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## METAL-INSULATOR TRANSITIONS IN CUPRATE HIGH-TEMPERATURE SUPERCONDUCTORS

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*We study the possible mechanisms of carrier localization and metal–insulator transitions (MITs) in inhomogeneous hole-doped cuprates. We show that the insulating state of doped cuprates is different from the Mott insulating state of undoped cuprates characterized by the charge-transfer (CT) gap. The dopant- and carrier-driven inhomogeneities favor the charge segregation and ordering in the form of a three-dimensional (3D) network of carrier-rich and carrier-poor stripes. The specific charge ordering results in the formation of different superlattices and energy bands of localized carriers in inhomogeneous high- $T_c$  cuprates. The energy bands associated with the charged stripes develop in the middle of the CT gap. We argue that the in-gap impurity and polaronic states (bands) develop into metallic states at some critical doping levels. We use the uncertainty relation to obtain the specific conditions for the MITs in doped cuprates. The applicability limits of these MITs in cuprates are clarified. Our results are in good agreement with the existing experiments on La-based and other cuprates.*

**Keywords:** metal-insulator transitions, doped cuprates, polaronic carriers, superlattices.

### Introduction

Understanding the mechanisms of carrier localization, metal–insulator transitions (MITs) and suppression of high- $T_c$  superconductivity in hole-doped cuprates remains one of the central issues in condensed matter physics. In the undoped cuprates, the carrier localization is caused by the strong on-site Coulomb repulsion and the Mott–Hubbard model is believed appropriate to describe the MITs in the cuprates [1]. As the doping level increased towards the underdoped level, the hole-doped cuprates become unusual metals and the importance of electron correlations diminishes [2] and other mechanisms may play a major role in carrier localization and MITs in these systems. For lightly doped cuprates within the insulating antiferromagnetic (AF) phase, the electron correlation is likely to dominate the carrier localization; when the underdoped region is approached, it is not obvious which process will dominate. In hole-doped cuprates, the phenomena of carrier localization and MITs seem to be much more complicated by carrier–dopant (defect)–lattice or carrier–lattice interaction effects, which usually were ignored in the existing theoretical models. In reality, there are important unexplained differences between the MITs observed in lightly doped [1,3], underdoped [4,5] and optimally doped [6] cuprates. Another unresolved issue in the physics of high- $T_c$  cuprates is the role of the electronic inhomogeneity and charge ordering in the phase separation in the form of alternating stripes associated with the two distinct electronic phases (i.e. metallic/superconducting and insulating phases) [7-12], which is intimately related to carrier localization and MITs in these materials. In this work, we address the carrier localization and MITs by providing important new insight into the physics of these phenomena in inhomogeneous hole-doped cuprates.

### 3. Specific charge ordering and formation of different superlattices

The hole-doped cuprates are inhomogeneous systems (where the dopants and charge carriers are distributed inhomogeneously) and the underdoped cuprates are more inhomogeneous than overdoped ones [10]. The dopant-driven and carrier-driven inhomogeneities may produce regions

with different doping levels or with spatially varying local density of charge carriers. The doped cuprates have a rich phase diagram and their metallic and superconducting states are quite different from those of conventional metals and superconductors [13-16]. The insulating state of these materials is also substantially different from that of undoped cuprates [15,17]. Instead of strong electron correlations (i.e., on-site Coulomb interactions), which play a dominating role only in undoped cuprates and drive these systems into a CT-type Mott insulating state [18], the carrier-defect-phonon and carrier-phonon interactions and the electronic inhomogeneities play an important role in doped cuprates and are responsible for the carrier localization and pairing and the charge segregation, which may manifest itself via local phase separation and stripe formation (i.e., the charge carriers form metallic stripes, “rivers of charge carriers” [15], separated by insulating stripes). In particular, in doped high- $T_c$  cuprates the inhomogeneous spatial distribution of charge carriers leads to their segregation into carrier-rich and carrier-poor regions.

