Impurity-induced Mott transitions in partially filled antiferromagnetic states

Hisatoshi Yokoyama¹, Ryo Sato¹ and Kenji Kobayashi²

¹Department of Physics, Tohoku University, Aoba-ku, Sendai, 980-8578, Japan
²Department of Natural Science, Chiba Institute of Technology, Narashino, 275-0023, Japan
E-mail: yoko@cmpt.phys.tohoku.ac.jp

Abstract. In view of cuprate superconductors, effects of point-type repulsive impurity potential \(V\) on partially filled antiferromagnetic (AF) states are studied for a strongly correlated square-lattice Hubbard model \((U/t = 12)\) with a diagonal transfer \(t' = -0.3t\), using a variational Monte Carlo method. To simplify interpretation, here we focus on the case of \(\delta_{\text{imp}} = 0\), where \(\delta_{\text{imp}}\) and \(\delta\) are densities of impurity sites and doped holes, respectively, with \(\delta = 0.04 - 0.20\). As a result, a filling-control-type Mott transition occurs at \(V = V_M (\sim 2t)\) irrespective of \(\delta\); a partially filled AF state becomes Mott insulating for \(V > V_M\). Such impurity-induced Mott transitions extensively appear beyond the above parameter settings.

1. Introduction

Recent numerical studies on uniform Hubbard-type models using refined techniques [1, 2] argued that antiferromagnetic (AF) long-range orders (or phase separation) prevail in wide ranges of doping rate \(\delta\) and frustration \(t' = t\). To cope with the inconsistency with experiments on cuprate superconductors, in which AF orders rapidly vanish on carrier doping, we have paid attention to the effects of impurity potential caused by the dopant ions inherent in cuprates. Another source of impurity effects is apical oxygen in the T'-structure cuprates (typical for electron-doped systems), which is considered to induce robust AF orders, especially, in as-grown samples [3].

In the preceding report [4], we studied effects of point-type impurity potential \(V\) on an AF state, mainly regarding \(U/t\) dependence and cases of attractive potential \((V < 0)\). As a result, screening of \(V\) owing to \(U\) is so effective in a strongly correlated regime \((U \gtrsim W, W: \text{band width})\) with \(-U/2 \lesssim V < 0\) that the effects of \(V\) thoroughly disappear, namely, the properties of uniform AF state \((V = 0)\) are preserved in the above range of \(V\) [5]. A doped AF state remains metallic for \(V < 0\).

As a continuation, in this report, we discuss the cases of repulsive impurity potential \((V > 0)\), which actually correspond to the potential owing to the carrier dopants and apical oxygen atoms in cuprates, in a strongly correlated regime. It is found that, in contrast with the attractive cases, filling-control-type Mott transitions extensively arise at \(V \sim 2t\), and partially filled AF states often become insulating.
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:

\[
\mathcal{H} = \mathcal{H}_{\text{kin}} + \mathcal{H}_U + \mathcal{H}_{\text{imp}} = -\sum_{(i,j),\sigma} t_{ij} \left( c_i^{\dag} c_j + \text{H.c.} \right) + U \sum_j n_{j\uparrow} n_{j\downarrow} + V \sum_{\ell=1}^{N_{\text{imp}}} \sum_{\sigma} n_{\ell\sigma},
\]

where \( n_{j\sigma} = c_j^{\dag} c_j^{\sigma}, (i,j) \) indicates the sum of 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 parameters typical of hole-doped cuprates, namely, \( t = \) use nearest neighbors \((=)\) for diagonal neighbors, and 0 otherwise. We apply a many-body variation theory to the above model. In computing expectation values, we use the data of the best wave function among what we checked (six types, not shown).

As a trial wave function, we use a Jastrow type: \( \Psi = \mathcal{P} \Phi \). Below, we use the data of the best wave function among what we checked (six types, not shown).

