Insulator-to-superconductor transition by exciting half-filled $d$-wave pairing state beyond Mott gap

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Abstract. In connection with recent tera-Hertz photoexcitation experiments in which strongly correlated materials are changed from Mott-insulating to metallic, we statically study the properties of excited states beyond the Mott gap in the $d_{x^2-y^2}$-wave superconducting ($d$-SC) branch at half filling, by applying a variational Monte Carlo method to a two-dimensional Hubbard model. It is found that the first-order Mott transition previously found at $U = U_c$ for the lowest-energy state vanishes for the excited states. As a result, an insulator-to-$d$-SC transition occurs as the strength of excitation (corresponding to light intensity) is increased for $U/t$ ($> U_c/t$) fixed. As a mechanism, this conduction is led by newly created free carriers (unbound doublons and holons) in the excited states.

1. Introduction

Recently, many researchers have taken notice of insulator-to-conductor transitions found in various Mott insulators induced by tera-Hertz photoexcitation [1] and by applying pulse electric fields [2], in connection with the application to high-speed switching devices. In the former experiments [1], parent (nondoped insulating) compounds of cuprate superconductors (SCs) were compared between electron-doped (Nd$_2$CuO$_4$) and hole-doped (La$_2$CuO$_4$) cases. As a result relevant to the preset study, it was found that Nd$_2$CuO$_4$, which has a smaller Mott (charge transfer) gap (1.65 eV) than La$_2$CuO$_4$ (2.2 eV), needs weaker light intensity (0.027 photons/Cu site) than the latter (0.055 photons/Cu site) for metallization.

So far, most related theoretical studies had shed light on the dynamical properties after such excitation [3]. The state after excitation develops or relaxes with ultra-high speeds, but it is significant from a basic point of view to treat the state as a quasi-stationary state, as previously studied using $t$-$J$-type models [4]. It is not trivial whether the “fresh” (right after pumping) excited state remains insulating or has been changed to conductive. In this study, we assume that the state excited beyond the Mott gap is stationary for sufficiently long time for electronic processes. This assumption is not necessarily irrelevant, because relaxation processes through phonons and light emission have much longer time scales. Here, we consider these excited state in the light of variation theory. Into the trial excited state, we intentionally introduce additional doublons (doubly occupied sites, abbreviated as D) and holons (empty sites, as H) by prohibiting the total number of doublons ($D$) from being smaller than our set lower bound $D_L$ ($D \geq D_L$). This operation is roughly corresponds to creating $D_L$ additional doublons, in relation to the
above photoexcitation experiments [1]. As a pilot study, here we concentrate on a \(d_{x^2-y^2}\)-wave superconducting (d-SC) state \(\Psi_d\); because \(\Psi_d\) brings about a Mott transition at \(U = U_c \sim 6.6t\) [5], we will be able to clarify how the Mott transition evolves as \(D_L\) increases.

2. Formalism

In this study, we consider a single-band Hubbard model \((U \geq 0)\) on a square lattice with diagonal transfer:

\[
\mathcal{H} = \mathcal{H}_{\text{kin}} + \mathcal{H}_U = - \sum_{(i,j),\sigma} t_{ij} \left( \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \text{H.c.} \right) + U \sum_j n_{j\uparrow} n_{j\downarrow},
\]

where \(n_{j\sigma} = \hat{c}_{j\sigma}^\dagger \hat{c}_{j\sigma}\) and \((i,j)\) indicates the pairs on sites \(i\) and \(j\). We set the hopping integral \(t_{ij} = t\), \(t'\) for diagonal (next-nearest) 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.

In this proceedings, we concentrate on the cases at half filling \((n = N/N_0 = 1\) or \(\delta = |1-n| = 0, N: \text{number of electrons}, N_0: \text{number of sites})\). The value of \(t'/t\) is fixed at \(-0.3\). For the systems into which the band renormalization effect (see below) is introduced, it is known \((\text{for } \delta = 0)\) that the lowest-energy state \((D_L = 0)\) in each branch \([\text{d-SC, AF and normal (paramagnetic)}\) states\] does not depend on the value of \(t'/t\) at least for \(-0.5 \leq t'/t \leq 0.5\) and \(U > U_c [6, 7]\). However, such a feature is nontrivial for the excited states \((D_L > 0)\); we leave \(t'/t\) dependence for a future problem.