Whereas the short- and long-range carrier-defect-phonon and carrier-phonon attractive interactions may dominate and give rise to charge aggregation in carrier-rich metallic regions together with charge depletion in spatially separated carrier-poor regions with no mobile carriers. In high- $T_c$  cuprates, the local inhomogeneity and the competitions between the kinetic energy, the above attractive interactions and the long-range Coulomb interaction produce nanoscale self-organized structures called stripes. Further, the anisotropy of the dielectric constants ( $\epsilon_\infty$  - high frequency dielectric constant and  $\epsilon_0$  - static dielectric constant) and the smallness of  $\eta$  ( $=\epsilon_\infty/\epsilon_0$ ) in the cuprates favor such phase separation into alternating carrier-rich and carrier-poor regions. As the doping level increases towards underdoped regime, specific charge ordering in the form of a 3D network of dynamic (metallic) and static (insulating) stripes takes place in these regions, where distinctly different superlattices and energy bands of dopants and self-trapped carriers (intrinsic large polarons) are formed at their inhomogeneous spatial distribution. In particular, the hydrogenic impurity centers (impurities with loosely bound quasi-free carriers or large polarons) and extrinsic large polarons (or impurities with tightly bound large polarons) are assumed to form the superlattices with the lattice constant  $a_I$  and coordination number  $z$ . The charge ordering in carrier-poor and carrier-rich domains results in the formation of simple cubic, body-centered cubic and face-centered cubic superlattices with coordination numbers  $z = 6, 8$  and  $12$ , respectively, and the formation of different impurity bands in the CT gap of the cuprates. In the tight-binding approximation the widths of the impurity bands can be determined from the relation

$$W_I = 2zJ, \quad (1)$$

where  $J$  is the hopping integral between nearest-neighbor impurity centers.

For the case of the hydrogenic impurity centers, the hopping integral in Eq. (1) may be approximated as [19]

$$J = \frac{e^2 \lambda}{\epsilon_0} \exp[-\lambda R], \quad (2)$$

where  $\lambda$  is the reciprocal radius of an impurity state,  $R$  is the distance between the impurity centers.

With increasing  $R$ , the impurity band is continuously narrowed, finally ending in the discrete levels of individual impurity centers ( $R \gg 1/\lambda$ ). In this case carriers are localized to the dopant sites or confined to the potential wells at impurity sites and the system is converted into an insulator. Therefore, conduction takes place in a narrow impurity band by hopping of carriers from one to another impurity site. When the number of impurity states increases, their overlap increases, thus increasing  $W_I$  and the charge transport in sufficiently broadened impurity bands becomes band-like

(i.e., metal-like). Determination of the critical carrier concentration at which such a transition from the hopping-like to the metal-like charge transport occurs in cuprates is a very intricate problem, which is at present not completely solved. We believe that the intrinsic large polarons like impurity centers form different superlattices and the widths of the polaronic bands just as the widths of the impurity bands are determined from a relation just like Eq. (1) [20].

## 2. Possible mechanisms of metal-insulator transitions and stripe formation in cuprates

In this section we discuss the different mechanisms that cause carrier localization in hole-doped cuprates. The question now arises how, under certain conditions, a system with  $n$  carriers in cuprates may undergo a phase transition from a delocalized into a localized state. In inhomogeneous hole-doped cuprates, the strong carrier-carrier interactions, structural disorder, carrier-defect-phonon and carrier-phonon interactions may play an important role in carrier localization and MITs. We first examine the condition for the existence of the localized bipolaronic states in cuprates. At very low doping, the separate levels of extrinsic and intrinsic large bipolarons are formed in the CT gap of the cuprates. As the doping increases towards underdoped region, the binding energies of such large bipolarons start to decrease gradually and become zero at some doping levels. In particular, the binding energy of the intrinsic large bipolaron is now defined as

$$\Delta_b = E_{bB} - 2\varepsilon_F, \quad (3)$$

where  $E_{bB}$  is the binding energy of the large bipolarons,  $\varepsilon_F$  is the Fermi energy of large polarons.

