Filament Dynamics in Resistive Switching

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Abstract. The hallmark of nonequilibrium phase transitions is the strong spatial inhomogeneity associated with bi-stability of macroscopic phases. We consider the resistive switching in transition metal oxides as a prototypical nonequilibrium phase transition associated with an equilibrium phase transition. We analyze the behavior of the hysteretic behavior of I-V relations in the resistive switching, especially the negative differential resistance regime during the reverse sweep of applied voltage. We show that the peculiar I-V behavior is closely related to the bi-stability of the metallic and insulating phases and the dynamics of the metallic filament, and investigate the filament dynamics in terms of the minimum entropy production principle in the nonequilibrium theormodynamics.

1. Introduction

Controlling materials property with electromagnetic input is one of the central themes in modern condensed matter physics. We focus here on the phase transition in transition-metal oxides controlled by static bias applied across the sample, driving the system from an insulator to metal and vice versa. This phenomenon known as the resistive switching (RS) has been studied over the last half century, and the interest on this topic has piqued recently [1, 2, 3] due to its application possibilities as a resistive logic device. Production of commercial resistive memory is imminent. The systems exhibiting the resistive transition are quite diverse – ranging from intrinsic semiconductors to transition-metal oxides/chalcogenides (TMO/TMC) – with varying mechanisms with switching fields over the range of 1 ~ 1000 kV/cm.

The origin of the underlying RS mechanism in small-gap systems of TMO/TMC is very much debated until today. One of the main questions is the magnitude of the switching electric field for the insulator-to-metal transition (IMT) at the order of a few kV/cm, which translates to the energy scale of sub-meV of potential drop across a unit cell. From the point of the band-structure theory, this seems too small to create a fundamental transformation of electronic structure from an insulator to a metal. This raises a question of whether the appropriate approach is to consider the electric field as effectively coupled to collective degrees of freedom.

The scenarios for the RS can be divided into two categories: electronic and thermal mechanisms. The electronic scenario advocates direct action of electric field [4] through acceleration of electron kinetic energy directly impacting the electronic structure and leading to...
the resistive transition. Such scenarios include impact ionization, avalanche, electro-migration of metallic filaments. These mechanisms are considered fast in the regime of femto-seconds [5]. However, as pointed above, the predicted energy scales for the switching field are generally considered too large for the observed energy gap.

The other group of scenarios of thermal switching rely on the indirect role of the electric field via the hot-electron effect created by the electric field [6, 7, 8]. Electric field pumps energy into the system via coupling to electrons which then dump their excess energy to phonons. In a steady-state the heat balance is maintained between the power generated by the electric field and the energy dissipation into phonons whose energy is eventually dissipated into environment. At the nonequilibrium steady-state, the effective temperature of electrons, elevated from the bath temperature, is controlled by the electric field and a phase transition may take place due to this hot-electron temperature. This scenario has been advocated by many groups, in particular, with a result demonstrating that the hot-electron temperature at an RS in VO$_2$ matching the equilibrium critical temperature [6]. However, it is widely considered that the thermal process would be too slow to account for the RS phenomena and the controversies remain.

The goal of this theoretical effort is to provide a minimal dissipation model on gapped solids and reconcile the differences between the electronic and thermal RS scenarios based on a quantum mechanical microscopic model. Previous theories have largely been semi-classical resistive network model which set up a network of resistors following an empirical resistance $R(T)$ and establish local energy balance due to the Joule heating [1, 3]. This theory has been successful in reproducing the experimental $I$-$V$ relations, but is fundamentally incapable to address the mechanism of the RS.

2. Filament Dynamics and Nonequilibrium Steady-State

In the past few decades, the debate in the RS phenomena has centered around the question of whether the IMT (insulator-to-metal transition) is electronic or thermal. Recent experiments have supported both scenarios. The experiments supporting electronic mechanism have shown that the switching mechanism is ultra-fast and is independent of phonon-pumping [5]. We have shown recently [9] that the nonequilibrium excitation by electric field is given by the Landau-Zener excitations suggesting that the switching mechanism is fundamentally electronic. However, some experiments have made arguments supporting the thermal switching ideas with the most definitive argument coming from the local temperature measurement in the vicinity of metallic filament during the switching. This work showed that the hot-electron temperature matched the equilibrium critical temperature, which has also been confirmed by our previous work. The authors have concluded that the RS in ordered solids is fundamentally an electronic process while it permits a thermal interpretation.