We study this model using a variational Monte Carlo (VMC) method [8, 9, 7], which enables us to exactly treat many-body wave functions. To construct trial excited states in which doublon-holon pairs are induced, we extend the previously used Jastrow form of the lowest-energy \(d\text{-SC state}\) \(\Psi = \mathcal{P} \Phi_d = \mathcal{P}_G \mathcal{P}_Q \Phi_d [7]\) to

\[
\Psi_{D_L} = \mathcal{P}_{D_L} \Psi,
\]

where \(\mathcal{P}_{D_L}\) is a projector that imposes the condition \(D \geq D_L\), as explained below.

First, we review the lowest-energy part \(\Psi\). \(\mathcal{P}_G\) is the celebrated onsite \((\text{Gutzwiller})\) projector with a parameter \(g [10]\), \(\mathcal{P}_Q\) is a nearest-neighbor doublon-holon \((\text{D-H})\) binding factor [11, 12] vital for Mott physics: \(\mathcal{P}_Q = \prod_j (1 - Q_j),\) where

\[
Q_j = \zeta_d d_j \prod_{\tau} (1 - h_{j+\tau}) + \zeta_h h_j \prod_{\tau} (1 - d_{j+\tau}),
\]

\(d_j = n_{j\uparrow} n_{j\downarrow}, h_j = (1 - n_{j\uparrow})(1 - n_{j\downarrow}), \zeta_d\) and \(\zeta_h\) are D-H binding parameters, and \(\tau\) runs over all the nearest-neighbor sites of site \(j\). At half filling, a relation \(\zeta_d = \zeta_h\) holds in the D-H parameters owing to the electron-hole symmetry. For the one-body state, we employ an \(N\)-electron \(d_{x^2-y^2}\)-wave BCS wave function,

\[
\Phi_d = \left( \sum_k a_k \hat{c}_{k\uparrow} \hat{c}_{-k\downarrow} \right)^{N/2} |0\rangle,
\]

where \(a_k = \frac{\varepsilon_k}{u_k} \frac{\Delta_k}{\varepsilon_k - \mu + \sqrt{\left(\varepsilon_k - \mu\right)^2 + \Delta^2_k}}\),

with \(\Delta_k = \Delta_d (\cos k_x - \cos k_y)\). In \(\Phi_d\), band renormalization effects are introduced by optimizing a tight-binding band up to fifth neighbors [7]: \(\varepsilon_k = \gamma_k + \varepsilon_1(k) + \varepsilon_2(k) + \varepsilon_3(k) + \varepsilon_4(k)\), with \(\gamma_k = -2t(\cos k_x + \cos k_y), \varepsilon_1(k) = -4t_2(\cos k_x \cos k_y), \varepsilon_2(k) = -2t_2(\cos 2k_x + \cos 2k_y), \varepsilon_3(k) = -4t_3(2\cos 2k_x \cos 2k_y + \cos k_x \cos 3k_y),\) and \(\varepsilon_4(k) = -2t_4(3\cos 3k_x + \cos 3k_y)\). Because the parameters in \(\Phi_d (\Delta_d, \mu, t_1-t_4)\) have considerable redundancy, they are not optimized at unique values; however, the correlation parameters \((g\) and \(\zeta_d)\) as well as the minimal energy and corresponding physical quantities are uniquely determined.
Now, we explain the projector $\mathcal{P}_{D_L}$ in Eq. (2) for D-H excitations. It is known that the doublon density $d = D/N_s$ ($D$: total number of doublons) in the lowest-energy state $\Psi$ is finite even in the Mott-insulating regime ($6.6t < U_c < U < \infty$): $d = 0.032$ (0.020) for $U/t = 8$ (12) [12]. In this regime, however, the doublons are tightly bound to the counter holons [13]; therefore, there is no free charge carrier. In the limit of $U/t \to \infty$, doublons and holons completely vanish. By applying $\mathcal{P}_{D_L}$, we force this state to have at least $D_L$ doublons, in other words, $\mathcal{P}_{D_L}$ restricts the space of $\Psi$ to $D \geq D_L$. Therefore, $\Psi_{D_L=0}$ indicates the original $\Psi$, and $\Psi_{D_L}$ for $U/t = \infty$ is the state in which correctly $D_L$ doublons (and holons) are excited. To treat different system sizes, it is substantial to use the lowest doublon density $d_L = D/L$ instead of $D_L$.