Actually, large bipolarons can exist in cuprates in carrier-poor domains at  $\eta < \eta_c$  and remain localized. At a certain doping level  $n = n_c$  or  $x = x_c = n_c/n_a$  (where  $n_a = 1/V_a$  is the density of the host lattice atoms,  $V_a$  is the volume per  $\text{CuO}_2$  unit in cuprates),  $\Delta_b = 0$  and the large bipolaron will dissociate into two large polarons. Therefore, the critical carrier concentration  $n_c$  determined from Eq. (3) is

$$n_c = \frac{(m_p E_{bB})^{3/2}}{3\pi^2 \hbar^3}, \quad (4)$$

where  $m_p$  is the polaron mass.

For the  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) system we can evaluate  $n_c$  using the parameter values  $m_p = 2.1m_e$  ( $m_e$  is the free electron mass),  $\varepsilon_\infty = 3.5-4.5$ ,  $\eta = 0.04-0.08$ ,  $E_{bB} \approx (0.01-0.10)$  eV. Then we obtain  $n_c \approx (0.0514-1.6238) \cdot 10^{20} \text{ cm}^{-3}$ . Taking into account that the value of  $V_a$  in the orthorhombic LSCO is  $190 \text{ \AA}^3$ , we find the doping levels  $x_c = 0.001-0.031$  at which large bipolarons dissociate into large polarons. This means that such localized 3D bipolarons can exist only in the lightly doped LSCO. At  $x > x_c$  the energy bands of extrinsic and intrinsic large polarons may exist, thus permitting charge transport by means of intra-band conduction. In case of narrow polaronic bands, charge transport becomes hopping-like and is caused by intra-band hopping processes [21]. One can assume that if the bandwidth of the extrinsic large polarons exceeds some critical value, their intra-band conduction becomes metal-like. We attempt to find the conditions under which such a insulator-to-metal transition occurs in doped materials.

The conditions for carrier localization or delocalization can be obtained by using the uncertainty principle:  $\Delta p \Delta x \geq \hbar/2$ , where  $\Delta p$  and  $\Delta x$  are the uncertainties in the momentum and coordinate of a carrier, respectively. This uncertainty relation can be written as

$$\Delta x \cdot \Delta E \simeq \frac{\hbar^2 (\Delta k)^2}{2m^*} \frac{1}{2\Delta k}, \quad (5)$$

where  $\Delta E$  and  $\Delta k$  are the uncertainties in the energy and wave vector of the carrier,  $m^*$  is the effective mass of a carrier.

The expression  $\hbar^2(\Delta k)^2/2m^*$  in Eq. (5) represents the uncertainty in the kinetic energy of quasi-free carriers. Taking into account that in the impurity band the uncertainties in the energy of carriers is of order  $W_I/2$  and the uncertainty in their wave vector is about  $1/a_I$ , the relation (5) can be rewritten in the form (cf. [22])

$$\Delta x \cdot \Delta E \simeq \frac{W_I a_I}{4}. \quad (6)$$

On the other hand, the uncertainty in the energy  $\Delta E$  of carriers at the hydrogen-like impurity centers is of the order of the repulsive Hubbard energy  $U$ , whereas  $\Delta x$  would be of the order of  $a_H$  ( $a_H$ -Bohr radius). In this case the condition for carrier localization can be written in the form

$$\frac{U}{W_I} > \frac{0.25 a_I}{a_H}, \quad (7)$$

which is useful in determining the quantitative criteria for the Mott MITs. From Eq. (7) it follows that the Mott MIT is governed by the ratios  $U/W_I$  and  $a_I/a_H$ .

We believe that the strong carrier-defect-phonon and carrier-phonon interactions can initiate MITs in cuprates. If the uncertainty in the energy of large polarons bound to impurities is of order  $E_{pl}$  and the uncertainty in the coordinate  $\Delta x$  of these carriers is the order of the radius of the impurity centers  $R_I$ , the condition for carrier localization or a new type of MIT can be written as

$$\frac{E_{pl}}{W_I} > \frac{0.25 a_I}{R_I}, \quad (8)$$

from which it follows that the MIT is governed by the ratios  $E_{pl}/W_I$  and  $a_I/R_I$ .

