and many-body correlation factors do not depend on the attribute of sites (impurity or host). This trial state has another merit, namely, simple structure with respect to an often-used electron-hole transformation. Consequently, the properties of repulsive potential, especially at half filling, are expected from those of attractive potential, and...
vice versa. $\Psi(\theta)$ basically exhibits behavior similar to $\Psi(\#x)$ for $V < V_M$. In this article, using $\Psi(\theta)$, we discuss insulator-to-metal transitions occurring in strong impurity potential and other properties at half filling.

2. Formalism

2.1. Model

In this study, we consider a Hubbard model ($U \geq 0$) with a point-type spin-independent impurity potential ($V$) on extended square lattices:

$$\mathcal{H} = \mathcal{H}_{\text{kin}} + \mathcal{H}_U + \mathcal{H}_{\text{imp}} = - \sum_{(i,j),\sigma} t_{ij} (c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.}) + U \sum_j n_{j\uparrow} n_{j\downarrow} + V \sum_{\ell=1}^{N_{\text{imp}}} \sum_{\sigma} n_{\ell\sigma},$$

(1)

where $n_{j\sigma} = c_{j\sigma}^\dagger c_{j\sigma}$, $(i, j)$ indicates the pairs on sites $i$ and $j$, and $N_{\text{imp}}$ is the number of impurity sites, which we will explain shortly. Here, we set the hopping integral $t_{ij}$ as $t$ for nearest neighbors ($\geq 0$), $t'$ for diagonal neighbors, and 0 otherwise ($\mathcal{H}_{\text{kin}} = \mathcal{H}_t + \mathcal{H}_{t'}$). We use $t$ and the lattice spacing as the units of energy and length, respectively. Here, we focus on the cases at half filling ($n = N/N_s = 1$ or $\delta = |1 - n| = 0$, $N$: number of electrons, $N_s$: number of sites) and of typical impurity densities $\delta_{\text{imp}} = N_{\text{imp}}/N_s \sim 0.08$ in strong correlation ($U/t = 12$). We assume that the impurity potential $V$ comes from a single species, for example, apical oxygen atoms in a $T'$-structure system [1], and act on a nearby single site (“impurity site”) for simplicity. If we grant this simplification, the index $\ell$ in $\mathcal{H}_{\text{imp}}$ runs over the impurity sites $r_{\ell}$ ($\ell = 1, \cdots, N_{\text{imp}}$). Furthermore, we assume that $r_{\ell}$ are randomly distributed. We widely varies $V$ (even for $V < 0$) to grasp overall features of impurity effects. Although the above setting of impurity potential is possibly simplistic, we believe that fundamentals of impurity effects with strong correlation can be captured.

2.2. Method

We apply a many-body variation theory to the above model. In computing expectation values, we employ a VMC method [6, 7, 8], which enables us to exactly treat many-body wave functions. As a trial wave function, we use a Jastrow type: $\Psi = \mathcal{P}\Phi$. In the previous studies [3, 4], we distinguished the impurity and host sites in the (many-body) projection factor $\mathcal{P}$ in $\Psi(\#x)$. We found, however, that this leads to the uncontrollable fluctuation in the VMC optimization for $V > V_M$, as mentioned. Therefore, in the new trial state $\Psi(\theta)$, we do not distinguish the impurity and host sites in the many-body projection factors, $\mathcal{P} = \mathcal{P}_H \mathcal{P}_\sigma \mathcal{P}_Q \mathcal{P}_G$, where $\mathcal{P}_H$, $\mathcal{P}_\sigma$, $\mathcal{P}_Q$, and $\mathcal{P}_G$ are a nearest-neighbor charge [4] and spin ($\mathcal{P}_\sigma = \beta^\sigma$ with $\beta$ being a parameter and $\sigma = 4 \sum_{(i,j)} S_i^z S_j^z$) correlators, a nearest-neighbor doublon-holon binding factor, and onsite (Gutzwiller) factor, respectively, where doublon (holon) indicates a doubly occupied (empty) site. For $\mathcal{P}_Q$ and $\mathcal{P}_G$, we adopt the same forms as those used for the uniform systems [8]. Instead of controlling the effect of impurity potential using many-body correlators, we introduce a one-body projector [5]

$$\mathcal{P}_\theta = \theta^\kappa, \quad \text{with} \quad \kappa = \sum_{\ell,\sigma} n_{\ell\sigma},$$

(2)

and $\theta$ being a parameter. Thus, the new trial state becomes $\Psi(\theta) = \mathcal{P}\mathcal{P}_\theta\Phi$.