We compute expectation values with respect to $\Psi_{d_L}$ using a VMC method similar to that in Ref. [7]. $\mathcal{P}_{D_L}$ can be easily treated in this method. We use systems of $N_s = L \times L$ sites of $L = 10$ and 12 with the periodic-antiperiodic boundary conditions. In an optimization sweep, we iterate typically 160 times for each parameter with $5 \times 10^4$ samples. To search for the global minimum within $\Psi_{d_L}$, we performed 20 sweeps at most in the cases of $U \sim U_c$.

3. Results
As shown in a previous study [5], $\Psi$ exhibits a Mott transition at $U = U_c \sim 6.6t$. We start with the confirmation of this transition. In Fig. 1(a), we show the doublon density $d = D/N_s$ for several values of $d_L$ as a function of $U/t$. For the lowest-energy state ($d_L = 0$), $d$ exhibits a virtual discontinuity at $U = U_c \sim 6.6t$; $d$ still has finite small values for $U > U_c$, and decreases slowly as $U/t$ increases. The remaining of $d$ at a finite value is caused by bound D-H pairs [5]. For $d_L > 0$, $d$ seems to increase semi-regularly as $d_L$ increases. In Fig. 1(b), we show the difference of $d$ between the states of $d_L$ and the lowest-energy state ($d_L = 0$). We find that the restriction as to $d_L$ roughly results in increase of $d$ by $d_L$, especially correctly, for large $d_L$ and/or $U/t$. Because this quantity converges at $d_L$ for $U/t \to \infty$, the state of $U/t = 20$ is regarded as sufficiently strongly correlated. On the other hand for $U < U_c$, $d$ is almost independent of $d_L$ in the present range ($d_L < 0.05$) owing to $d \gg d_L$, indicating that moderate strength of D-H excitation ($d_L < 0.05$) is ineffective in a weakly correlated conductive regime.

To consider the conductivity for $U > U_c$ in the $d$-SC state, we introduce a notion of “free doublon”. For $d_L = 0$ and $U > U_c$, almost all doublons are paired with holons (D-H bound pairs), and there is no free charge carrier (unpaired doublon or holon). For $d_L > 0$, however, such

![Figure 1](image-url)
an unpaired doublon may appear, which we call a free doublon. In the present wave function, a doublon without a holon(s) in the nearest-neighbor sites is regarded as a free doublon, because the present D-H binding factor \( P_Q \) is restrict to the nearest-neighbor sites. Henceforth, the total number (density) of free doublons is denoted by \( D_F \) (\( D_F = D_F/N_s \)). In Fig. 2(a), we show \( U/t \) dependence of \( d_F \) for various values of \( d_L \). For \( d_L = 0 \), \( d_F \) becomes substantially zero for \( U > U_c \) [14]. Namely, almost all doublons [see Fig. 1(a)] exist as D-H pairs. As \( d_L \) is increased, free doublons come to appear; for small \( d_L \) (\( \leq 0.02 \)), \( d_F \) almost vanishes at some values of \( U/t < 20 \), whereas for \( d_L \geq 0.03 \), \( d_F \) remains finite even for \( U/t = 20 \) [15]. It suggests that the state becomes conductive, if some additional doublons (and holons) are created in excitation.

Next, we confirm that these free doublons actually contribute to conductivity. We analyze the kinetic energy \( \langle \mathcal{H}_{\text{kin}} \rangle = E_{\text{kin}} \) into two contributions of electron hopping that changes \( D \) \( \langle E_{\text{kin}}^{(L)} \rangle \) and preserves \( D \) \( \langle E_{\text{kin}}^{(F)} \rangle \). The former corresponds to the local process in which a D-H pair is created or destroyed and does not contribute to conductivity, whereas the latter corresponds to the global motion of free doublons or holons. In Fig. 2(b), the free part \( E_{\text{kin}}^{(F)} \) is shown as a function of \( U/t \). One may notice that the behavior of \( |E_{\text{kin}}^{(F)}| \) is quite similar to that of free-doublon density [Fig. 2(a)], indicating that free doublons directly contributes to the electronic motion. In Fig. 3(a), we show \( d_F \) and \( |E_{\text{kin}}^{(F)}| \) as functions of \( d_L \) for \( U/t = 8 \) and 12 (\( > U_c/t \)). A threshold value of \( d_L \) (excitation strength) seems to exist beyond which the state becomes conductive, and the threshold increases as \( U/t \) increases. This tendency is consistent with the experiments in hole-doped and electron-doped cuprate superconductors mentioned in Sect. 1.

