Interfaces of correlated electron systems: Proposed mechanism for colossal electroresistance

Takashi Oka¹ and Naoto Nagaosa¹,²,³

¹Correlated Electron Research Center (CERC), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba Central 4, Tsukuba 305-8562, Japan.
²Department of Applied Physics, University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan.
³CREST, Japan Science and Technology Corporation (JST), Saitama, 332-0012, Japan.

(Dated: September 15, 2018)

Interfaces of correlated electron systems: Proposed mechanism for colossal electroresistance

Mott’s metal-insulator transition at an interface due to band bending is studied by the density matrix renormalization group (DMRG). We show that the result can be recovered by a simple modification of the conventional Poisson’s equation approach used in semi-conductor heterojunctions. A novel mechanism of colossal electroresistance is proposed, which incorporates the hysteretic behavior of the transition in higher dimensions.

PACS numbers: 71.30.+h, 73.40.-c, 71.10.Fd

Strongly correlated electron systems (SCES) – a group of materials in which the effect of Coulomb repulsion is large – is one of the major candidates on which the next-generation electronics may be built. This strong hope put on to SCES electronics stems from the richness of its phase diagram. Various electronic and magnetic phases transitions are reported with high sensitivity to external conditions. Inside electric devices, the electrons behave collectively, so the high sensitivity of SCES may lead to drastic functionalities.

Yet, SCES electronics faces a strong conceptual barrier to widespread acceptance and application, since it is essentially many-body where the useful concepts such as band structure are believed to fail. It is expected to be the case especially at the interfaces. In this Letter, we study interface Mott transition near an interface between a metal electrode and SCES in terms of the density matrix renormalization group (DMRG) method. Surprisingly, we found that the conventional band bending picture based on Poisson’s equation is valid with a small modification (eq. (3)). Namely, the conventional concepts in semiconductor devices are still useful and valid to design the SCES devices. As an application, we propose a novel mechanism of the colossal electroresistance (CER), i.e., the large switching of resistance.

Among many possibilities to industrial applications, perhaps CER in SCES heterostructures is closest to realization, e.g., nonvolatile resistance random access memories (RRAM). The device consists of a film of perovskite manganite such as Pr₁₋ₓCaₓMnO₃, δ = 0.3 (PCMO), which is a holed-doped Mott insulator, sandwiched by two metallic electrodes. The current-voltage curve shows large hysteresis at room temperature, where the resistivity of the on and off states differ by a large factor. Although this effect has been explored in details, the understanding of the mechanism is still missing. Baikalov et al. pointed out from multileads resistance measurements, that the switching take place at the interface. Then, Sawa et al. reported that the CER behavior depends on the work-function of the electrode metal. They interpreted the I-V characteristics using a Schottky contact model (metal/p-type semiconductor) accompanied with an interface state. Another mechanism was proposed by Rozenberg et al.

The mechanism of CER we propose here do not assume any interface states but attributes the large non-linearity of the I-V characteristics to interface Mott transition, where a layer of Mott insulator blocks the current. In dimensions higher than one, the transition is first order, and the width of the Mott insulator layer depends on how the voltage is changed. This explains the hysteretic behavior of the I-V characteristics.

The Letter consists of two parts. We first study a one-dimensional model of a metal/SCES interface using DMRG combined with Poisson’s equation. We show that interface Mott transition can be understood on the basis of local equilibrium, i.e., electric state is determined by the local value of the potential. Then, we explore the hysteresis loop of the I-V characteristics on a phenomenological basis assuming the a hysteretic density-potential curve for a system in higher dimensions. Along with the mechanism of CER, another interesting consequence of interface Mott transition is proposed: A quantum well structure emerges spontaneously (Fig.1(a)) at the interface of a hole (electron) doped Mott insulator and an electrode with small (large) work-function. Our results may lead to fabrication of clean 2D metallic systems analogous to the high mobility transistor in semiconductor physics.