To obtain the Slater-determinantal part $\Phi$, we diagonalize the following mean-field Hamiltonian of spin $\sigma$,

$$\mathcal{H}_{\text{MF}}^{(\sigma)} = \mathcal{H}_{\text{kin}}^{(\sigma)} + U \sum_j \langle n_{j\sigma} \rangle n_{j-\sigma} + \mathcal{H}_{\text{imp}}^{(\sigma)},$$

(3)
Table 1. Relationship of elements in Hamiltonian Eq. (1), related quantities (defined later), and variational (and fixed) parameters in trial wave function $\Psi(\theta)$ through unitary transformation Eq. (4) at half filling. In the column of physical quantities, the quantity in the left[right]-hand side indicates, say, $n_{\text{imp}}(V,t') [2 - n_{\text{imp}}(-V,-t')]$.

| Hamiltonian | Physical quantities | Variational (fixed) parameters |
|-------------|---------------------|-------------------------------|
| $\mathcal{H}_t \leftrightarrow \mathcal{H}_t$ | $n_{\text{imp}} \leftrightarrow 2 - n_{\text{imp}}$ | $g \leftrightarrow g$ |
| $\mathcal{H}_t' \leftrightarrow -\mathcal{H}_t'$ | $n_{\text{hst}} \leftrightarrow 2 - n_{\text{hst}}$ | $\zeta_d \leftrightarrow \zeta_h$ |
| $\mathcal{H}_U \leftrightarrow \mathcal{H}_U$ | $m \leftrightarrow m$ | $t_1 \leftrightarrow -t_1$ |
| $\mathcal{H}_V \leftrightarrow -\mathcal{H}_V + 2VN_{\text{imp}}$ | $d \leftrightarrow h$ | $\eta \leftrightarrow \eta$ |

In the kinetic part $\mathcal{H}_{\text{kin}}^{(\sigma)}$, a band-renormalization effect is introduced in the same manner as in Refs. [3, 4, 8]. In the second term, we take account of an AF long-range order of the nesting vector $\mathbf{Q} = (\pi, \pi)$ also in the same manner as in the previous studies. In the last term, we use the raw impurity term in the Hamiltonian to avoid the redundancy of the screening effect, that is, because $V_{\text{eff}}/V$ in $\Psi(\#x)$ and $\mathcal{P}_\theta$ play similar roles. Thus, $V$ in $\Psi(\theta)$ is a constant given in Eq. (1). Diagonalizing $\mathcal{H}_{\text{MF}}^{(\sigma)}$ of $N_s \times N_s$ matrices, we obtain eigenenergies $\varepsilon_n$ and eigenvectors $\phi_{n\sigma}$ with corresponding quasiparticle operators $\hat{a}_{n\sigma}^\dagger$, which are linear combinations of $c_{\sigma}^{\dagger}$. Using $\phi_{n\sigma}$, one-body part is constructed as $\Phi = \prod_n (\varepsilon_n \leq \varepsilon_F) \prod_{\sigma} \phi_{n\sigma}^\dagger |0\rangle$, where $\varepsilon_F$ indicates the energy of the highest occupied level (HOMO). $\Psi(\theta)$ is reduced to the exact noninteracting ground state of Eq. (1) for $U = 0$ and to the corresponding uniform states for $V = 0$. The AF state $\Psi_{\text{AF}} = \mathcal{P}_\theta \Phi_{\text{AF}}$ is reduced to the PM state $\Psi_{\text{PM}} = \mathcal{P}_\theta \Phi_{\text{PM}}$ for $\delta_{\text{AF}} \to 0$ [4] and $\beta \to 1$.