Similarly, the MIT may be also driven by varying the Fermi energy  $\varepsilon_F$  and the lattice constant  $a_p$  of intrinsic large polarons. The condition for such a MIT can be written as

$$\frac{E_{pl}}{\varepsilon_F} \simeq \frac{0.5 a_p}{R_p}. \quad (9)$$

We examine below the possibility of the Mott and other MITs in  $La$ -based cuprates.

### 2.1 The Mott-type metal-insulator transitions

Knowing the Hubbard energy  $U$  and the bandwidth  $W_I$  one can find the criterion for the Mott MIT from the condition (7). For the case of hydrogen-like impurity centers, the value of  $U$  is equal to  $(55/64)E_I^H$  [23], while the bandwidth  $W_I$  is given by [19]

$$W_I \simeq 2z \frac{\varepsilon^2}{\varepsilon_0 a_H} \exp[-\lambda R], \quad (10)$$

where  $\lambda = a_H^{-1}$ .

For  $a_I = 4a_H$ , we obtain from the relation (7) the familiar localization picture described by the Mott criterion  $W_I/U \simeq 1$ . If we assume  $a_I \simeq 3.4783a_H$ , we find  $W_I/U \simeq 1.15$ , which is known as the Hubbard criterion for the MIT. Then, the Mott MIT point is determined from the condition

$$\frac{W_I}{U} = \frac{64z}{55} \exp\left[-\frac{R}{a_H}\right] = \frac{4a_H}{a_I}. \quad (11)$$

If the impurity centers form simple cubic, body-centered cubic and face-centered cubic superlattices with  $a_I = R$  (for  $z = 6$ ),  $a_I = 2R/\sqrt{3}$  (for  $z = 8$ ) and  $a_I = \sqrt{2}R$  (for  $z = 12$ ), the appropriate densities of carriers per unit cells of such superlattices are  $n = 1/a_I^3$ ,  $n = 2/a_I^3$  and  $n = 4/a_I^3$  respectively. It follows that the distance between nearest-neighbor impurity centers are equal to  $R = 1/n^{1/3}$  (for  $z = 6$ ),  $R = \sqrt{3}/(4n)^{1/3}$  (for  $z = 8$ ) and  $R = 2^{1/6}/n^{1/3}$  (for  $z = 12$ ). Thus, Eq. (11) can be written as

$$\frac{64z}{55n^{1/3}a_H} \exp\left[-\frac{1}{n^{1/3}a_H}\right] = 1, \text{ for } z=6, \quad (12)$$

$$\frac{64\sqrt[3]{2}z}{55n^{1/3}a_H} \exp\left[\frac{\sqrt{3}}{\sqrt[3]{4}n^{1/3}a_H}\right] = 1, \text{ for } z=8, \quad (13)$$

$$\frac{64\sqrt[3]{4}z}{55n^{1/3}a_H} \exp\left[\frac{2^{1/6}}{n^{1/3}a_H}\right] = 1, \text{ for } z=12, \quad (14)$$

from which we obtain the following criteria for the Mott MITs:  $n_c^{1/3}a_H \simeq 0.3265$  (for  $z = 6$ ),  $n_c^{1/3}a_H \simeq 0.2968$  (for  $z = 8$ ),  $n_c^{1/3}a_H \simeq 0.2504$  (for  $z = 12$ ).

If we take  $m^* = m_e$  and  $\varepsilon_0 = 30$ , we find  $a_H = 15.87 \text{ \AA}$  in LSCO, so that the Mott MITs would occur at the hole concentrations  $x_c = n_c/n_a \simeq 0.00075 - 0.00166$ . These values of  $x_c$  are much smaller than the value of  $x_c \simeq 0.02$  at which the destruction of the AF order is observed in LSCO [1]. However, the large-radius dopant and large polaron may form the hydrogenic impurity center in LSCO. The hydrogenic state of a large polaron in LSCO has an effective Bohr radius  $a_H = 0.529\varepsilon_0(m_e/m_p) \text{ \AA}$ . If we take  $m_p = 2.1m_e$  and  $\varepsilon_0 = 28$ , we find  $a_H \simeq 7.06 \text{ \AA}$ . In this case the Mott transitions would occur at  $x_c = 0.0141$  (for  $z = 8$ ) and  $x_c \simeq 0.0188$  (for  $z = 6$ ). These values of  $x_c$  are close to the experimental values  $x \simeq 0.018 - 0.02$  at which the AF order is destroyed in LSCO [1, 24].