DMRG study of a 1D interface — We start with the one-dimensional model of a metal/SCES interface on a lattice with total Hamiltonian

\[ H_{\text{tot}} = H_{\text{ele}} + H_{\text{SCES}} + H_{\text{jnc}}, \]

where the electrode

\[ H_{\text{ele}} = -t \sum_{\sigma} \sum_{i=1}^{L-1} \left( c_{i+1\sigma}^\dagger c_{i\sigma} + H.c. \right) \]

and SCES

\[ H_{\text{SCES}} = -t \sum_{\sigma} \sum_{i=L-1}^{L} \left( c_{i+1\sigma}^\dagger c_{i\sigma} + H.c. \right) + \sum_{i=L+1}^{2L} \left( U n_{i\uparrow} n_{i\downarrow} + V n_{i} \right) \]

is connected by a junction

\[ H_{\text{jnc}} = -t \sum_{\sigma} \left( c_{L+1\sigma}^\dagger c_{L/2\sigma} + H.c. \right). \]

\( t \) is the hopping

\[ \sigma \in \{ \uparrow, \downarrow \}. \]
element, \( L \) the total size of the system with the interface at the center, and \( U \) denotes the on-site Coulomb repulsion (for a related work, see [12]). We take the units \( \hbar = \varepsilon_0 = 1 \). The potential \( V_i \) defined in the SCES region obeys 1D Poisson’s equation, whose discretised solution is

\[
V_i = -\alpha \sum_{j=1}^{L} \sum_{k=1}^{L} (n_i^n - n_i^+) + V_\infty, \quad i \in [L/2+1, L], \tag{2}
\]

where \( n_i = \langle \sum_{\sigma} \epsilon_{i\sigma} c_{i\sigma} \rangle \) is the electron density and \( n_i^+ \) the positive background related to the hole doping ratio \( \delta \) by \( n_+ = 1 - \delta \), and \( \alpha = \frac{\varepsilon_\infty}{\varepsilon} \) (\( \varepsilon \) : lattice const., \( \varepsilon_\infty \) : dielectric constant). \([a, b] \) stands for the interval between \( a \) and \( b \). \( H_{\text{SCES}} \), without \( V_i \), is identical to the Hamiltonian of the one-dimensional Hubbard model, which exhibits a Mott transition at half-filling if \( U > 0 \) [13]. If \( n \neq 1 \), the ground-state is metallic, a state known as the Tomonaga-Luttinger liquid (e.g. [14]).

The DMRG calculation is performed as follows. In order to obtain the ground-state consistent with the potential determined by Poisson’s equation, we update the potential using eq. (2) at each step of the finite size method in the DMRG procedure [10]. This is repeated more than 50 times to obtain total convergence. The typical size of the block Hilbert space used here is \( m = 250 \).

We fix the total number of electrons \( \sum_{i=1}^{L} n_i = Ln_+ \), thus, the Fermi level of the electrode changes as electrons (holes) are injected to the SCES region when we vary \( V_\infty \). The interface is characterized by the work-function difference \( V_\infty \equiv V_{L/2+1} - V_L = \text{(work-function of electrode)} - \text{(work-function of SCES)} \). We note that \( V_\infty \) achieving a given \( V_D \) depends on \( L \).

In Fig. 1 we plot the electron density and the potential in a metal/SCES interface. Three typical solutions are plotted in an increasing order of \( V_D \) from (a) to (c).

(a) **Interface Mott transition:** When the electrode’s work-function is small enough, a quantum well is formed at the interface, i.e., two insulating (MI) and BI layers with width \( d_{\text{MI}}, d_{\text{BI}} \), sandwiches a metallic region (M) with width \( d_{\text{M}} \). The \( V_D \) dependences of these widths are plotted in Fig. 2(b).

(b) **Ohmic junction:** The Fermi surface of the electrode and SCES balances and no barrier is formed.

(c) **Schottky barrier:** A Schottky barrier is formed as in conventional metal/semiconductor interfaces.

It is noted here that the qualitative features of the results are all captured well by the conventional band bending picture by replacing the valence (conduction) band by the lower (upper) Hubbard band as shown in Fig. 1. We also note that in the three cases, \( n(x) \) shows an oscillatory behavior in the electrode regime, which is the 1D Friedel oscillation \( \delta n(x) = \cos(2k_F r + \eta_F)/r \) with \( k_F \) the Fermi wave number, \( \eta_F \) a phase shift, and \( r \) the distance from the interface.

In Fig. 2(a), we plot the electron density \( n(x) \) against \(-(V(x) - V_\infty)\) in the SCES regime \( x \in [51, 90] \), where data near the boundary \( x \in [91, 100] \) were omitted to avoid the boundary effect. The data falls onto a universal density-potential curve, which increases as the potential becomes deeper. In the middle of the curve, there is a plateau at half-filling whose width is \( \Delta(U) \). The in-
interface Mott transition is an analog of the filling driven Mott transition [12, 16]. There the filling $n(\mu)$ of a grand canonical system is studied while the chemical potential $\mu$ is varied. The universal density-potential curve is an analogue of the filling driven Mott transition follows the $n(\mu)$ curve in the bulk transition.