To put the previous theoretical findings in a broader context of thermodynamics, we examine here the scenario of nonequilibrium pattern formation based on the nonequilibrium thermodynamics argument [10] of the minimum entropy production, following Ridley’s work [11]. In comparison with the numerical results, we find discrepancies from Ridley’s argument to suggest that the bulk resistivity has to be significantly modified to include the spatial variation of the order parameter to reproduce the microscopic theory prediction.

Let us first review Ridley’s seminal work [11] from 1963 for nonequilibrium pattern formation. The sample geometry is depicted in FIG. 1(a) where the so-called $S$-type material develops a metallic filament during the insulator-to-metal transition process. This is typical signature of spatial inhomogeneity in nonequilibrium phase transition. With the bulk resistivity relation empirically given by the resistance $R(T)$, as sketched in FIG. 1(b), between the electric field $E$ and the current density $J$, the current values are given as multi-valued at given electric field such as shown. For the middle solution in the region of the negative differential resistance (NDR, $dJ/dE < 0$), the bulk solution spontaneously breaks into spatially inhomogeneous regions.
Figure 1. (a) Schematic drawing of the resistive switching devices. Given an electric current $I$ the device supports bi-stability of the insulating and metallic phases with the metallic filament carrying the majority of the current. (b) Sketch of the bulk $I-V$ relation of the $S$-shape RS materials. For a given $I$, the electric field $E$ is determined to minimize the entropy production. For a current density $J$ (averaged over the sample) in the range $J_1 < J < J_2$, the multiple-phase solution at $E = E_m$ minimizes the Joule heating. (c) Sketch of theoretical $I-V$ from with a uniform order parameter. The (black) dashed part is only available in spatially inhomogeneous solution. The electric field which allows a stable uniform metallic solution is the $E_{MIT}$ at the metal-to-insulator transition.

with metallic and insulating mixed phases [11]. The metallic phase appears in the form of a conducting filament, as shown in (a).

The criterion for the multi-phase solution relies on the minimum entropy production principle of the nonequilibrium thermodynamics for a steady-state. Since the $S$-shaped curve is single-valued as a function of the current, we imagine that we map out the $I-V$ relation of the devices by fixing the total current flowing through the device, hence the name current-controlled NDR device [12]. The nonequilibrium thermodynamics has that the entropy change of the system is written in a continuity equation of entropy $S$ (per volume) as

$$\frac{dS}{dt} = -\nabla \cdot J_s + \dot{s},$$

(1)

where $J_s$ is the entropy flow and $\dot{s}$ is the entropy production. $J_s$ accounts for the heat flow into environment in the form of Joule heating and $\dot{s}$ is the entropy generation mainly due to the external force, and the temperature and chemical potential gradient. Ignoring the nonequilibrium effect of the interface between the metallic and insulating phases through the temperature and chemical potential gradient, we then express the entropy production in terms of the bulk power generation $EJ$. When multiple phases exist, the total $EJ$ is the sum from each phases. At a nonequilibrium steady-state $dS/dt = 0$ and the entropy flux and the entropy production are balanced out.
The solution for the filament formation is to find the minimum entropy production given the total current $I$ while the electric field is treated as adjustable. We also allow the fractional cross-section $a$ of the metallic phase to vary. Since the electric field inside the metallic and insulator is identical we have the entropy production as

$$T \dot{s} = E[J_1(1-a) + J_2a],$$

with the current density $J_1$ and $J_2$ inside the insulating and metallic phases, respectively. For current density in the interval $J_1 < J < J_2$ [with $J_1$ and $J_2$ defined by the NDR condition as marked as in (b)] a uniform phase is unstable, and the minimum electric field which allows multiple solutions is $E_m$ as shown in FIG. 1(b). At $E = E_m$, $J_1$ and $J_2$ are read off from the curve $R(T)$. By imposing the constraint

$$I/A = J_1(1-a) + J_2a,$$

with the device cross-section $A$, one can determine the filament cross-section $a$.

