

## Field-Driven Mott Gap Collapse and Resistive Switch in Correlated Insulators

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Mott insulators are “unsuccessful metals” in which Coulomb repulsion prevents charge conduction despite a metal-like concentration of conduction electrons. The possibility to unlock the frozen carriers with an electric field offers tantalizing prospects of realizing new Mott-based microelectronic devices. Here we unveil how such unlocking happens in a simple model that shows the coexistence of a stable Mott insulator and a metastable metal. Considering a slab subject to a linear potential drop, we find, by means of the dynamical mean-field theory, that the electric breakdown of the Mott insulator occurs via a first-order insulator-to-metal transition characterized by an abrupt gap collapse in sharp contrast to the standard Zener breakdown. The switch on of conduction is due to the field-driven stabilization of the metastable metallic phase. Outside the region of insulator-metal coexistence, the electric breakdown occurs through a more conventional quantum tunneling across the Hubbard bands tilted by the field. Our findings rationalize recent experimental observations and may offer a guideline for future technological research.

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*Introduction.*—The conventional description of the electric breakdown, i.e., the field-driven formation of a conductive state in an otherwise insulating system, is based on the well-known Landau-Zener mechanism of quantum tunneling across the insulating gap [1,2]. However, the existence of a “rigid” band gap to be overcome by the field sets a lower bound of the threshold field and limits the density of excited carriers promoted across the gap. This ultimately leads to large density fluctuations at the nanoscale, one of the bottlenecks in device miniaturization [3]. These limitations are intrinsic in band insulators, whose gap is fixed as long as the chemical composition and lattice structure do not change.

Because of the collective nature of their gap, Mott insulators have recently emerged as ideal candidates to overcome the above issues, providing a potential alternative to semiconductor-based microelectronics [3,4]. The possibility of driving a gap closure in a Mott insulator by means of an electric field could enable us to access a much larger carrier density than in semiconductors, potentially overcoming most limitations of conventional devices.

Experimental evidences for such an appealing field-driven *resistive switch* have been recently found in several Mott insulators [5–12] and Mott-based devices [13,14]. Remarkably, these experiments ubiquitously report a whole novel scenario for the electric breakdown that cannot be reconciled with the standard Landau-Zener description. For instance, the breakdown occurs through the abrupt formation of a conductive channel at anomalously small threshold fields [5] as opposed to the smooth activation

at fields of the order of the gap as predicted by across-gap tunneling. In addition, VO<sub>2</sub> electric double-layer transistors formed at a solid-electrolyte interface show massive conducting channels that suddenly open up above a threshold gate voltage, with an extension much beyond the fundamental electrostatic screening length [13,14].

While several mechanisms of resistive switch beyond the tunneling paradigm are known, including thermally driven electric breakdown [15,16], valence-change-driven insulator-to-metal transitions [17,18], and resistive transitions generated by the nonlinear propagation of ionic defects [19], we believe that the above experiments still suggest another possible scenario of a genuine insulator-to-metal Mott transition triggered by the external field, as proposed in Refs. [5,6] and modeled phenomenologically through a resistor network mimicking the competition between a stable insulating phase and a metastable metallic one. Nonetheless, a true microscopic description in model Mott insulators is still lacking. Indeed, theoretical studies within the single-band Hubbard model, the paradigm of strongly correlated systems, have so far highlighted a breakdown that is essentially due to the tunneling across the Mott gap, as if the latter were as rigid as the band gap in semiconductors [20–24]. Even if in some cases the tunneling breakdown can be anticipated by the formation at large fields of in-gap states due to the Wannier-Stark effect [25–27], still the agreement with the above experiments remains poor.

Motivated by the strong evidence that the first-order character of the Mott transition plays a major role in the

aforementioned experiments [5,6,13], in this Letter, we explore the route to the electric breakdown that opens whenever a stable Mott insulator coexists with a metastable metal which is not connected to the insulating solution.

We realize this situation including extra degrees of freedom to the Hubbard model [28,29], and we choose an orbital degree of freedom which is ubiquitously relevant in actual Mott insulating materials and increases the coexistence region with respect to the single-orbital model. In particular, we consider the simplest modeling of a Mott insulator with a  $d-d$  gap [30], which we study in a slab geometry and in the presence of a constant electric field, i.e., an open circuit setup mimicking a FET [13] with a gate voltage but without a source-drain bias.

We show that, within the insulator-metal coexistence region, an electric field can drive a discontinuous transition from the insulator to a gap-collapsed metal at threshold fields much smaller than those expected in a Zener breakdown.