Now, we discuss aspects of superconductivity using the \( d_{x^2-y^2} \)-wave long-distance pairing correlation function \( P_d \) (for definition of \( P_d \), see Ref. [12]), which measures the strength of d-SC. Figure 3(b) shows the \( U/t \) dependence of \( P_d \) for various values of \( d_L \). For \( d_L = 0 \), as discussed in Ref. [12], \( P_d \) exhibits a sharp peak immediately under \( U_c/t \), but suddenly vanishes in the Mott-insulating regime \( (U > U_c) \), where charge fluctuation is strongly suppressed. For \( d_L > 0 \), \( P_d \) has a peak with comparable height at the same \( U/t \), but exhibits a robust tail in the Mott-insulating regime, corresponding to the behavior of \( d_F \) and \( E_{\text{kin}} \). Considering that the carrier density is roughly \( 2d_F \) (free doublons+holons) here, we notice that the behavior of \( P_d \) is similar to that in (chemically) doped cases somewhat quantitatively (compare with Fig. 9 in Ref. [12]).

Finally, we check that the above argument is consistent with the behavior of momentum...
Figure 3. (a) Free-doublon density (left axis) and absolute value of free part of kinetic energy (right axis) as functions of $d_L$ for two values of $U/t$ in Mott insulating regime. (b) $U/t$ dependence of $d$-wave superconducting correlation function for the same values of $d_L$ and $L$ as in Fig. 1 (a). The appreciable values of $P_d$ for $U/t \lesssim 5$ are spurious owing to system-size dependence characteristic of weakly correlated cases [12]. The Mott transition point for $d_L = 0$ is indicated by a red arrow on the lower axis.

Figure 4. Momentum distribution function for various values of $U/t$ along path of $k$, $(0, 0) - (\pi, 0) - (\pi, \pi) - (0, 0)$ for (a) $d_L = 0.0$ and (b) $d_L = 0.0417$.

distribution function:

$$n(k) = \frac{1}{2} \sum_\sigma \langle c_{k\sigma}^\dagger c_{k\sigma} \rangle,$$

and charge density structure factor:

$$N(q) = \frac{1}{N_s} \sum_{i,j} e^{i\mathbf{q}(\mathbf{R}_i-\mathbf{R}_j)} \langle n_i n_j \rangle - n^2.$$

Figure 4 depicts the evolution of $n(k)$ as $U/t$ is varied. In the case of $d_L = 0$ [panel (a)], $n(k)$ for $U/t = 5$ exhibits almost normal metallic behavior, in which a Fermi surface (FS) (discontinuity) is clear both at near the antinodal point $(\pi, 0)$ and near $(\pi/2, \pi/2)$ in the nodal direction $(0, 0)$ - $(\pi, \pi)$. For $5t < U < U_c \sim 6.6t$, FS near $(\pi, 0)$ vanishes owing to the SC gap formation but FS in the nodal direction is preserved. This behavior is characteristic of the $d_{x^2-y^2}$-wave SC. For $U > U_c$, FS in both points vanishes and $n(k)$ becomes smooth for any $k$, suggesting a Mott...
maximum value of behavior of increases. (3) This conduction is led by the free carriers (unbound doublons and holons). The weak and intermediate strength (\(d_{\text{weakly correlated (conductive) regime}}\) \(U < U_c\)) preserves for \(U > U_c\) always shows the feature of \(d\)-SC mentioned above. Thus, the behavior of \(n(k)\) is found to be consistent with the above argument.

Similarly, in Figs. 5 (a) and (b), we show the evolution of \(N(q)\) as \(U/t\) increases for \(d_L = 0\) and \(d_L \sim 0.04\), respectively. According to the single-mode approximation [16], the state is gapless (gapped) in the charge sector, if \(N(q)\) is linear (quadratic) for \(|q| \to 0\). For \(d_L = 0\), \(N(q)\) is linear (likely quadratic) for small \(|q|\) for \(U < U_c\) (\(U > U_c\)), which behavior is consistent with a Mott transition \([5]\). On the other hand for \(d_L = 0.0417\), \(N(q)\) for small \(|q|\) is linear for any \(U/t\), indicating \(\Psi_{0.0417}\) is always gapless in the charge sector, and then conductive \([17]\).