Let us examine this scenario for the filament formation based on the numerical microscopic calculations performed on finite lattices of dissipative tight-binding model with anti-ferromagnetic (AFM) ordering. The details of the model is described in Ref. [9]. The dissipation is introduced by the fermion bath model [13, 14] and the mean-field approximation is used to treat the anti-ferromagnetic correlation. Once the Hamiltonian is set, we have used the Keldysh Green’s function method to solve for the self-consistent condition for AFM order parameter.

The $I$ – $V$ characteristics as sketched in FIG. 1(c) describes the bulk calculations, with an infinite size lattice of uniform order parameter following the formalism in Ref. [15]. The red solid lines are metallic (top) and insulating (bottom) solutions. The black dashed line connecting the two phases is only accessible with mixed-phase solutions and a uniform bulk calculation is not accessible to this NDR part of the $I$ – $V$. As the electric field is turned on from zero, the current density follows the lower curve until the threshold field $E_{\text{IMT}}$ for the insulator-to-metal transition (IMT) and the system makes a strongly discontinuous transition to a metallic solution (following the red dashed line). Upon decreasing the field, the metallic state makes a transition at the field $E_{\text{MIT}}$ for the metal-to-insulator transition (MIT). The calculation showed that the nature of the two transitions are quite different with the threshold field dependence,

$$E_{\text{IMT}}/\Delta_0 \ll E_{\text{MIT}}/\Delta_0 \sim \Delta_0/t \ll 1.$$

The switching fields are much smaller than the energy scale of the energy gap, and in particular, the MIT field is orders of magnitude smaller than the IMT field.

Figure 2 shows the calculated $I$ – $V$ relations for a square lattice oriented in the $(1, 1)$ direction with respect to the electric field. The lattice of size $(60a)/\sqrt{2} \times (30a)/\sqrt{2}$ has $I$ – $V$ relation with good qualitative agreement with experiments on VO$_2$ [6]. The agreement includes the resistive hysteresis, existence of the NDR in the reverse voltage sweep, and the match of the hot-electron temperature at the RS with the equilibrium critical temperature. However, calculation shows a cusp in the $I$-$V$, between the points marked as 1 and 2 in (a). This is an artifact from a finite-size calculation. As shown in (c), as one passes from 1 to 2, the insulating phase begins to appear in from the edges of the sample. The black area is for zero gap (metal) with the arrow
Figure 2. $I-V$ relation and the domain dynamics for resistive hysteresis in the dissipative AFM lattice. (a) $I-V$ curve for a lattice size of $(60a)/\sqrt{2} \times (30a)/\sqrt{2}$. The two-dimensional square lattice is oriented in the (1,1) direction where its group velocity is aligned with the field direction. The IMT shows sensitive dependence on the impurity concentration $c$ while the MIT is nearly independent of impurities. The IMT and MIT threshold fields calculated for an infinite and uniform lattice are marked as $E_{\text{IMT}}^\infty$ and $E_{\text{MIT}}^\infty$, respectively. (b) $I-V$ for a $(120a)/\sqrt{2} \times (30a)/\sqrt{2}$. The part of the curve showing the NDR behavior extends further vertically, due to more room for filament to fit in the lattice. It also shows rough steps (and slight hysteresis) in the vertical part due to the pinning of order parameters by impurities. Otherwise, the overall behavior of the hysteresis is very similar to (a). (c) Snapshots of the domain structure on the reverse sweep of total voltage in (a) at $c = 0.03$ (blue curve). The black area is for zero gap (metal) and orange area for a finite gap (insulator), and the arrows denote the current density.

showing the current density. As we move from 2 to 3, the metallic region shrink like a thinning filament. This filament dynamics is confirmed in a larger calculation with twice the width of the previous calculation with $(120a)/\sqrt{2} \times (30a)/\sqrt{2}$, as shown in (b). The nearly vertical part of the NDR curve extends further into high current without the sign of a cusp. In this run, the filament emerges with a width smaller than the lattice width. A small hysteresis loop appeared in the vertical $I-V$ part, probably due to the pinning of domain walls by impurities. A similar behavior has been observed in $\text{V}_2\text{O}_3$ samples [8]. In the limit of large filament thickness, one expects that the bulk properties of the metallic and insulating domains may determine the mixed-phase properties, as argued by Ridley [11]. In the numerical results, the electric field indeed seems to approach a limit $E_m$, as marked in Figure 2(b). As seen in (a), the limit $E_m$ is insensitive to the impurity concentration, and by comparing with (b) $E_m$ does not depend strongly on the width of the system either. As the filament narrows, the $I-V$ deviates from the vertical line and it is speculated [9] to come from inter-domain-wall scattering of electrons, leading to an increased voltage for thinner filament, i.e., smaller current.