*Model.*—We consider a half-filled two-orbital Hubbard model in a three-dimensional lattice and in the presence of a crystal field  $\Delta > 0$  that lifts the orbital degeneracy. The generic Hamiltonian reads

$$\mathcal{H} = \sum_{\mathbf{k}\sigma} \sum_{\alpha,\beta=1}^2 t_{\mathbf{k}}^{\alpha\beta} c_{\mathbf{k}\alpha\sigma}^\dagger c_{\mathbf{k}\beta\sigma} - \frac{\Delta}{2} \sum_i (n_{i,1} - n_{i,2}) + \frac{U}{2} \sum_i (n_i - 2)^2, \quad (1)$$

where  $n_{i,\alpha} = \sum_{\sigma} c_{i\alpha\sigma}^\dagger c_{i\alpha\sigma}$  and  $n_i = \sum_{\alpha} n_{i,\alpha}$  are density operators at site  $i$ . At  $U = 0$  and for not too large  $\Delta$ , the model (1) describes a two-band metal. At large  $U$ , the ground state is a nonmagnetic Mott insulator, with one filled and one empty orbital. Such an insulating state is stable against spin and/or orbital ordering due to the finite value of  $\Delta$ . This is in sharp contrast to the single-band Mott insulator description, in which the extensive spin entropy inevitably favors the onset of a magnetic order. Although the presence of a symmetry-broken phase at weak or intermediate coupling in model (1) cannot be excluded, its very existence and its properties critically depend on the model details. Therefore, to maintain the discussion as general as possible, in what follows we shall not account for possible broken symmetry phases [31].

In order not to weigh the Hamiltonian with too many parameters, we choose for our analysis  $t_{\mathbf{k}}^{11} = t_{\mathbf{k}}^{22} = -2t(\cos k_x + \cos k_y + \cos k_z)$ , the intraorbital dispersion on a three-dimensional cubic lattice, and  $t_{\mathbf{k}}^{12} = t_{\mathbf{k}}^{21} = v(\cos k_x - \cos k_y) \cos k_z$ , a nonlocal hybridization that leaves the local single-particle density matrix diagonal in the orbital index even though the occupation of each orbital is not a conserved quantity. We observe that, because of our very specific choice, the model possesses an orbital  $U(1)$  symmetry that can be broken by a Stoner instability at weak

coupling [32]. However, as we previously mentioned, we shall restrict our analysis only to the symmetry invariant subspace. The energy unit is such that the intraorbital hopping is  $t = 0.5$  and we take  $v = 0.25$ . We solve the model by means of the dynamical mean-field theory (DMFT) [33,34] using an exact diagonalization solver (see Supplemental Material [35]).

Under the above assumptions, the model (1) undergoes a first-order Mott transition at a critical value of  $U = U_c$  monotonically decreasing with increasing  $\Delta$  [see Fig. 1(a)]. For the sake of definiteness, in the following we fix the crystal-field splitting to  $\Delta = 0.4$ , for which  $U_c \approx 8.05$ . For  $U_c < U < U_s \approx 8.3$ , the insulating solution is stable, but the metal continues to exist as a metastable solution up to the spinodal point  $U_s$ . The state variable that better characterizes the transition is the orbital polarization  $m = n_1 - n_2$ , i.e., the population imbalance between the lower and upper orbitals. At  $U = 0$ , the model describes a partially polarized metal ( $m < 2$ ). A finite interaction  $U$  reduces the effective bandwidth and induces a repulsion between occupied and unoccupied states, leading to an effective enhancement of the crystal field  $\Delta_{\text{eff}} > \Delta$  that increases the orbital polarization  $m$ . At the first-order transition, the metal turns abruptly into an almost fully polarized insulator ( $m \approx 2$ ) [36,40], with a finite gap separating the occupied lowest band from the empty upper one [35], a sort of Mott insulator “disguised” as a conventional band insulator.

In order to study the effect of an applied electric field, we consider a layered slab of our idealized material subject to a static and uniform electric field  $E = \Delta V/N$  (Coulomb gauge) directed along the slab direction [see Fig. 1(b)], namely, an open circuit configuration with  $\Delta V$  playing the role of a gate voltage.

*Results.*—Starting from the equilibrium insulator at  $U > U_c$ , we increase the electric field  $E$  and monitor the ground state evolution until, above a threshold value  $E_{\text{th}}$ , a *conducting* state with a finite density of states at the Fermi level is established throughout the slab. We find that such a field-induced conducting state has strikingly different properties depending on whether the insulator at zero field is within the coexistence region,  $U_c < U \lesssim U_s$ , or outside,  $U > U_s$ . This is highlighted by Figs. 1(c) and 1(d), showing the electric-field dependence of the layer-resolved spectral density outside [Fig. 1(c)] or inside [Fig. 1(d)] the coexistence region. In both cases, we show the spectra immediately before and after the formation of the conducting state.