Poisson’s equation and local equilibrium approximation

— Since the metal/SCES interface determines the transport properties of the device, the width of the layers $d_{\text{MI}}, d_{\text{M}},$ and $d_{\text{BI}}$ in Fig. 1(a) is of practical interest. Here, we derive the width by solving the modified Poisson’s equation

$$\frac{d^2 V(x)}{dx^2} = -\frac{e}{\varepsilon} (n(V(x)) - n_{+}),$$

where we assume local equilibrium, that is, we assume that the electron density only depends on the local value of the potential. In the following, we evaluate eq. (3) to obtain the widths expressed solely by the potential difference $V_D$, hole doping ratio $\delta$, band width $W$, and the Mott gap $\Delta$.

We adopt a simplified density-potential relation by linearizing the DMRG result Fig. 2(a); A constant compressibility $-dn(V)/dV = \kappa = 2/(W - \Delta)$ is assumed for $-(V(x) - V_\infty) \in [\delta - (1 - \kappa)/\delta, \delta/\kappa + \Delta, (1 + \delta)/\kappa + \Delta]$ and $n(V) = 1$ for $-(V(x) - V_\infty) \in [\delta/\kappa + \Delta, \delta/\kappa + \Delta]$ and $n(V) = 2$ for $-(V(x) - V_\infty) \in [(1 + \delta)/\kappa + \Delta, \infty]$. We seek for a solution with a fixed density at the bulk SCES $n(x) = n_+ = 1 - \delta$. Mott insulator layer (MI) $n(x) = 1$ and band insulator layer (BI) $n(x) = 2$, but varies in the metallic region (M). The width of MI layer is nonzero when $V_D < -\delta/\kappa$

$$d_{\text{MI}} = \sqrt{2\varepsilon(-V_D - \delta/\kappa)}/\varepsilon\delta$$

and saturates at $V_D = -\delta/\kappa - \Delta$. When the MI layer saturate, the metallic region appears whose width is

$$d_{\text{M}} = \sqrt{\varepsilon/\kappa \cosh^{-1} \left[ \left\{ (V_D + \Delta)\delta/\kappa + \sqrt{2\Delta\delta/\kappa(V_D + \Delta + \delta/\kappa)^2 - 2(\delta/\kappa)(V_D + \delta/\kappa)} \right\} / \{2\Delta\delta/\kappa - (\delta/\kappa)^2 \} \right]$$

which saturates when $V_D < -\frac{1+\delta}{\delta} - \Delta$. Finally, the width of the BI layer is nonzero when $V_D < -\frac{1+\delta}{\delta} - \Delta$

$$d_{\text{BI}} = -F_2 + \sqrt{(F_2)^2 - 2\varepsilon(1+\delta)(1+\delta/\kappa + V_D + \Delta)}$$

with $F_1 = \sqrt{2\varepsilon\Delta/\varepsilon}$ and $F_2 = F_1 \cosh \left( \sqrt{\varepsilon k/\varepsilon d_M} \right)$

$$\sqrt{\varepsilon k/\varepsilon} \left( \frac{\delta}{\kappa} \right) \sinh \left( \sqrt{\varepsilon k/\varepsilon d_M} \right)$$

where $d_M$ here is obtained by substituting $V_D = -\frac{1+\delta}{\delta} - \Delta$ in eq. (4). In Fig. 2(b), we plot the $V_D$ dependence of the widths and compare them with the DMRG results, and both agrees surprisingly well. This agreement is highly nontrivial; In the insulating phase the localization length $\xi \sim W/\Delta(U)$ is of the order of few sites, but in the metallic phase it
should diverge. So, in the metallic phase the local approximation is not a priori justified. However, our numerical calculation shows that it works remarkably well. Based on this success, we apply this local approximation to more generic cases below.

![Diagram](image)

**Application to CER** — In CER devices, the interface between the electrode and SCES is two dimensional. In systems with dimensions higher than one, the filling effect between the electrode and SCES is two dimensional. A region near the transition point [15, 16, 17]. We expect similar hysteresis in the density-potential relation of the interface Mott transition (Fig. 3 (a)). This is the case, coexistence of the metallic state and the insulating state is possible. Then, the I-V characteristics of the device shows high non-linearity as well as a memory effect, i.e., RRAM behavior, when a layer of coexistence state exist in the SCES region. The condition for this to take place is \( V_D < - (\delta - \Delta \delta)/\kappa \). For example, if \( V_1 < V_D < V_\infty < V_2 \) is satisfied, the region nearest to the interface is a phase coherence layer, as shown in Fig. 3 (c) and (d). If this is the case, the junction may be insulating (c) or metallic (d). They correspond to the off and on states, respectively. We can switch between them by applying a voltage \( V_A \) on the electrode forming a hysteresis loop in the I-V characteristics Fig. 3 (b).

