



A simple metal–insulator criterion for the doped Mott–Hubbard materials

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ABSTRACT

A simple metal–insulator criterion for doped Mott–Hubbard materials has been derived. Its readings are closely related to the orbital and spin nature of the ground states of the unit cell. The available criterion readings (metal or insulator) in the paramagnetic phase reveal the possibility of the insulator state of doped materials with the forbidden first removal electron states. According to its physical meaning, the result is similar to the Wilson's criterion in itinerant electron systems. The application of the criterion to high- T_c cuprates is discussed.

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1. Introduction

It is known that some of perspective doped transition-metal oxides: cuprates [1,2] and manganites [3] show mysterious pseudogap states. It is surprising that the pseudogap states in such different oxides have common features [4,5]. Extensive dielectric regions on the phase T/χ -diagram of manganites [6,7] also attract attention (primarily in the paramagnetic phase(PM)). If the reasons for the insulator states in these different doped oxides are general by nature, one can try to detect them directly following the quasiparticle picture established by Hubbard [8].

The purpose of our work is to construct a metal–insulator criterion based on Wilson's ideas [9] concerning a system of itinerant electrons in the analytical form for the doped Mott–Hubbard materials. Indeed, this approach includes the statement that if the electron system consists of completely occupied and empty bands, it is an insulator, otherwise, it is a metal. Here, there are some features. Due to many-electron effects the spectral density of quasiparticle states in the Mott–Hubbard materials depends on the carrier concentration. Secondly, it does not make much sense to apply the Wilson's criterion to the doped materials because of the fluctuations of the impurity potential which create new states. However, it is known that because of the similarity of the phase T/χ diagrams of the high- T_c cuprates, with the carriers having different origins, the carrier concentration is a crucial factor. Consideration is given to the criterion taking into account the many-electron effects only.

2. Method

The generalized approach uses the fact that the optical intracell transitions with their (l -orbital, S -spin)-selection rules in the transparency window and optical charge transfer transitions in the oxides can be observed at the same d -states [10,11]. In the first approximation one can assume that the quasiparticles are unit cell excitations which can be represented graphically as single-particle transitions between different sectors $N_h = \dots(N_{h0}-1), N_{h0}, (N_{h0}+1), \dots$ of the configuration space of the unit cell (N_{h0} -hole number per cell in the undoped material, see Fig. 1) [12]. Each of these transition forms a r -th quasiparticle band, where the root vector $r = ii'$ in the configuration space numerates the initial i and final i' many-electron states in the transition. The transitions, with the number of electrons increasing or decreasing, form the conduction or valence bands, respectively.

For our purposes it is convenient to start with Lehmann's representation for the Green's function $G_{fg\sigma}^{\lambda\lambda}$ of the intracell Hamiltonian H_0 with respect to the family of single-particle operators $c_{f\lambda\sigma}^{(+)}$ and their matrix elements in the basis of $|(N_h, M_S)_i\rangle$ -eigenstates of H_0 (S and M are the spin and spin projection of the multi-electron cell eigenstate):

$$\begin{aligned} G_{fg\sigma}^{\lambda\lambda} &= \langle\langle c_{f\lambda\sigma} | c_{g\lambda\sigma}^+ \rangle\rangle = \sum_{rr'} \gamma_{f\lambda\sigma}(r) \gamma_{g\lambda\sigma}(r') D_{0fg}^{rr'}(E) \\ &= \delta_{fg} \sum_{rr'} \delta_{rr'} \frac{\gamma_{\lambda\sigma}^2(r) F_r(x)}{E - \Omega_r}, \end{aligned} \quad (1)$$

where matrix elements

$$\begin{aligned} \gamma_{\lambda\sigma}(r) &= \langle(N_h + 1, M'_S)_\tau | c_{f\lambda\sigma} | (N_h, M_S)_\mu \rangle \\ &\times \delta(S', S \pm |\sigma|) \delta(M', M + \sigma), \end{aligned} \quad (2)$$

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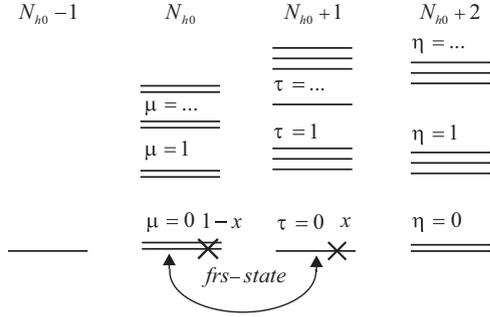