Outside coexistence, i.e.,  $U > U_s$  [Fig. 1(c)], we observe a marked tilting of lower (LHB) and upper (UHB) Hubbard bands, just as we would expect in a band insulator. When the field is large enough, the LHB crosses the Fermi energy at the left side of the slab, which is thus doped with holes, while the UHB does the same at the right side, which is instead doped with electrons. As a result, two facing and

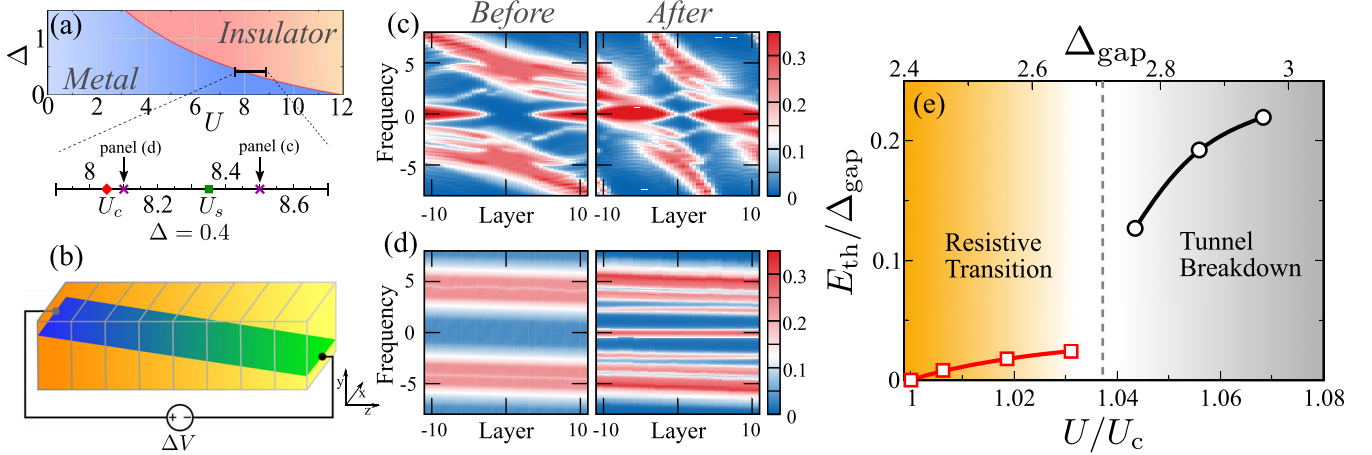


FIG. 1. (a) Zero-field phase diagram. Schematic representation of the equilibrium phase diagram. The enlargement at  $\Delta = 0.4$  highlights the range of the interaction relevant for this study.  $U_c$  marks the Mott transition critical value (red diamond).  $U_s$  marks the spinodal point (green square). The arrows indicate the interaction strengths used in (c) and (d) (purple crosses). (b) Sample geometry. The sample is a layered slab subject to a linear voltage drop  $\Delta V$ , corresponding to a uniform and static electric field along the slab direction. (c),(d) Electric-field-induced insulator-to-metal transition. Layer-resolved local spectral densities before (left) and after (right) the field-driven insulator-to-metal transition for two values of the interaction strength:  $U = 8.5$  (c) and  $U = 8.1$  (d), respectively, outside and inside the coexistence region. The intensities of the applied fields are  $E = 0.2$  (left) and  $E = 0.6$  (right) for (c) and  $E = 0.01875$  (left) and  $E = 0.025$  (right) for (d). (e) Electric-field versus interaction phase diagram. Threshold field  $E_{\text{th}}$  in units of the zero-field insulating gap  $\Delta_{\text{gap}}$  versus  $U/U_c$ . We also show  $\Delta_{\text{gap}}$  for each value of  $U/U_c$  (upper x axis).

oppositely charged surface layers with finite spectral weight at the Fermi level appear [35], whose thickness grows with the field until, above the threshold value  $E_{\text{th}}$ , they touch in the center of the slab. Before touching, the two surface layers are tunnel coupled through an insulating barrier, the central part of the slab [35], and only above  $E_{\text{th}}$  is a sizable density of states at the Fermi level established throughout all the sample. We note that this behavior is essentially that originally uncovered by Zener [1] in the Coulomb gauge, though here in an open circuit configuration, as already discussed in the case of Mott insulators [20,41].