As a many-body factor, a form \( \mathcal{P} = \mathcal{P}_H \mathcal{P}_Q \mathcal{P}_G \) is used. In the onsite (Gutzwiller) factor \( \mathcal{P}_G \) [8] and the nearest-neighbor doublon-holon (D-H) binding factor \( \mathcal{P}_Q^\dagger \), we distinguish impurity and ordinary (non-impurity) sites as,

\[
\mathcal{P}_G = \prod_{j \neq \ell} \left[ 1 - (1 - g) n_{j\uparrow} n_{j\downarrow} \right] \prod_{\ell=1}^{N_{\text{imp}}} \left[ 1 - (1 - g_{\text{imp}}) n_{\ell\uparrow} n_{\ell\downarrow} \right], \tag{2}
\]

where \( g \) and \( g_{\text{imp}} \geq 0 \) are variational parameters, and \( \ell (j) \) runs over impurity (ordinary) sites. If the optimized value of \( g \) (or \( g_{\text{imp}} \)) is smaller \( \text{larger} \) than 1, the effective onsite interaction is repulsive \( \text{attractive} \). As a D-H factor [9, 10], which is relevant for Mott physics, we use

\[
\mathcal{P}_Q^\dagger = \prod_{j \neq \ell} \left( 1 - Q_j \right) \prod_{\ell=1}^{N_{\text{imp}}} \left\{ \left( 1 - \tilde{Q}_\ell \right) \prod_{\tau} \left[ 1 - (1 - \eta_\ell) h_{\ell\tau} d_{\ell+\tau} \right]\right\}, \tag{3}
\]
with
\[ Q_j = \zeta_\eta \, d_j \prod_{\tau'} (1 - h_{j+\tau'}) + \zeta_h \, h_j \prod_{\tau'} (1 - d_{j+\tau'}) \]  \tag{4}
\[ \tilde{Q}_\ell = \zeta_\ell \, d_\ell \prod_{\tau} (1 - h_{\ell+\tau}) \]  \tag{5}
where \( d_j = n_{\uparrow} n_{\downarrow}, \ h_i = (1 - n_{\uparrow})(1 - n_{\downarrow}) \), and impurity sites are excluded in the product of \( \tau' \) in Eq. (4). \( \eta, \ \zeta_\eta, \ \zeta_h \) and \( \zeta_\ell \) are variational parameters. We assume in constructing Eq. (3) that holons occupying impurity sites are isolated (doped) holes and do not compose D-H pairs. In this sense, a two-body Jastrow-type factor seems favorable for holons at impurity sites. Similarly, doublons in the ordinary sites are not assumed to combine with holons in the nearest-neighbor impurity sites. In addition, we take account of a nearest-neighbor repulsive correlation between holons (doublons), which are effective, especially, for high doping rates:
\[ P_H = \prod_{\langle i,j \rangle} \left[ 1 - (1 - \eta) (d_i d_j + h_i h_j) \right] \]  \tag{6}
where \( \langle i,j \rangle \) indicates a nearest-neighbor pair of sites, and \( \eta \) (0 \leq \eta \leq 1) is a variational parameter. In \( P_H \), we do not distinguish impurity and ordinary sites for simplicity.

For obtaining the one-body part \( \Phi \), we diagonalize the following mean-field Hamiltonian,
\[ H^{\sigma}_{\text{MF}} = H^\text{BR}_{\text{kin}} + U \sum_j \langle n_{j\sigma} \rangle n_{j-\sigma} + H^{\text{scr}}_{\text{V}}. \]  \tag{7}