However, the value of $E_m$ does not agree with the value derived from Ridley’s argument as discussed in Figure 1. Based on the minimum entropy production principle, $E_m$ is expected to be the minimum electric field which stabilizes a metallic phase, that is, $E_{\text{MIT}}^\infty$. As shown in Figure 2(a), $E_{\text{MIT}}^\infty$ is smaller than calculated $E_m$ roughly by an order of magnitude. As discussed above, the reason for the discrepancy is not the impurities nor the size of the system. We speculate that the problem lies in the negligence of the nonequilibrium flow of entropy due to
the temperature and chemical potential, along with the energy cost due to the order parameter stiffness. It is suggestive that the domain walls grow out of the sample edges where the energy cost can be minimized. Therefore, the insulating phase may be created from the edges with lowered energy barrier.

3. Conclusion
Nonequilibrium quantum phase transition is a tremendous challenge in coming ages of condensed matter physics. We have provided some key understanding to the long-standing problem of resistive switching (RS) in ordered solids, and resolved an important issue regarding the mechanism of the RS. Quantum modeling showed that the segregation of quantum phases driven by nonequilibrium is critical to understand the nonequilibrium physics. This also indicates, however, that such calculations are going to be computationally costly, if the theory only relies on the Keldysh diagrammatics. In this work, we have tested the RS switching condition calculated from the Keldysh theory with the nonequilibrium thermodynamics argument based on the minimum entropy production principle. The discrepancy indicates that microscopic understanding of nonequilibrium entropy is essential before we begin to apply the rules of the nonequilibrium thermodynamics in quantum systems.

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References
[1] Janod E, Tranchant J, Corraze B, Querré M, Stoliar P, Rozenberg M, Cren T, Roditchev D, Phuoc V T, Besland M P and Carlo L 2015 Adv. Funct. Mater. 25 6287–6305
[2] Stoliar P, Carlo L, Janod E, Corraze B, Guillot-Deudon C, Salmon-Bourmand S, Guiot V, Tranchant J and Rozenberg M 2013 Adv. Mater. 25 3222–3226
[3] Lee J S, Lee S and Noh T W 2015 Appl. Phys. Rev. 2 031303
[4] Mayer B, Schmidt C, Grupp A, Bühler J, Oelmann J, Marvel R E, Haglund R F, Oka T, Brida D, Leitenstorfer A and Pashkin A 2015 Phys. Rev. B 91 457
[5] Jager M F, Ott C, Kraus P M, Kaplan C J, Pouse W, Marvel R E, Haglund R F, Neumark D M and Leone S R 2017 Proc. Nat. Aca. Sci. 114 9558–9563
[6] Zimmers A, Aigouy L, Mortier M, Sharoni A, Wang S, West K G, Ramirez J G and Schuller I K 2013 Phys. Rev. Lett. 110 056601
[7] Singh S, Horrocks G, Marley P M, Shi Z, Banerjee S and Sambandamurthy G 2015 Phys. Rev. B 92 155121
[8] Guénon S, Scharinger S, Wang S, Ramirez J G, Koelle D, Kleiner R and Schuller I K 2013 EPL (Europhysics Letters) 101 57003
[9] Li J, Aron C, Kotliar G and Han J E 2017 Nano Lett. 17 2994
[10] de Groot S R and Mazur P 1984 Non-equilibrium Thermodynamics (New York: Dover Publications)
[11] Ridley B K 1963 Proc. Phys. Soc. 82 954
[12] Kim H T, Kim B J, Choi S, Chae B G, Lee Y W, Driscoll T, Qazilbash M M and Basov D N 2010 J. Appl. Physics. 107 023702
[13] Han J E 2013 Phys. Rev. B 87 085119
[14] Han J E and Li J 2013 Phys. Rev. B 88 075113
[15] Li J, Aron C, Kotliar G and Han J E 2015 Phys. Rev. Lett. 114 226403