Fig. 1. (Colour Online) $E_i(N_h, M_S)$ – energy level scheme of the Hilbert space based on cell states (7) and (8) with hole numbers per cell $N_h = N_{h0} - 1, N_{h0}, N_{h0} + 1, \dots$, where $i = \mu, \tau, \eta$ and N_{h0} is a hole number per cell in the undoped material. The cross indicates the occupied ground cell states of hole doped material. A solid line with arrows corresponds to the first removal electron states with a lowest binding energy.

the total space of the root vectors $\{r\} = \dots + \{r_{12}\} + \{r_{23}\} + \dots, (\{r_{12}\} = \{\mu\tau\}, \{r_{23}\} = \{\tau\eta\})$ and so on, Fig. 1). An occupation factor $F_r(x)$ is the probability to detect a cell in any of the i, i' states participating in the r -th transition, and $\Omega_r = E_i(N_h, M_S) - E_{i'}(N_h + 1, M_{S'})$ is a quasiparticle energy in the r -th band. For example, in the PM phase of the doped material the occupation factor has the form:

$$F_{r_{12}}(x) = \frac{1 - \alpha x}{2S + 1}, \quad (3)$$

where $\alpha = 1 - (2S + 1)/(2S' + 1)$ is proportional to the ratio of the spin multiplets of the i, i' states participating in the r_{12} - (from the subspace $\{r_{12}\}$) transition between the ground states $|(N_{h0}, M_S)_{i=0}\rangle$ and $|(N_{h0} + 1, M_{S'})_{i'=0}\rangle$ indicated by the arrow in Fig. 1.

The Green's function in its simplest form (1) is quite insufficient to study the \vec{k} -physics of quasiparticles (e.g., superconductivity properties of high- T_c cuprates). However, this approach is free from the shortcomings of the hydrogen-like (s -)representation and low-energy approximations, because we do not restrict ourselves to choosing the intracell Hamiltonian H_0 , and we are ready to work with all $|(N_h, M_S)_i\rangle$ states in the framework of the Russell-Saunders scheme. The total number of the valence states is equal to the sum over all the quasiparticle valence states:

$$N_V(x) = \sum_{\lambda\sigma} \sum_r \gamma_{\lambda\sigma}^2(r) \int dE \left(-\frac{1}{\pi} \right) \text{Im} D_0^r(E)_{E+i0} = N_V^{12}(x) + N_V^{23}(x), \quad (4)$$

where $N_V^{12}(x)$ and $N_V^{23}(x)$ are the contributions from the quasiparticles with the root vectors r from the $\{r_{12}\}$ and $\{r_{23}\}$ subspaces because the other states of $|(N_h, M_S)_i\rangle$ in the doped material are not occupied, and there is a zero probability $F_r(x) = 0$ to detect a cell in these states at a low temperature. The key condition at the insulating state, which we are interested in, is

$$N_e - x = N_V(x), \quad (5)$$

where $(N_e - x)$ is the total electron number per cell of the hole doped material. That is, if the number of electrons in a cell equals to the number of the valence states, the material is an insulator.

To obtain the Fermi level position in the degenerate doped material at zero temperature one could carry out the integration on the right side of the equation:

$$x = \sum_{\lambda\sigma} \sum_r \gamma_{\lambda\sigma}^2(r) \int_{E_F} dE \left(-\frac{1}{\pi} \right) \text{Im} D_0^r(E)_{E+i0}, \quad (6)$$

over the top valence band of the first removal electron states (*frs*) with the lowest binding energy (see Fig. 1), and this is sufficient at the actual concentrations $x \sim 0.1$, as a rule. However, this is not sufficient, when the hole concentration x exceeds the number of quasiparticle states in the top valence band $x \gg N_{frs}$. And this is not due to the hole concentration being too large, but because the number of *frs* quasiparticle states N_{frs} may be very small, which is

to be seen later. It is not obvious that the Fermi level with doping will be immersed into the following energy valence band in the doped Mott-Hubbard material. To understand the features of the solutions of Eq. (6) in the doped materials with the forbidden *frs* quasiparticle states (i.e. $N_{frs} = 0$), one derives the total number of the valence states in (5) as a function $N_V(x, N_{frs})$.

