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Supporting Information

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Universal Electric-Field-Driven Resistive Transition in Narrow-Gap Mott Insulators

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Universal electric-field-driven resistive transition in narrow gap Mott insulators

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1 Experimental details

The samples used for electric pulses experiments are single crystals (typical sizes 200x200x50 μm^3). The electrode contacts were made on freshly cleaved surfaces using gold wires and carbon paste (Electrodag PR-406). A subsequent annealing in vacuum at 150 °C during 30 min was necessary to achieve good contact resistances. We checked that the contact resistances were much smaller than the sample resistance. Voltage pulses were applied using an Agilent 8114A. During the pulses, the voltage and current across the sample were measured with a Tektronix DPO3034 oscilloscope associated with a IeS-ISSD210 differential probe (see Ref. [S1] for details). All three systems are narrow gap Mott insulators. Their resistivity, measured as a function of temperature using a Keithley 236 source-measure unit, is shown in Figure S1.

The experimental temperatures for our experiments were chosen so to have a large ON/OFF resistance ratio and optimize the signal. Electric pulses were applied in the paramagnetic Mott insulating phases of $GaTa_4Se_8$ (at 77 K) and $V_{2-x}Cr_xO_3$ (at 164 K), and in the antiferromagnetic Mott insulating phase of $NiS_{2-x}Se_x$ (at 4 K).

The voltage thresholds were defined by the sudden change of slope in the V_S vs t data shown in Figure 1. The experimental investigation of the applied voltages close to V_{th} is complicated by the long delay times t_d, which result in the smearing of the resistive transition. This is likely due to heating effects of the sample. Nevertheless, those effects become negligible as soon as the applied voltage on the sample V_S gets well above V_{th} and t_d becomes short (in the order of tens of μsec).

2 Numerical simulations

2.1 Algorithm

We model the active layer as an array of cells arranged in a square lattice. Each cell corresponds to a small portion of the sample such that its electronic state can be well defined. We assume that each cell can be in either a correlated metal (CM) or a Mott insulator state (MI). In this way, we turn the problem into a site percolation problem, *i.e.*, the Mott resistive transition occurs when the CM sites form a percolation path.[S2, S3] The dynamical character of the system (evolution in time) is simulated by switching back and forth the states of the cells according to the transition probabilities given by $P_{MI\rightarrow CM}$ and $P_{CM\rightarrow MI}$. Thus the external voltage applied on the network creates local electric fields that spatially modulate the fraction of CM cells by means of $P_{MI\rightarrow CM}$.

In contrast to uncorrelated percolation process, here there are strong correlations that build up in the local transition probabilities, by way of the resistor network. In fact, as a segment becomes CM, then its voltage drop decreases with the concomitant increase of the potential differences on the sites neighbor to the segment. This promotes their successive MI to CM transition, and eventually gives place to an avalanche and a percolation path. Resistor-network-based numerical simulations are an efficient method to study percolation problems involving electrical conduction [S4], as analytical approaches are often impractical. Each cell in the network has a central node with four resistors that connect it with the first-neighbors, as depicted in Figure 2 (in the manuscript). When the cell is in CM state, all its four resistors adopt the value R_{CM}, whereas they are set to R_{MI} when the cell is in MI state.

The time is discretized in timesteps and the external voltage $V(t)$ is applied at each interval. Then the following procedure is followed at each step:

1. The distribution of voltages in all the nodes of the resistor network is calculated by nodal analysis.

2. The local voltage drops $\Delta V = \left((V_1 - V_3)^2 + (V_2 - V_4)^2 \right)^{1/2}$ are calculated for each cell, where $V_1 ... V_4$ are defined in each cell as depicted in Figure S3. With these ΔV's the local transition rates $P_{\mathrm{MI}\rightarrow\mathrm{CM}}$ and $P_{\mathrm{CM}\rightarrow\mathrm{MI}}$ are recomputed.

3. At every cell a transition is attempted by acceptance-rejection sampling [S5] using $P_{\mathrm{MI}\rightarrow\mathrm{CM}}$ and $P_{\mathrm{CM}\rightarrow\mathrm{MI}}$. Specifically, if the cell is in MI state and $rnd < \nu e^{-(E_B - q\Delta V)/kT}$ it is switched to the CM state. Where *rnd* is the uniform distribution in the interval [0:1). Similarly, a cell in the CM state is switched to MI if $rnd < \nu e^{-(E_B - E_{CM})/kT}$.