## 2.2 The other metal-insulator transitions and stripe formation

From the relation (8) we obtain the following criteria for the new MITs:

$$\frac{0.5z}{n^{1/3}R_I} \exp\left[-\frac{1}{n^{1/3}R_I}\right] = 1, \text{ for } z=6, \quad (15)$$

$$\frac{0.5\sqrt[3]{2}z}{n^{1/3}R_I} \exp\left[-\frac{\sqrt{3}}{\sqrt[3]{4}n^{1/3}R_I}\right] = 1, \text{ for } z=8, \quad (16)$$

$$\frac{0.5\sqrt[3]{4}z}{n^{1/3}R_I} \exp\left[-\frac{2^{1/6}}{n^{1/3}R_I}\right] = 1, \text{ for } z = 12, \quad (17)$$

from which it follows that  $n^{1/3}R_I \simeq 0.651$  (for  $z = 6$ ),  $n^{1/3}R_I \simeq 0.453$  (for  $z = 8$ ),  $n^{1/3}R_I \simeq 0.3354$  (for  $z = 12$ ).

We are now able to evaluate  $n_c$  in La-based cuprates using the above values of  $n^{1/3}R_I$ . For  $\varepsilon_\infty = 4.5$ , and  $\eta = 0.04 - 0.12$  the value of  $R_I$  is found to vary between 8.44 and 9.91  $\text{\AA}$ . In these cases, the MITs occur at  $x_c \simeq 0.0539 - 0.0872$  (for  $z = 6$ ) and  $x_c = 0.0182 - 0.0294$  (for  $z = 8$ ) in La-based cuprates with small-radius dopants. When  $\varepsilon_\infty = 3.5$ ,  $\eta = 0.04 - 0.12$  and  $R_I \simeq 6.41 - 7.56$ , the MITs and stripe formation occur in these systems at  $x_c \simeq 0.1213 - 0.1990$  (for  $z = 6$ ) and  $x_c \simeq 0.0409 - 0.0671$  (for  $z = 8$ ). For  $\varepsilon_\infty < 3.5$  and  $z = 6$ , the metal-insulator boundary of La-based cuprates containing small-radius dopants lies in the deeply over doped region ( $x_c > 0.2$ ).

Further, the double substitution of smaller cations for host lattice ions and dopants may also favor the MITs and stripe formation occurring in a wide range of doping of  $La$ -based cuprates. In  $La_{2-x-y}Nd_ySr_xCuO_4$  and  $La_{2-x-y}Eu_ySr_xCuO_4$  with small-radius dopants  $Nd^{3+}$  and  $Eu^{3+}$ , the static stripe phases are observed in a wide range of carrier concentration [25].

## Conclusion

We argued that the inhomogeneous spatial distribution of dopants and hole carriers favor the specific charge ordering in the form of a 3D network of carrier-rich and carrier-poor stripes and the formation of different superlattices and in-gap bands of dopants and large polarons. We have shown that the localized in-gap states develop into metallic states at some critical doping level. We have succeeded in deriving the quantitative criteria for the MITs caused by strong carrier-carrier (i.e., Coulomb) interaction on the same impurity center, disorder, carrier-dopant-phonon and carrier-phonon interactions. The applicability limits of the Mott and other MITs in hole-doped cuprates are determined. Our results show that the other MITs in the cuprates are accompanied by the formation of static (insulating) stripes in carrier-poor domains and dynamic (metallic) ones in carrier rich domains.

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