By following this approach, one obtains a simple metal-insulator criterion, which is characterized by the condition: $N_{frs} = 0$ (-insulator) or $N_{frs} \neq 0$ (-metal) irrespective of the hole concentration x . Consideration is given to the case with one hole per cell $N_{h0} = 1$ in the undoped materials and an arbitrary number N_λ of the occupied λ orbitals, i.e. $N_e = 2N_\lambda - 1$. This is relevant for the high- T_c cuprates. In this case of one hole per cell, the $|(N_h, M_S)_i\rangle$ cell states are a superposition of different hole configurations of the same orbital l -symmetry:

$$|(N_{h0}, M_S)_\mu\rangle = \sum_\lambda \beta_\mu(h_\lambda) |h_\lambda, M_S\rangle \quad (7)$$

Thus, there are one-hole spin doublet states, $C_{2N_\lambda}^1 = 2N_\lambda$, where $C_{2N_\lambda}^k$ is the number of combinations. Besides, there are $C_{2N_\lambda}^2 = N_S + 3N_T$ of the spin singlets $N_S = C_{N_\lambda}^2 + N_\lambda$ and triplets $N_T = C_{N_\lambda}^3$:

$$|(N_{h0} + 1, M_{S'})_\tau\rangle = \sum_{\nu\nu'} B_\tau(h_\nu, h_{\nu'}) |h_\nu, h_{\nu'}, M_{S'}\rangle \quad (8)$$

in the two-hole sector (Fig. 1) in the N_λ -orbital approach. Using the intracell Hamiltonian H_0 in the cell function representation the configuration weights $\beta_\mu(h_\lambda)$ and $B_\tau(h_\lambda, h_{\lambda'})$ can be obtained by the exact diagonalization procedure for the matrices $(\hat{H}_0)_{\lambda\lambda'}$ and $(\hat{H}_0)_{\lambda\lambda'}^{\nu\nu'}$ in the $E_i(N_h, M_S)$ -eigenvalue problem in different sectors N_h . [12] The sum (4) over all the r -th excited states with $\mu \neq 0$ in the sector $N_h = N_{h0}$ is omitted, and only the excited states with any $\tau(\eta)$ index in the nearest $N_h = (N_{h0} + 1)$ and $(N_{h0} + 2)$ sectors are summed up. To calculate the matrix elements (2) the eigenfunctions (7) and (8) in different sectors must be expressed through each other. It is possible due to the rules for the addition of the angular momenta [13]. The expressions for high- and low-spin two-hole partners (with $S' = S \pm |\sigma|$) can be combined into a single expression:

$$|h_\lambda, h_{\lambda'}, M_{S'}\rangle = \Gamma_\uparrow(S'_M, S) c_{\lambda'\downarrow} \left| h_\lambda, M' - \frac{1}{2} \right\rangle + \text{sgn}(\Delta S) \Gamma_\downarrow(S'_M, S) c_{\lambda'\uparrow} \left| h_\lambda, M' + \frac{1}{2} \right\rangle \quad (9)$$

where $\Delta S = S' - S = \pm |\sigma|$, and the coefficients

$$\Gamma_\sigma^2(S'_M, S) = \frac{S + \eta(\sigma) \text{sgn}(\Delta S) M' + \frac{1}{2}}{2S + 1} \quad (10)$$

have a completeness property for the contributions from the identical spin states of a doped hole to different high- and low-spin two-hole partners:

$$\sum_{\Delta S = -|\sigma|}^{+|\sigma|} \Gamma_\sigma^2(S'_M, S) = \sum_\sigma \Gamma_\sigma^2(S'_M, S) = 1, \quad (11)$$

and also

$$\sum_{M=-S}^S \Gamma_\sigma^2(S'_M, S) = S + \frac{1}{2} \quad (12)$$

Taking into account relations (7), (8) and (12) one can determine the matrix element in (4) by the sum:

$$\begin{aligned} & \langle (N_{h0} + 1, M_{S'})_\tau | c_{\nu\sigma} | (N_{h0}, M_S)_\mu \rangle \\ & = \sum_{\lambda, \lambda'} \langle h_{\lambda'}, h_{\lambda'}, M_{S'} | c_{\nu\sigma} | h_\lambda, M_S \rangle \beta_\mu(h_\lambda) B_\tau(h_{\lambda'}, h_{\lambda'}) \Gamma_\sigma(S'_M, S) \end{aligned} \quad (13)$$