4. The values of the resistors of the network are updated according to the state of the cells that have changed.

2.2 Parameters

The number of cells in the network is 128 x 40. This size is the maximal one that allows for a reasonable simulation time to perform the full study. All the simulations presented in this work where done with the following choice of parameters. The sole exception is the multipulse simulations in Figure 5, as will be indicated later.

The unit of energy is adopted as kT=1 and the attempt rate ν that enters in the probabilities $P_{\mathrm{MI}\rightarrow\mathrm{CM}}$ and $P_{\mathrm{CM}\rightarrow\mathrm{MI}}$ is set to unity.

The energy of the states are E_{MI} = 0 (reference state), then E_{CM} =10 kT and E_B =20 kT, which provides reasonable simulation times of about 10^4 - 10^5 timesteps.

The load resistor in the electric circuit of the network is taken as unity. We set R_{MI} = 16, which gives an equivalent total resistance to the network of a similar order, and roughly mimics the actual experimental situation. We set R_{CM} = 0.3, which makes $R_{\mathrm{MI}} >> R_{\mathrm{CM}}$ according to the assumptions of a Mott insulator and a correlated metal states.

In the case of the study of the multipulse experiment shown in Figure 5, the simulations require a rather long computational time, specially, Figure 5d. Therefore, we set E_{CM} =15 kT and the resistances R_L= 2, R_{MI} = 16 and R_{CM} = 0.01.

2.3 Additional Figures

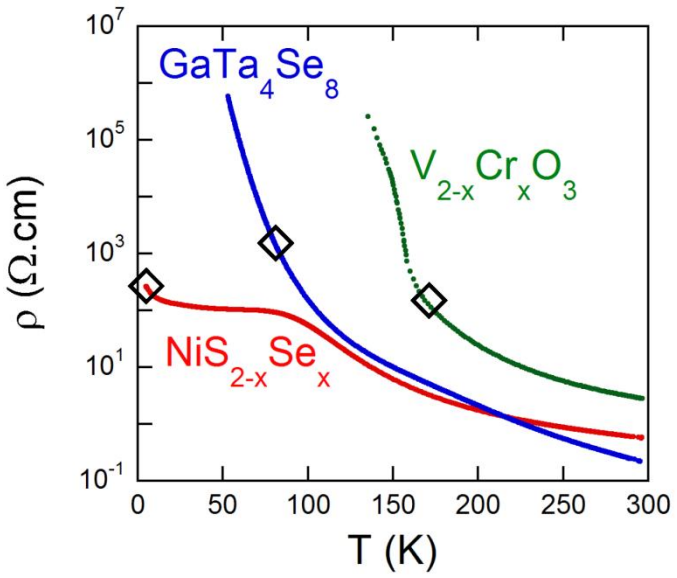

Figure S1. Resistivity as a function of temperature for the three experimental samples of our study. Open diamonds indicate the temperature of application of electric pulses for each compound.

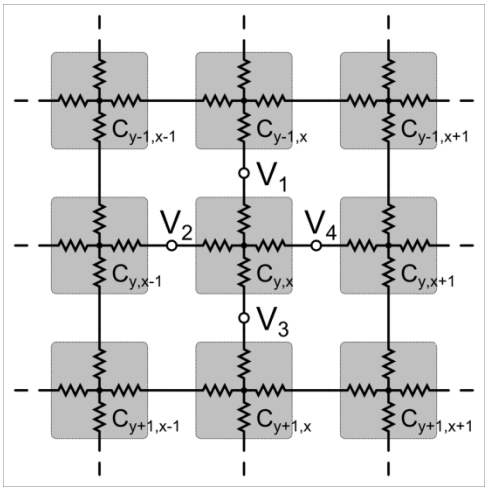

Figure S2. Detail of the resistor configuration of each cell and the inter-cell connections.

2.4 Supporting Videos

The following video files containing animations of the simulations are included in the repository `https://www.dropbox.com/sh/l2qkrit45tgt9zf/0yBZWy-ueb`

- "videoFig3"
- "videoFig4a"
- "videoFig4b"
- "videoFig4c"
- "videoFig4d"

Additional References

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[S5] S. Chib, E. Greenberg, *The American Statistician* **1995**, *49*, 327–335.