Hole-driven MIT theory, Mott transition in VO₂, MoBRiK*

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For inhomogeneous high-\(T_c\) superconductors, hole-driven metal-insulator transition (MIT) theory explains that the gradual increase of conductivity with increasing hole doping is due to inhomogeneity with the local Mott system undergoing the first-order MIT and the local non-Mott system. For VO₂, a monoclinic and correlated metal (MCM) phase showing the linear characteristic as evidence of the Mott MIT is newly observed by applying electric field and temperature. The structural phase transition occurs between MCM and Rutile metal phases. Devices using the MIT are named MoBRiK.

It has been generally accepted that conductivity, \(\sigma\), and \(T_c\) for inhomogeneous high-\(T_c\) superconductors [1] as strongly correlated systems gradually increase with doped hole density in a Mott insulator from under-doping to critical doping [2], although a first-order transition near the Mott insulator had been theoretically suggested [3]. These phenomena seemed to be explained by the Mott-Hubbard (MH) continuous metal-insulator transition (MIT) theory. However, since the MH theory was established for homogeneous system, the theory does not explain the phenomena in inhomogeneous system. The first-order Mott MIT without the structural phase transition (SPT) has not clearly been proved.

This paper briefly describes important ideas and physical meanings of the hole-driven MIT theory (extended Brinkman-Rice (BR) picture [4]) named by a reviewer in ref. 7 and explains the above phenomena. An experimental observation is also presented to clarify the Mott MIT.

Metal has the electronic structure of one electron per atom in real space, i.e. half filling, which indicates that \(\delta q=\sum(q_q,q_j)=0\) where \(q_q\) and \(q_j\) are charge quantities at \(i\) and \(j\) nearest neighbor sites, respectively [4]; there is no charge density wave. The BR picture explains physical properties of a strongly correlated metal, which was developed when \(n=l\) for a homogeneous system with one electron per atom, where \(n\) is the number of electrons in the Mott system with the electronic structure of metal (Fig. 1(a) left) and \(l\) is the number of lattices in the measurement region [3]. The Mott insulator becomes metal via MIT.

When \(n<1\) (inhomogeneous) (Fig. 1(a) right), Fourier transform from K-space to real-space is not satisfied. The local Mott insulator (system) becomes metal after MIT. When the inhomogeneous system is measured, local metal (Mott) regions are averaged over all lattice sites (measurement region); the measured physical quantity is an averaged one. Then the effective fractional charge is given by \(e'\) = \(\rho e\), where \(0<\rho=n/l\leq1\) is band filling. The fractional Coulomb energy between quasiparticles is defined by \(U=\rho U' = \kappa \rho^2 U_c\), where \(0<\kappa<1\) (\(\kappa=U'/U_c\) is the correlation strength), \(0<\kappa \rho^2<1\), \(U_c\) is the critical on-site Coulomb interaction in the BR picture [3]. The system is satisfied with Fourier transform. The physical meaning of the fractional \(e'\) and \(U\) is the effect of measurement (average) and not true effect.

The averaged system has an electronic structure of one effective charge per atom: \(\rho' = n'/l=1\) where \(n'\) is the number of the effective charges. This is the same electronic structure as one of the BR picture with \(\kappa=1\) [3,4]. The effective mass of the quasiparticle is derived from Gutzwiller’s variational method and has the same form as that in the BR picture. This is because the averaged system with \(\rho'=1\) and the true system satisfied with the BR picture are mathematically self-consistent. The effective mass is given by

\[
\frac{m^*}{m} = \frac{1}{1 - (\frac{U_c}{\rho})^2} = \frac{1}{1 - \kappa^2 \rho^4}, \quad 0 < \kappa \rho^2 < 1.
\]

In the experimental analysis for an inhomogeneous system, \(\kappa\) was estimated as one [4]. The observed effective mass is given by

\[
\frac{m^*}{m} = \frac{1}{1 - \rho^4}.
\] (1)