After substituting expression (13), (2) and Green's function (1) into relation (4) one obtains

$$N_{\nu}(x) = N_{\nu}^{12} + N_{\nu}^{23} = N_{s,\nu}^{12} + 3N_{t,\nu}^{12} + N_{\nu}^{23}, \quad (14)$$

where instead of the “blind” sum over the root vectors r , we use the summation over the physically meaningful indices τ, M and ΔS (i.e. the sum over all low- spin (s) and high-spin (t) two-hole states):

$$N_{s(t),\nu}^{12} = \sum_{\nu\sigma} \sum_{\tau} F_{\tau}^{s(t)}(x) \sum_{MM'} \left\{ \sum_{\lambda} \Gamma_{\sigma}(S'_M, S) \beta_{\mu=0}(h_{\lambda}) B_{\tau}(h_{\lambda}, h_{\nu}) \delta(S', S \pm |\sigma|) \delta(M', M + \sigma) \right\}^2, \quad (15)$$

where the (+) and (–) on the right side are used with the indices t and s , respectively, and the occupation factor in the PM phase reads

$$F_{(0,\tau)}^{s(t)}(x) = \begin{cases} \frac{1}{2}(1 - \alpha_{s(t)}x), & \tau = 0 \\ \frac{1}{2}(1 - x), & \tau \neq 0 \end{cases}, \quad (16)$$

with $\alpha_{s(t)} = 1 - 2/(2S' + 1)$ and $S' = 0, 1; S = 1/2$. Let us start with the contribution from the spin singlet f_{rs} states $N_{f_{rs}}^s$:

$$N_{\nu}(x) = (2N_{\lambda} - 1) - x(1 - N_{f_{rs}}^s) = N_e - x(1 - N_{f_{rs}}^s) \quad (17)$$

where the low and high spin contributions are

$$N_{s,\nu}^{12} = (1/2)[(N_{\lambda} + 1)(1 - x) + 2xN_{f_{rs}}^s] \quad (18)$$

and

$$N_{t,\nu}^{12} = (1/2)(N_{\lambda} - 1)(1 - x) \quad (19)$$

respectively. The relation $N_{\nu}^{23} = x(2N_{\lambda} - 2)$ for the contributions from the quasiparticle with the root vectors from $\{r_{23}\}$ subspace is derived similar to the previous expression for contribution (15). The number of possible singlet f_{rs} states is in the range $0 \leq N_{f_{rs}}^s \leq 2$, where

$$N_{f_{rs}}^s = 1 - \sum_{\lambda} \beta_0^2(h_{\lambda}) \times \sum_{\lambda'\lambda''} [1 - \delta_{\lambda\lambda'} - \delta_{\lambda\lambda''}] B_{\tau=0}^2(h_{\lambda'}h_{\lambda''}), \quad (20)$$

and $\tau = 0$ corresponds to the f_{rs} -quasiparticles. In deriving (17) we also use relation (12) and identity $\sum_{\lambda} \beta_{\mu}^2(h_{\lambda}) \sum_{\lambda'\lambda''} B_{\tau}^2(h_{\lambda'}h_{\lambda''}) = 1$ at any μ and τ . Since the sum

$$\sum_{\tau} [\beta_0(h_{\lambda}) B_{\tau}(h_{\lambda}, h_{\nu})] [\beta_0(h_{\lambda'}) B_{\tau}(h_{\lambda'}, h_{\nu})] = 0 \quad (21)$$

at any ν and $\lambda \neq \lambda'$, the contribution from the cross-term from (15) to the total number of the valence states is absent. In the case of the triplet nature of f_{rs} states, one obtains a similar expression to (17) with the contribution:

$$N_{f_{rs}}^t = 1 - \sum_{\lambda} \beta_0^2(h_{\lambda}) \sum_{\lambda' \neq \lambda'' \neq \lambda} B_{\tau=0}^2(h_{\lambda'}h_{\lambda''}), \quad (22)$$

where $0 \leq N_{f_{rs}}^t \leq 1$.

3. Results and discussion

From relation (17) it follows that the doped material can show both the metallic $N_{\nu}(x) > (N_e - x)$, and dielectric properties $N_{\nu}(x) = (N_e - x)$ at $N_{f_{rs}}^{s(t)} > 0$ or $N_{f_{rs}}^{s(t)} = 0$, respectively. This result is similar to the Wilson's criterion for the itinerant electron systems [9]. The f_{rs} states can be prohibited at $\delta(S', S \pm |\sigma|) = 0$ (the s -forbidden f_{rs} states) as well as when the doped hole changes the initial orbital configuration of $|(N_{h_0}, M_S)_{\mu=0}\rangle$ ground cell states as a whole (1-forbidden f_{rs} states). Surprisingly, only one forbidden

transition in the cell leads to the insulating state of the whole material. How this transition is different from many others? Nothing changes in the undoped material. The transition is distinguished only in the doped material by a specific symmetry of the ground states of doped carriers.