We compute expectation values with respect to $\Psi(\theta)$ using a VMC method similar to that in Ref. [8]. We use systems of $N_s = L \times L$ sites with $L = 10 - 16$ and the periodic-antiperiodic boundary conditions. In a VMC sweep, we use 20,000 samples, because the convergence of $\Psi(\theta)$ is much better than $\Psi(\#x)$. A configuration of impurities $\{\mathbf{r}_i\}$ is randomly chosen and fixed throughout a single sweep of VMC calculation (optimization and measurement). In most cases, we will display data of 4 kinds of $\{\mathbf{r}_i\}$ simultaneously.

2.3. Electron-hole transformation

The electron-hole (unitary) transformation $\mathcal{U}$ defined below is often used to treat a more-than-half-filled system, especially in cases with $\mathcal{H}_t'$, in a less-than-half-filled band:

$$\mathcal{U}^{-1} \hat{c}_{j\sigma}^\dagger \mathcal{U} \rightarrow e^{i\mathbf{Q} \cdot \mathbf{r}_j} \hat{c}_{j\sigma}^\dagger, \quad \text{or} \quad \mathcal{U}^{-1} \hat{c}_{\mathbf{k}\sigma}^\dagger \mathcal{U} \rightarrow \hat{c}_{\mathbf{Q} - \mathbf{k}\sigma}. \quad (4)$$

The expectation value of an observable $O$ with respect to a wave function $|\Psi\rangle$ is transformed as

$$O_{\text{avr}} = \langle O \rangle = \frac{\langle \Psi | O | \Psi \rangle}{\langle \Psi | \Psi \rangle} = \frac{\langle \tilde{\Psi} | \tilde{O} | \tilde{\Psi} \rangle}{\langle \tilde{\Psi} | \tilde{\Psi} \rangle} = \langle \tilde{O} \rangle,$$

with $\tilde{O} = \mathcal{U}^{-1} O \mathcal{U}$ and $|\tilde{\Psi}\rangle = \mathcal{U}^{-1} |\Psi\rangle$. At half filling, the original forms and transforms of entities often have simple relations (Table 1), which are useful for connecting the cases of attractive and repulsive potentials and for checking the correctness of calculations. The trial wave function $\Psi(\theta)$ is transformed by simply substituting the variational parameters as in the right column of Table 1, in contrast with the former wave function $\Psi(\#6)$. As for the Hamiltonian Eq. (1),
\[ E(V, t') - V \delta_{\text{imp}} \equiv E(V, t') - (-V \delta_{\text{imp}}) = E(-V, -t') \],

where the model parameters other than \( V \) and \( t' \) are fixed. As another example, the staggered magnetization (an AF order parameter),

\[ m = \frac{2}{N_s} \left| \sum_j e^{i\mathbf{Q}\cdot \mathbf{r}_j} \langle S_j^z \rangle \right|, \]

satisfies the relation

\[ m(V, t') = m(-V, -t'). \]

Thus, \( \mathcal{E} \) and \( m \) are symmetric with respect to \( V = 0 \), if the sign of \( t' \) is reversed along with the sign of \( V \). Figs. 1(a) and 1(b) show the \( V/t \) dependence of \( E(V, t') \) and \( m(V, t') \), respectively, actually calculated by the VMC method for the AF state. The cases for \( t'/t = -0.3 \) and \( +0.3 \) are simultaneously plotted with blue (red) symbols and lines. We can confirm that the above symmetry, namely Eqs. (6) and (8) are satisfied, indicating the VMC calculations are correct.

### 3. Insulator-to-metal transitions

In the uniform cases (\( V = 0 \)), the AF and PM (\( U > U_c \approx 8t \)) states are insulating. In Fig. 1(a), \( E \) exhibits rapid variations of slope at \( V = V_U^{(+)} \approx \pm 11t \); correspondingly, in Fig. 1(b), \( m \) rapidly drops at \( V = V_U^{(+)} \) as \( |V| \) increases. In particular for \( V > 0 \) and \( t'/t = -0.3 \) (equivalently for \( V < 0 \) and \( t'/t = +0.3 \)), the behavior at \( V = V_U \), namely discontinuity of \( m \), suggests first-order transitions.