Eq. (1) is defined at \(\rho \neq 1\) (Fig. 1(b)) and has a first-order discontinuous MIT between a Mott insulator with \(U_c\) at \(\rho=1\) and a metal at \(\rho_{\text{max}} < 1\). The MIT is caused due to breakdown of \(U_c\) by hole doping of a very low density, \(n_c = 1 - \rho_{\text{max}}\), into the Mott insulator (Fig. 1(c)); this is a hole-driven MIT. \(n_c\) is a minimum constant hole density where the MIT occurs. Conversely, control of \(n_c\) makes the Mott insulator switch between insulator and metal. After the MIT, the local Mott insulators become strongly correlated local metals with a \(\kappa \neq 1\) value in the BR picture, \(m^*\) in Eq. (1) is an average of the true effective mass in the BR picture.

For inhomogeneous superconductors, the gradual increase of conductivity with doping [2] is that the local Mott insulators in (Fig. 1(a right)) continuously change into metal with hole doping; \(\sigma \propto (m^*/m)^2\) with doping density, \(\rho\), dependence in Eq. (1). The reason why the measured conductivity with doping is continuous is...
that the magnitude of a local metal conductivity after the MIT is very small because the local area is within about 3nm [1].

**FIG. 1.** (a) Left: homogeneous Mott system. Right: inhomogeneous system with local Mott insulators where the first-order MIT occurs and non-Mott insulator regions where the MIT does not occur. Example: Mott insulator of VO$_2$, non-Mott insulator of V$_2$O$_5$. (b) The effective mass of Eq. 1 with divergence near band filling $\rho = 1$. Red-dash line indicates the first-order MIT. Yellow region is critical hole density $n_c$ in which MIT occurs. (c) The physical meaning of Eq. 1. Breakdown of $U_c$ is shown by hole doping of a low density into Mott insulator; this is the MIT. (d) Electric field and temperature dependence of the MIT measured for a VO$_2$-based two terminal device (Inset, brown: VO$_2$; yellow: electrodes). The MCM metal phase showing the linear characteristic is clearly shown.

Furthermore, the Mott MIT [5-7] and the Peierls MIT with the SPT [8,9] have been controversial even in VO$_2$ as a representative Mott insulator. This is due to ambiguity of relation between MIT and SPT. Fig. 1(d) clearly shows the relation. VO$_2$ was known to simultaneously undergo the MIT and the SPT near 68°C (similar to V=1 case in Fig. 1(d)). Actually, even in this case, the MIT was not simultaneous with the SPT in our work [7]. When voltage is applied to a VO$_2$-based device [7], the MIT occurs between T (monoclinic semiconductor phase, transient triclinic) [5,6] and MCM (monoclinic and correlated metal) phase with the increase of $\sigma$. The SPT happens between MCM and R (rutile tetragonal metal phase) (SPT instability, red-dash line in Fig. 1(d)); this was confirmed by micro-Raman experiment [10]. This indicates that the MIT is not related to the SPT. The MCM phase as evidence of the Mott transition clarifies the controversial problem. MCM differs from Pouget et al.’s M$_2$ Mott-Hubbard insulator phase [5,6]. We consider that T can be the paramagnetic Mott insulator with the equally spaced chain structure because T and MCM have the same structure.

MCM is caused by $n(E) = n_c(T, E) - n(T)$, where $n(E)$ is the hole density excited by electric field (voltage), $n(T)$ is the hole density excited by temperature, $n_c(T, E)$ is the critical hole density in which the MIT occurs by the electric field and temperature excitations [7]. Hole carriers were confirmed by Hall measurement [7]. For constant $n_c$, $n(T)$ decreases as $n(E)$ increases. Thus, $T_{MIT}$ decreases, which is evidence of the fact that the MIT is controlled by doped holes. This is predicted in Eq. 1. Further, MCM is regarded as a non-equilibrium state because metal exists at the divergence in Eq. 1 (Fig. 2(b)). First-order MIT devices using the transition between T and MCM are called MoBRiK from names of Mott-Brinkman-Rice-Kim physicists who have established and extended the MIT.

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