In the kinetic part \( H^\text{BR}_{\text{kin}} \), a band-renormalization effect is introduced by following Ref. [2], as
\[ H^\text{BR}_{\text{kin}} = -t \sum_{\langle i,j \rangle} \left( c^\dagger_{i\sigma} c_{j\sigma} + \text{H.c.} \right) - \sum_{\langle i,j \rangle} t_\nu \left( c^\dagger_{i\sigma} c_{j\sigma} + \text{H.c.} \right), \]  \tag{8}
where the first term is the nearest-neighbor hopping and the second term indicates virtual hoppings up to fifth neighbors (see Fig. 1(b) in Ref. [2]). Here, \( t_\nu \)'s (\( \nu = 1\ldots4 \)) are variational parameters, which are crucial for stabilizing, especially, the AF state. In the second term of Eq. (7), we take account of an AF long-range order of the nesting vector \( \mathbf{Q} = (\pi, \pi) \) as
\[ \langle n_{j\sigma} \rangle = \frac{n}{2} + \delta_{\text{AF}} \text{sign}(\sigma) e^{i\mathbf{Q} \cdot \mathbf{r}_j}, \]  \tag{9}
where \( n = N/N_s \), \( \text{sign}(\sigma) = 1 \) or \( -1 \) according to \( \sigma = \uparrow \) or \( \downarrow \), and \( \delta_{\text{AF}} \) is a variational parameter that controls the magnitude of AF order. In the last term of Eq. (7), we consider screening effects of impurity potential \( V \) owing to electron correlation,
\[ H^{\text{scr}}_{\text{V}} = \sum_{\ell} \left( V_{\text{eff}} n_{\ell\sigma} + V_{\text{nn}} \sum_{\tau} n_{\ell\tau,\sigma} \right), \]  \tag{10}
where \( \tau \) runs over the nearest-neighbor sites of impurity site \( \ell \), and \( V_{\text{eff}} \) and \( V_{\text{nn}} \) are variational parameters. The first term \( (0 \leq V_{\text{eff}}/V \leq 1) \) represents a direct screening effect in the impurity sites; for \( V_{\text{eff}}/V = 0 \), the effect of impurity potential \( V \) is considered to be completely screened out by the electron correlation. By \( V_{\text{nn}} \), the electron density in the adjacent sites of impurity sites can be optimized. Diagonalizing \( H^{\sigma}_{\text{MF}} \) of \( N_s \times N_s \) matrices, we obtain eigenenergies \( \varepsilon_n \) and
eigenvectors $\phi_{n\sigma}$ with corresponding quasiparticle operators $\alpha^\dagger_{n\sigma}$, which are linear combinations of $c^\dagger_{j\sigma}$. Using $\alpha^\dagger_{n\sigma}$, one-body part is constructed as

$$\Phi = \prod_n (\varepsilon_n \leq \varepsilon_F) \prod_{\sigma} \alpha^\dagger_{n\sigma}|0\rangle,$$

where $\varepsilon_F$ indicates the energy of the highest occupied level (HOMO). The correlated AF state $\Psi_{\text{AF}} = \mathcal{P}\Phi_{\text{AF}}$ is reduced to a correlated paramagnetic (PM) state $\Psi_{\text{PM}} = \mathcal{P}\Phi_{\text{PM}}$ for $\delta_{\text{AF}} \to 0$.

We compute expectation values with respect to $\Psi$ using a VMC method similar to that in Ref. [2]. We use systems of $N_s = 10 \times 10$ sites with the periodic-antiperiodic boundary conditions, because the system-size dependence is small and only quantitative. A configuration of impurities $\{r_\ell\}$ is randomly chosen and fixed through a single sweep of VMC calculation (optimization and measurement). $\{r_\ell\}$ dependence exists to some extent and its degree depends on the model parameters. In a VMC sweep, we use 250,000 samples. In most cases, we will display data of 4 – 8 kinds of $\{r_\ell\}$ simultaneously.

3. Results

In the preceding report [4], we showed that properties of the uniform cases ($V = 0$) are preserved for $V_L \lesssim V \lesssim V_M$ in a strongly correlated regime ($U \gtrsim W$, $W = 8t$: band width), because the effect of impurity potential is almost completely screened out, namely $V_{\text{eff}}/V \sim 0$ and $V_{\text{un}}/V \sim 0$. Here, $V_L \sim -U/2$ and $V_M \sim t - 2t$. We concluded that the impurity potential is ineffective in weakly repulsive and moderately attractive cases for a large $U/t$.