We assume a tunneling form for the current

\[ I(V_A) = T(V_A)J_0(V_A), \]

where \( T(V_A) \) is the tunneling probability of the barrier and \( J_0(V_A) \) the current at the metal/SCES junction without any barrier. \( J_0(V_A) \) reflects the details of the device and may be Ohmic, i.e. \( J_0(V_A) \propto V_A \), or if space charge limited current is realized, \( J_0(V_A) \propto (V_A)^2 \) [18]. We assume that the tunneling probability decreases exponentially as the width of the MI layer grows, i.e., \( T(V_A) = e^{-d_{MI}(V_A)/\xi} \), where \( \xi \) is the decay length (here the temperature dependence is neglected). Neglecting the jump of \( n \) in Fig. 3 (a), the width of the MI layer can be obtained by replacing \( -V_D - \delta/\kappa \) in eq. (4) with \( V_i - V_D - V_\infty - V_A \), \( i = 1, 2 \) for the on and off state, respectively. Thus, a hysteresis loop is realized in the I-V characteristics as in Fig. 3 (b). If we define the CER ratio by \( \Delta R/R \equiv (R_{\text{off}} - R_{\text{on}})/R_{\text{on}} \), where \( R_{\text{on,off}} \) is the resistivity of the on and off states, we obtain \( \Delta R/R = e^{(2V_2-V_D-V_\infty)/e\delta} - 1 \). In a Ti/PCMO based CER device, however, neither the on nor the off states show Ohmic I-V characteristics [8]. This can be also understood by our model with \( V_D + V_\infty < V_1 \). In such cases, the CER ratio becomes \( \Delta R/R = e^{(2(V_2-D-V_\infty)/e\delta)} - 1 \). In either case, we can design a CER device with larger CER ratio by decreasing the doping ratio \( \delta \) and making the phase coexistence region wider.

In summary, we have studied the interface Mott transition by the DMRG method and by Poisson’s equation combined with a local equilibrium ansatz. We proposed a novel mechanism of CER for materials with a first order metal-insulator transition.

We thank A. Sawa, I. H. Inoue, M. Kawasaki and Y. Tokura for illuminating discussions, and Y. Ogimoto for careful reading of the manuscript. TO acknowledges S. Onoda, and F. Ogushi for helpful comments.

[1] A. Asamitsu, Y. Tomioka, H. Kuwahara, and Y. Tokura, Nature 388, 50 (1997).
[2] V. Ponnambalam, S. Parashar, A. R. Raju, and C. N. R. Rao, Appl. Phys. Lett. 74, 206 (1999).
[3] H. Oshima, K. Miyano, Y. Konishi, M. Kawasaki, and Y. Tokura, Appl. Phys. Lett. 75, 1473 (1999).
[4] S. Q. Liu, N. J. Wu, and A. Ignatiev, Appl. Phys. Lett. 76, 2749 (2000).
[5] A. Baikalov, et al., Appl. Phys. Lett. 83, 957 (2003).
[6] A. Sawa, T. Fujii, M. Kawasaki, and Y. Tokura, Appl. Phys. Lett. 85, 4073 (2004).
[7] T. Hasegawa, K. Mattenberger, J. Takeya, and B. Bat-
logg, Phys. Rev. B 69, 245115 (2004).
[8] A. Ohtomo and H. Y. Hwang, Nature 427, 423 (2004).
[9] M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. 70, 1039 (1998).
[10] S. R. White, Phys. Rev. Lett. 69, 2863 (1992).
[11] M. J. Rozenberg, I. H. Inoue, and M. J. Sánchez, Phys. Rev. Lett. 92, 178302 (2004).
[12] K. Yonemitsu, J. Phys. Soc. Jpn. 74, to appear (2005).
[13] E. H. Lieb, and F. Y. Wu, Phys. Rev. Lett 21, 192 (1968).
[14] H. Schulz, Correlated electron systems V.J.Emery (ed) World Scientific (1992).
[15] D. S. Fisher, G. Kotliar, and G. Moeller, Phys. Rev. B 52, 17112 (1995).
[16] G. Kotliar, S. Murthy, and M. J. Rozenberg, Phys. Rev. Lett. 89, 046401 (2002).
[17] S. Onoda, cond-mat/0408207 (2004).
[18] M. A. Lampert, and P. Mark, Current Injection in Solids (Academic Press, 1970).