Under the conditions $N_{\lambda} = 1$ and $N_e = (1 - x)$ one always obtains a simple metal with the $N_{f_{rs}}^s = 2$ and $N_{\nu}(x) = (1 + x)$ valence states, as in the Hubbard model, where the high-spin (triplet) states are simply not available. Note, from the derivation of expression (17) it is clear that the orbital and spin degeneracy of the ground cell state in the sector $N_h = N_{h_0}$ does not result in the metallic character of the undoped material.

To apply the criterion to the high- T_c cuprates we choose the intracell Hamiltonian H_0 in accordance with the LDA+GTB approach [12]. This method generalizes the Hubbard's concept to real materials by using the Wannier cell function representation [14–17]. Let us calculate the magnitude of $N_{f_{rs}}^{s(t)}$ in the high- T_c cuprates, where $r = \{^2b_1, A_1\}$ –root vector which is relevant for (15) at $\mu=0$ and $\tau=0$ [15,17], i.e. it corresponds only to the A_1 singlet f_{rs} state. Using the exact diagonalization procedure with LDA parameters [18], one obtains the relation:

$$N_{f_{rs}}^s = 1 + [\beta_0^2(h_b) - \beta_0^2(h_{d_x})] \times [\beta_0^2(h_b^2) - \beta_0^2(h_{d_x}^2)] \approx 0.97 \quad (23)$$

for the singlet f_{rs} states, where doublet and singlet ground states (7) and (8) are

$$\begin{aligned} |^2b_1\rangle_0 &= \beta_0(h_b) |h_b, \sigma_{\frac{1}{2}}\rangle + \beta_0(h_{d_x}) |h_{d_x}, \sigma_{\frac{1}{2}}\rangle \\ |A_1\rangle_0 &= B_0(h_b^2) |h_b^2, 0_0\rangle + B_0(h_{d_x}^2) |h_{d_x}^2, 0_0\rangle \\ &\quad + B_0(h_{d_x}, h_b) |h_{d_x}, h_b, 0_0\rangle, \end{aligned} \quad (24)$$

h_b and h_{d_x} are the holes in the b -symmetrized cell states of oxygen and $d_{x^2-y^2}$ cooper states of the CuO_2 layer, respectively. There are no forbidden states, and the number of the valence states is almost constant: $N_{\nu}(x) \approx N_e - 0.03x$. However, if the doped holes for any physical reasons, avoid the states of the sector $N_h = (N_{h_0} + 1)$ and all of them are in the ground cell state as the hole pairs in the sector $(N_{h_0} + 2)$ (see Fig. 2), one has $N_{\nu}(x) = (1 - x/2)(2N_{\lambda} - 1) + (2N_{\lambda} - 3)(x/2) = N_e - x$, and, therefore, the material should be an insulator. In fact, at $T > T_c$ in the high- T_c cuprates there is a mysterious pseudogap state not associated with the fluctuations of the superconducting order [1,2,4]. The number of the f_{rs} states $N_{f_{rs}}$ is determined by the ability to correctly describe the interaction of the doped holes in the intracell Hamiltonian H_0 . In particular, to numerically evaluate the number of the valence states in (23) it is possible to choose an approach different from LDA.

We also believe that it is necessary to investigate doped manganites with the high temperature pseudogap in the PM phase [3] and cobaltites with the spin forbidden f_{rs} states $\delta(S', S \pm |\sigma|) = 0$ ($S=0$ and $S'=2$) at the Co^{3+} and Co^{4+} ground states [19] in connection with the subject under discussion.

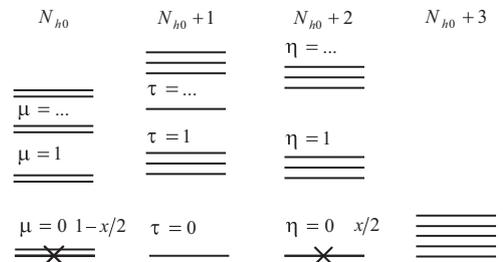


Fig. 2.

4. Conclusions

Formally, the criterion $N_{f_{fs}} = 0$ is valid at any hole concentration x and, unlike the Mott–Hubbard transition, a band crossover can occur at the ratio $t_{\lambda\lambda'}/U \ll 1$ between the hopping and the Coulomb interaction $\sim U(C_{N_{h0}+1}^2 + C_{N_{h0}-1}^2 - 2C_{N_{h0}}^2)$. A source of the metal–insulator transition could be any external effect resulting in the crossover between the top valence band and the forbidden f_{fs} state level, with the insulator becoming a metal. And we do not expect any effects from new quasiparticle states [20] in this range of Hamiltonian’s parameters.

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