![Figure 1](image.png)

Figure 1. Staggered magnetization as function of impurity potential for several doping rates. For each parameter set ($V$, $\delta$), data of four impurity configurations $\{r_\ell\}$ are plotted at the same time. The value at half filling for $V = 0$ ($m_0 \sim 0.88$) [2] is indicated by a cross and extended with a gray dash-dotted line. We also show $m_0(1 - \delta_{\text{imp}})$ for $\delta_{\text{imp}} = 0.08$ and 0.20 with dash-dotted lines of coordinate colors. A Mott transition point is indicated by a thick gray arrow. Incidentally, an AF order survives up to $\delta \sim 0.27$ for $V = 0$ [2].

Since Ref. [4] mainly treated $U/t$ dependence and cases of attractive potentials, here, we focus on cases of repulsive potential ($V > 0$) in the Mott-insulating regime of $U/t$ ($U > U_c \sim W$). In contrast with for $V_L \lesssim V \leq 0$, the value of $V_{\text{eff}}/V$ of the optimized AF state for $V > 0$ does not completely vanish (not shown). It is found that, when $V$ surpasses $V_M$ ($\sim t - 2t$), convergence in the VMC optimization suddenly becomes not easy and dependence on impurity configuration $\{r_\ell\}$ becomes serious, especially, for the PM state. Consequently, fluctuations in expectation values become large (sometimes too large to be managed). It is natural to consider that some transition arises at $V = V_M$ in the electronic state. Figure 1 shows $V$ dependence of the staggered magnetization (an AF order parameter)

$$m = \frac{2}{N_0} \left| \sum_j e^{iQ \cdot r_j} \langle S^z_j \rangle \right|,$$  

(12)
Figure 2. Average electron densities at impurity sites ($n_{\text{imp}}$, right axis) and ordinary sites ($n_{\text{ord}}$, left axis) as functions of impurity potential for several doping rates. As a reference, the data of $n_{\text{imp}}$ for $\delta = 0.08$ and $U = 0$ ($L = 10$ and 20) are also plotted with stars. A Mott transition point is indicated by a thick gray arrow.

Figure 3. (a) Schematic figure of real-space electron configuration in Mott insulating state realized for $V > V_{\text{M}}$. Steps at “Imp.” sites represent the repulsive impurity potential ($V > 0$). A blue circle represents an electron, and “H” indicates a holon (empty site). (b) Schematic figure for rough estimation of Mott-transition point $V_{\text{M}}/t$.

for several doping rates. We find, as $V/t$ increases, $m$ rapidly increases around $V = V_{\text{M}}$ and approaches $m_0(1 - \delta_{\text{imp}})$ for $V > V_{\text{M}}$, where $m_0$ ($\sim 0.88$) is the value of $m$ in the uniform case at half filling [2]. It follows that the staggered moment of the magnitude at half filling arises in the ordinary sites. Figure 2 shows $V/t$ dependence of the average electron densities on the impurity sites ($n_{\text{imp}}$) and the ordinary sites ($n_{\text{ord}}$) calculated as

$$n_{\text{imp}} = \frac{1}{N_{\text{imp}}} \sum_{\ell=1}^{N_{\text{imp}}} \sum_\sigma \langle n_{\ell\sigma} \rangle, \quad n_{\text{ord}} = \frac{N - N_{\text{imp}}n_{\text{imp}}}{N - N_{\text{imp}}},$$

respectively. The behavior of $n_{\text{imp}}$ is almost independent of $\delta$; as $V/t$ increases from $V/t = 0$, $n_{\text{imp}}$ rapidly decreases around $V = V_{\text{M}}$ and almost vanishes for $V > V_{\text{M}}$. Correspondingly, $n_{\text{ord}}$ rapidly increases and approaches 1, the value at half filling. In fact, the behavior in Figs. 1 and 2 reflects the relation $\delta_{\text{imp}} = \delta$, and can be interpreted as a filling-control-type Mott transition as we explain in the following.