A completely different behavior arises instead when the system is perturbed within the coexistence region between the metal and insulator ( $U_c < U \lesssim U_s$ ). In this case, a sharp insulator-to-metal transition instead takes place at the threshold field  $E_{\text{th}}$ . This is illustrated by the abrupt change of the layer-resolved spectral density across the resistive transition reported in Fig. 1(d). As the threshold field is crossed, the gap abruptly collapses and a sizable spectral weight at the Fermi level appears. The absence of any precursor on the insulating side indicates that the system displays a true resistive switch in which the ground state sharply changes from an insulator to a metal. As opposed to the previous case, the field-induced metal is homogeneous with a sizable spectral weight quite uniform throughout the whole slab [35].

In Fig. 1(e), we plot the threshold field  $E_{\text{th}}$  in units of the zero-field insulating gap  $\Delta_{\text{gap}}$  as a function of the distance from the first-order Mott transition. A clear break is observed in the curve around  $U \approx 1.04U_c \approx U_s$  separating

the two distinct regimes. The threshold field is small and slowly increasing within the coexistence region, while it gets larger and more steeply increasing outside. Remarkably, the strong variation of  $E_{\text{th}}$  is in sharp contrast to the weak variation of the gap.

It is evident that the low-field resistive transition not extending beyond the spinodal point  $U_s$  cannot be accidental. Indeed, as neatly shown in Fig. 2(a), the field-induced metallic phase is adiabatically connected with the metastable metal at zero field. At  $E = 0$ , the insulating solution has lower internal energy and is separated from the metastable metal by a small energy difference. However, the metallic solution has a high electrical polarizability, and it gains more energy from the field  $E$  than the incompressible Mott insulator, whose energy remains essentially constant. As a consequence, the two energy curves eventually cross at the threshold field  $E_{\text{th}}$ . The first-order character of such a field-driven insulator-metal transition is further highlighted by the hysteresis loop of the equilibrium orbital polarization  $m$  across the Mott transition, plotted in Fig. 2(b). The electric field thus acts as a switch between two phases with no adiabatic connection across the first-order metal-insulator transition [42].

Despite the field  $E$  having the dramatic role of driving the first-order insulator-metal transition, from the viewpoint of the metastable metal it is just a weak perturbation whose effects are accounted by the linear response theory. Indeed, the energy of the metastable metal is well fitted with a simple parabola [see Fig. 2(a)] as predicted by the linear response theory,  $\langle H \rangle(E) = \langle H \rangle(0) - \chi E^2/2$ , where  $\chi$  is the polarizability whose estimate is  $\chi \sim 5.46$ . This is important