4. Summary and discussions

With in mind the high-energy quasi-stationary states induced by photoexcitation or tera-Hertz pulse electric field, we studied excited states exceeding the Mott gap in the \(d_{z^2}-y^2\)-wave superconducting branch for the half-filled-band Hubbard model, by imposing the minimum doublon densities \(d_L (\lesssim 0.04)\) on the variational wave functions. It is found that (1) the first-order Mott transition arising at \(U = U_c\) for \(d_L = 0\) \([5]\) vanishes for \(d_L > 0\), and the state comes to approach an insulator asymptotically as \(U/t\) increases. The coherence of the state is preserved for \(U > U_c\). (2) Therefore, an insulator-to-\(d\)-SC transition occurs as \(d_L\) is increased. The transition value of \(d_L\), which corresponds to the required light intensity, increases, as \(U/t\) increases. (3) This conduction is led by the free carriers (unbound doublons and holons). The behavior of \(d\)-SC correlation function \(P_d\) resembles those of (chemically) doped cases. (4) The maximum value of \(P_d\) just below \(U = U_c\) for \(d_L = 0\) is almost unchanging for \(d_L > 0\). (5) In a weakly correlated (conductive) regime \((U < U_c)\), excitation by D-H-pair formation at least of weak and intermediate strength (\(d_L < 0.05\)) is ineffective.

These results are qualitatively consistent with the experiments of the parent compounds of cuprate SCs \([1]\). Actually, however, these compounds are antiferromagnetic (AF) Mott insulators. In fact, calculations for an AF case are now in progress. According to the preliminary results, the energy of the AF state is sufficiently lower than that of the \(d\)-SC state also for \(d_L > 0\). Thus, the properties of the AF state will be more relevant to the comparison with the experiments.
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References
[1] Okamoto H et al. 2010 Phys. Rev. B 82 060513(R); 2011 Phys. Rev. B 83 125102
[2] Yamakawa H et al. 2017 Nat. Mater. 16 1100
[3] For instance, Aoki H, Tsuji N, Eckstein M, Kollar M, Oka T and Werner P 2014 Rev. Mod. Phys. 86 779
[4] For instance, Takahashi A, Gomi H and Aihara M 2002 Phys. Rev. B 66 151103
[5] Yokoyama H, Ogata M and Tanaka Y 2006 J. Phys. Soc. Jpn. 75 114706
[6] Watanabe T, Yokoyama H, Tanaka Y and Inoue J 2006 J. Phys. Soc. Jpn. 75 074707
[7] Sato R and Yokoyama H 2016 J. Phys. Soc. Jpn. 85 074701
[8] Yokoyama H and Shiba H 1987 J. Phys. Soc. Jpn. 56 1490
[9] Umrigar C J, Wilson K G and Wilkins J W 1988 Phys. Rev. Lett. 60 1719
[10] Gutzwiller M C 1963 Phys. Rev. Lett. 10 159
[11] Yokoyama H and Shiba H 1990 J. Phys. Soc. Jpn. 59 3669
[12] Yokoyama H, Ogata M, Tanaka Y, Kobayashi K and Tsuchiura H 2013 J. Phys. Soc. Jpn. 82 014707
[13] For instance, Miyagawa T and Yokoyama H 1990 J. Phys. Soc. Jpn. 80 084705
[14] In Fig. 2(a), $d_F$ for $d_L = 0$ is finite for the values of $U$ slightly larger than $U_c$ for $L = 10$. For $L \rightarrow \infty$, however, $d_F$ is expected to completely vanish for $U > U_c$ within the present $P_Q$.
[15] The reason why free doublons appear and increase as $d_L$ increases is considered as follows. When the lowest doublon number $D_L$ is imposed, it becomes difficult as $D$ approaches $D_L$ (along with the increase of $U/t$) that the total energy is reduced by the process of creating and, especially, destroying D-H pairs, namely, by $E_{\text{kin}}^{(L)}$. Therefore, to reduce the total energy, it becomes advantageous that D-H pairs are resolved and $E_{\text{kin}}^{(F)}$ is reduced by hopping of free doublons and holons.
[16] See for instance, Auerbach A 1994 Interacting Electrons and Quantum Magnetism (New York: Springer)
[17] We also checked the gap behavior in the spin sector similarly using $S(q)$ (figures not shown). Both for $d_L = 0$ and $\sim 0.04$, spin gap opens for $U/t > 5$, where the state is either in robust $d$-SC or Mott insulating as identified above. This feature of the spin gap is consistent with the other results.