To study these anomalies, let us consider the momentum distribution function,

\[ n(\mathbf{k}) = \frac{1}{2} \sum_{\sigma} \langle c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} \rangle. \]

Quantities defined in the wave number space, such as \( n(\mathbf{k}) \) and the charge-density structure factor \( N(\mathbf{q}) \) [Eq. (10)], are basically not well-defined in systems without translational symmetry.
Nevertheless, they provide us with useful information as far as the electronic states preserve some homogeneous nature, or information as to how the randomness affects the electronic states, as discussed below. In Fig. 2(a), we show \( n(k) \) for five different values of \( V/t \) on the indicated path of \( k \). For \( V_U^- < V < V_U^+ \), \( n(k) \) is smooth and does not have Fermi surface (FS) (discontinuity) at any point on this path, meaning that the state is insulating. Note that the values for \( V/t = -4, 0 \), and 4 almost coincide; the feature of the homogeneous case \( V = 0 \) as an insulator is preserved within this regime. This is understood by the fact that the screening of \( V \) is almost complete there. Because it is not easy to directly see the screening effect in \( \Psi(\theta) \), we show in Fig. 3 the screening parameter \( V_{\text{eff}}/V \) obtained for a similar wave function \( \Psi(#6) \) [4], in which the screening becomes complete for \( V_{\text{eff}}/V = 0 \) and vanishes for \( V_{\text{eff}}/V = 1 \). The screening of \( V \) is almost complete for \( V_U^- < V < 0 \), whereas the effect of \( V \) somewhat remains for \( V < V_U^- \). This difference of screening effect is marked in the charge-density structure factor

\[
N(q) = \frac{1}{N_v} \sum_{i,j} e^{iq(r_i - r_j)} \langle n_i n_j \rangle - n^2, \tag{10}
\]

as shown in Fig. 2(b). It shows that for \( |V| < \left| V_U^{(\pm)} \right| \), the electron configuration is hardly affected by \( V \), whereas for \( |V| > \left| V_U^{(\pm)} \right| \) the electron configuration largely depends on the impurity configuration.

Returning to Fig. 2(a), for \( V < V_U^- \), we find that a pocket FS appears around \((\pi/2, \pi/2)\) in the nodal direction \((0,0)-(\pi, \pi)\), as seen for \( V/t = -16 \). It means that an insulator-to-metal transition occurs at \( V = V_U^- \) and a metallic state is realized for \( V < V_U^- \). Because \( n(k) \) decreases in the pocket compared to the insulating cases, the carriers are expected to be holes. On the other hand for \( V > V_U^+ \), \( n(k) \) exhibits a pocket FS around the antinodal point \((\pi, 0)\), as seen for \( V/t = +16 \). A similar Mott transition occurs at \( V = V_U^+ \) and the state becomes metallic for \( V > V_U^+ \). In this case, the carriers are considered electrons.

**Figure 2.** (a) Momentum distribution function and (b) charge-density structure factor for AF state along path of \( k \) (or \( q \)), \((0,0)-(\pi,0)-(\pi,\pi)-(0,0)\) for five different values of \( V/t \) at half filling. The data for a value of \( V/t \) are computed for a fixed impurity configuration, which is different for different \( V/t \).
To clarify the nature of this Mott transition, we consider the average electron densities on the impurity sites \( n_{\text{imp}} \) and the host sites \( n_{\text{hst}} \) and the average doublon density \( d \) calculated as

\[
n_{\text{imp}} = \frac{1}{N_{\text{imp}}} \sum_{\ell=1}^{N_{\text{imp}}} \sum_{\sigma} \langle n_{\ell\sigma} \rangle, \quad n_{\text{hst}} = \frac{N - N_{\text{imp}}n_{\text{imp}}}{N_{s} - N_{\text{imp}}}, \quad d = \frac{1}{N_{s}} \sum_{j} \langle n_{j\uparrow}n_{j\downarrow} \rangle. \quad (11)
\]