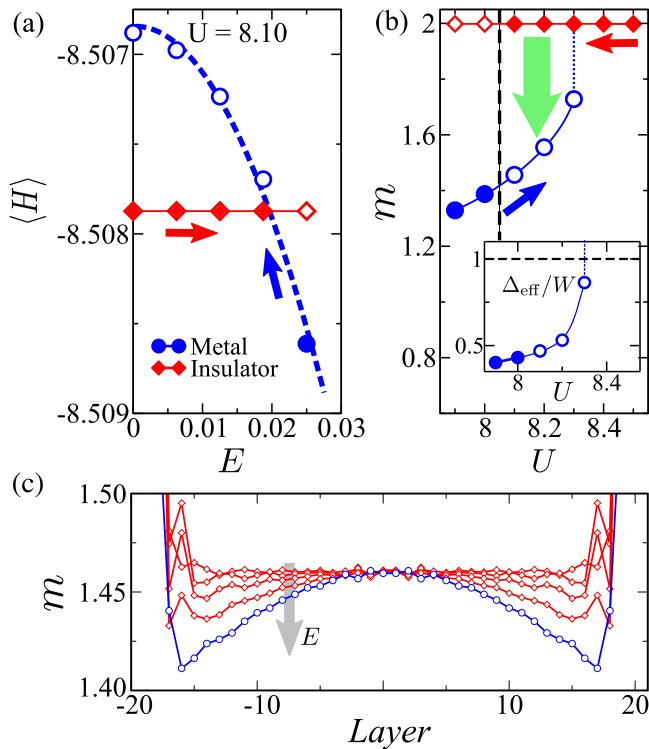


FIG. 2. Metal-insulator coexistence. (a) Internal energy  $\langle H \rangle$  of the metallic (blue circles) and the insulating (red diamonds) solutions, as a function of the electric field  $E = \Delta V/N$ . The solid (open) symbols mark the stable (metastable) character of the solution for each value of  $E$ . The dashed blue line is a quadratic fit to the energy of the metallic solution (see the text). The red full line is a guide to the eye. (b) Hysteresis loop for the orbital polarization  $m = n_1 - n_2$  across the zero-bias Mott transition. The red and blue arrows define the directions of continuous evolution for the insulating and metallic phases, respectively. The big green arrow indicates the electric-field-induced switch between the stable insulator and the metastable metal. Lines are guides to the eye. Inset: Effective crystal field  $\Delta_{\text{eff}}$  in units of the bandwidth  $W$  as a function of the correlation strength  $U$ . The metallic state is destabilized for  $\Delta_{\text{eff}} > W$ . (c) Polarization profile for the metastable metal for increasing electric field strength (gray arrow). Red diamonds (blue circles) refer to a system with an insulating (metallic) ground state.

for what concerns the relevance of our results once the circuit were closed or a weak probing bias applied, e.g., perpendicularly to the slab axis. Indeed, in the linear response regime we do not expect any prominent effect caused by a finite current flow [24] that could alter completely the physics with respect to the open circuit case.

In Fig. 2(c), we show that, as long as the system is within the coexistence region ( $U < U_s$ ), the energy gain is accompanied by a significant reduction of the value of  $m$  in the metastable metal, still in the linear response regime apart from finite size effects at the boundaries. This is remarkable, since it appears without a strong charge redistribution across the sample [35]. In other words, the net effect of the electric field is essentially to decrease the

orbital polarization of the metallic state, effectively moving in the phase diagram of Fig. 1(e) as if  $U$  or  $\Delta$  were reduced. In such a situation, the threshold field depends only on the metal polarizability, and we expect that it remains constant as a function of the slab length until the potential at the boundaries becomes so large that nonlinear effects appear, so that coupling with isopotential sources might become crucial to observe the same physics, an event we do not explore here.

Outside the coexistence region ( $U > U_s$ ), where a metastable metal no longer exists, the field modifies the insulating state through a strong charge redistribution, ultimately leading to the formation of the highly inhomogeneous conductive state in Fig. 1(c). The closure of the circuit in this case will induce more dramatic effects, even though we still expect the overall picture of a Landau-Zener tunnel breakdown to hold, possibly accompanied by other emergent effects [26,27].

*Conclusions.*—The above results unveil a so far unexplored pathway to the metallization of a correlated insulator, where the electric field drives a first-order transition from the insulator to a gap-collapsed metal phase, preexisting as a metastable state at zero field. Although the condition of being inside the insulator-metal coexistence region might be considered a rare circumstance in reality, we note that observed Mott transitions are often characterized by quite wide hysteresis loops; see, e.g., [43] in the popular case of  $V_2O_3$ .

Moreover, those results help clarify why a similar phenomenon has not yet been observed, at least within the DMFT, in the single-band Hubbard model, despite the latter also showing metal-insulator coexistence. In this model, in the whole coexistence region the metal differs from the insulator only by the presence of a narrow quasiparticle peak at the Fermi level that hosts only a tiny percentage of carriers. For the same reason, the region of parameters where a stable insulator coexists with a metastable metal is extremely narrow, actually vanishing at zero temperature, so that one ends up observing always the same behavior in a field as that of our model beyond the spinodal line [35].

Notwithstanding the obvious importance of the size of the coexistence domain on the insulating side of the Mott transition, we believe that the key issue to observe a genuine resistive switch is the competition of two neatly distinguishable phases, characterized by sharply different values of an extensive observable, in our case the orbital polarization  $m$ , which can be tuned by the external field. In turn, this property might also be the rationale of the abrupt gap collapse at the resistive transition, which reflects the very nature of the gap [29]. In our Mott insulator model, the gap separates two bands of different orbital character; i.e., it refers to the cost of redistributing electrons among the valence orbitals without changing valence, as opposed to the gap between the lower and upper Hubbard bands in an

idealized Mott insulator, which refers instead to the cost of changing the total charge within the valence shell.

A similar mechanism could be active in several known Mott insulators where the “polarization” of some degree of freedom starkly distinguishes the metal from the insulator. The physical nature of such an observable can be material dependent—for instance, in the case of VO<sub>2</sub> [44–46] or V<sub>2</sub>O<sub>3</sub> [47,48], it is supposed to be the relative occupancy of two different *d* orbitals, much like our simple case study—yet the correlation-driven Mott transition would be associated to a sharp change of its value. In such a context, the application of an external field would change this polarization favoring the metallic state and ultimately driving the resistive transition.

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