The uniform AF state at half filling is insulating for any $U/t$ ($>0$) and its character changes from a Slater type to a Mott type as a crossover around $U = W$ [11]. The density of a doubly occupied site (doublon) is suppressed in the Mott-insulating regime, e.g. down to 0.027 at $U/t = 12$ [12]. In an impurity system of $V > V_{\text{M}}$, almost all doped holes are trapped in the impurity sites to avoid raising energy by $V$. When the condition $\delta_{\text{imp}} = \delta$ is imposed, empty sites (holons) have one-to-one correspondence to impurity sites, and (almost) all ordinary sites are occupied by a single electron as shown in Fig. 3(a), namely, a half-filled band is virtually
Figure 4. Momentum distribution function of type-II AF state for (a) $\delta = 0.08$ and (b) $\delta = 0.20$ along path $(0, 0) \rightarrow (\pi, 0) \rightarrow (\pi, \pi) \rightarrow (0, 0)$. In each panel, evolution of $n(k)$ as $V/t$ changes is to be noticed. For each value of $V$, the result of a single $\{R_t\}$ is plotted. For comparison, the data for two typical reference values, a uniform AF Mott-insulating state at half filling (black open circles) and a uniform PM state (gray stars) are also drawn. The values of $U/t$, $t'/t$, and $L$ are common to all cases.

formed within the ordinary sites. In other words, some sites of the uniform half-filled lattice are depleted by the impurities, but the residual $N_e(1 - \delta_{\text{imp}})$ sites preserve the properties of the uniform half-filled band. Thus, a Mott-insulating state is realized at a partially filling by impurity potential.

Next, let us discuss why the Mott transition point is situated at $V_M/t \sim 1 - 2$ regardless of $U/t$ and $t'/t$ (data not shown). The variation of energy when an electron hops to an impurity site or hops to an impurity site and return to the original site, as shown in Fig. 3(b), is roughly estimated at $\Delta \varepsilon = V - t$ or $V - 2t$. For $\Delta \varepsilon < 0$ ($\Delta \varepsilon > 0$), an electron hops via an impurity site without (at) a loss of energy. In the case of $\delta_{\text{imp}} = \delta$ with large $U$ ($\gg t$), a global flow becomes possible only if electrons can hop to impurity sites. As a result, the state becomes conductive or insulating according as $\Delta \varepsilon$ is negative or positive, resulting in $V_M/t = 1 - 2$.

Finally, we corroborate this Mott transition by the momentum distribution function,

$$n(k) = \frac{1}{2} \sum_{\sigma} \langle \epsilon_{k\sigma} c_{k\sigma} \rangle. \quad (14)$$

Showing in Figs. 4(a) and 4(b) is evolution of $n(k)$ as $V/t$ changes for an underdoped ($\delta = 0.08$) and an overdoped ($\delta = 0.20$) cases, respectively. As discussed in Ref. [2], the uniform AF state for $t'/t = -0.3$ is classified as type II, which is insulating at half filling but becomes metallic for $\delta > 0$ with a pocket Fermi surface (FS) around $k = (\pi/2, \pi/2)$ and a gap near antinodal $k = (\pi, 0)$. Such metallic behavior—discontinuity with a dip near $(\pi/2, \pi/2)$ and smoothness near $(\pi, 0)$—can be confirmed by the curves for $V = 0$ in Fig. 4, and is preserved for $V \lesssim V_M$ (drawn with warm colors). For $V \gtrsim V_M$ (cold colors), however, the pocket FS near $(\pi/2, \pi/2)$ vanishes, and the behavior of $n(k)$ becomes similar to that of the uniform AF state at half filling. It indicates a Mott transition.
4. Conclusion
We studied effects of point-type repulsive impurity potential on a strongly correlated AF state, using a many-body variation theory. We showed that a filling-control-type Mott transition takes place in partially-filled AF states at $V = V_M \sim 2t$, if the densities of impurity is equal to the doping rate. We have found that such Mott transitions are not restricted to the present special case ($\delta_{\text{imp}} = \delta$) but manifest themselves widely ($\delta_{\text{imp}} \geq \delta$ and in attractive-potential cases). We will soon report them elsewhere. It is important to consider how the Mott transition discussed here is related to the behavior of cuprates, or more generally to phenomena in other materials.

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