In Fig. 4, \( n_{\text{imp}} \) and \( n_{\text{hst}} \) are shown as functions of \( V/t \). In the insulating area \( (V_{U}^{(-)} < V < V_{U}^{(+)}), \) \( n_{\text{hst}} \) is nearly 1, namely, half filling is realized in the host sites. On the other hand for \( V < V_{U}^{(-)} \) \( (V > V_{U}^{(+)}), \) \( n_{\text{hst}} \) approaches \( 1 - (+)|N_{\text{imp}}/N_{\text{hst}} (N_{\text{hst}} = N_{s} - N_{\text{imp}}): \) number of host sites, because \( |V| \gg U \) and the impurity sites are almost occupied by doublons (holons). Therefore, for \( V < V_{U}^{(-)} \) \( (V > V_{U}^{(+)}), \) the charge carriers (indicated by double-headed arrows in Fig. 4) are holons (doublons), which is consistent with the feature of \( n(k) \) discussed above. This feature is reflected in the average doublon density shown in Fig. 5. In the metallic regimes, \( d \) increases up
Figure 5. Average doublon (doubly occupied sites) densities in the whole system as a function of impurity potential for AF and PM cases. The contribution owing to that all impurity sites are occupied by doublons or holons is $N_{\text{imp}}/N_{\text{host}} \approx 0.087$ in this case. The residual part is the contribution from local doublon-holon-pair creation.

Figure 6. Explanatory drawing of impurity-induced insulator-to-metal transition at half filling in Mott regime ($U > U_c$). (a) Weakly and intermediately attractive cases ($|V| \lesssim U$), (b) strongly attractive cases ($|V| \gtrsim U$), (c) weakly and intermediately repulsive cases ($V \lesssim U$), and (d) strongly repulsive cases ($V \gtrsim U$).

Attractive

(a) $V_{U^\ast} < V < 0$

All electrons $\rightarrow$ localized

(b) $V < V_{U^\ast}$

H: Free holon (→ itinerant)
D: Trapped doublon

Repressive

(c) $0 < V < V_{U_{\ast}}$

All electrons $\rightarrow$ localized

(d) $V > V_{U_{\ast}}$

H: Trapped holon
D: Free doublon (→ itinerant)

Figure 6 schematically shows this mechanism of the Mott transition. In the cases of weak and intermediate potential ($|V| \lesssim U$) shown in (a) and (c), every site is occupied by an electron except when a local D-H pair is produced. These cases are basically in the same situation as the pure case ($V = 0$), in which an ordinary Mott insulating state is realized. On the other hand in the case of strong potential ($|V| \gtrsim U$), almost all impurity sites are occupied by doublons or holons. As a result, the counter holons (+-charged carriers) or doublons (-charged carriers) to 0.1; for attractive $V$, doublons are created and trapped in the impurity sites, and the pair-produced holons are in the host sites and become the carriers. For repulsive $V$, every impurity site is occupied by a holon and the pair-produced doublon freely moves in the host sites.
are created in the host sites and move around freely. From such an intuitive picture, the Mott transition point is roughly estimated to be $|V_U^{(±)}| ∼ U - |E_{\text{kin}}^{(D/H)}|$, where $E_{\text{kin}}^{(D/H)}(\sim -t)$ is the kinetic energy of free carriers (doublon/holon) in the host sites. This estimation seems appropriate for the AF case ($|V_U^{(±)}| ∼ 11t$), but somewhat larger for the PM case ($|V_U^{(±)}| ∼ 7t$).

4. Conclusion
We studied the effects of point-type impurity potentials on a strongly correlated AF and PM states at half filling, using a VMC method. We showed that a filling-control-type Mott transitions take place for strong impurity potential ($V = V_U^{(±)} ∼ ±U$). The mechanism is intuitively expected. In fact, we have found that such insulator-to-metal transitions are not restricted to the present case of $\delta = 0$ but manifest themselves widely for $\delta_{\text{imp}} > \delta > 0$ with $V > 0$. We will deliver a more systematic report elsewhere. Similar Mott transitions have been also found in a superconducting state [10]. It is important to consider how the Mott transitions discussed here are related to the behavior of cuprates, or more generally to phenomena in other materials.

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