

Percolative transitions with first-order characteristics in the context of colossal magnetoresistance manganites

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The unusual magnetotransport properties of manganites are widely believed to be caused by mixed-phase tendencies and concomitant percolative processes. However, dramatic deviations from “standard” percolation have been unveiled experimentally. Here, a semiphenomenological description of Mn oxides is proposed based on coexisting clusters with *smooth* surfaces, as suggested by Monte Carlo simulations of realistic models for manganites, also briefly discussed here. The present approach produces fairly abrupt percolative transitions and even first-order discontinuities, in agreement with experiments. These transitions may describe the percolation that occurs after magnetic fields align the randomly oriented ferromagnetic clusters believed to exist above the Curie temperature in Mn oxides. In this respect, part of the manganite phenomenology could belong to a new class of percolative processes triggered by phase competition and correlations.

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I. INTRODUCTION

The understanding of the self-organized behavior of transition-metal oxides, such as manganites and cuprates, has developed into one of the dominant scientific themes of condensed-matter physics. Phase coexistence and competition between different kinds of orders involving charge, orbital, lattice, and spin degrees of freedom leads to physical “complexity” as a characteristic of their behavior. The balance between competing phases is subtle, and small changes in composition, magnetic fields, or temperatures lead to large changes in the material properties. In the context of colossal magnetoresistance (CMR), these phenomena motivated the nanoscale phase separation theory of manganites,¹ and its concomitant percolative transition from the metal to the insulator. Many experiments have reported results compatible with phase separation and percolation in Mn oxides.^{2–4} Recently, CMR has been observed in simulations of simple models of competing phases, between T_C , the ordering temperature, and $T^* > T_C$, the temperature where coexisting clusters start developing upon cooling.⁵ In the presence of magnetic fields, the preformed ferromagnetic (FM) clusters align their magnetic moments, and they percolate. This encouraging agreement between theory and experiments suggests that the basic concept behind the CMR phenomenon—phase competition—has been unveiled.

However, several experimental facts still await for a rationalization within the phase-separation scenario. Distinct among them is the abrupt character of the metal-insulator transition (MIT) in some manganites, which is incompatible with *standard* percolative phenomena where the transition is smooth. The results in Fig. 1 illustrate this paradox: (a) In $\text{La}_{(5/8-y)}\text{Pr}_y\text{Ca}_{3/8}\text{MnO}_3$, the residual resistivity ρ_{dc} at zero temperature T changes fast with y , with a conductivity exponent t much larger than predicted by standard percolation.² (b) Far more challenging, ρ_{dc} of $\text{Sm}_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ changes

from insulator to metal in a *first-order* transition.⁶ Although most Mn oxides have continuous MITs, their ρ_{dc} vs T curves often show rapid changes and hysteresis loops, in the same regime where cluster percolation seems to occur. (c) Varying magnetic fields, $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ also presents a first-order-like MIT.⁷ These abrupt transitions cannot be rationalized within standard percolation theory,⁸ but they are found in regimes that exhibit some aspects of percolation.⁴

The theory recently presented by Burgy *et al.*⁵ relies on a CMR state above T_C made out of preformed FM clusters, with random magnetic-moment orientation (a similar picture was also qualitatively discussed by Uehara *et al.*²). This inhomogeneous state is stable due to the presence of quenched disorder. The random orientation of the cluster magnetic moments can be achieved by having “walls” between them made out of the competing insulating phase. Otherwise,

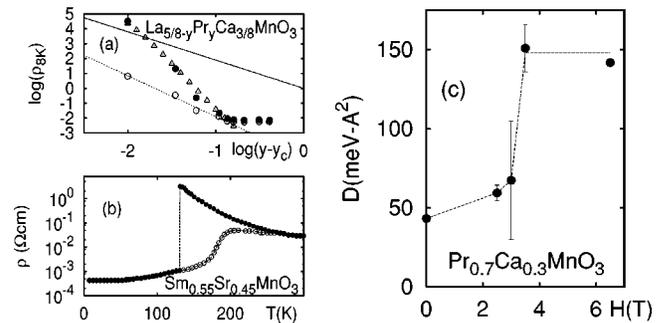


FIG. 1. Results illustrating the “abrupt” character of the metal-insulator transition in some manganites (compositions indicated). (a) Dependence of residual ρ_{dc} on chemical substitution at 0 (●) and 4 kOe (○) (Ref. 2). The solid line represents the prediction of standard percolation, while the dotted one is a guide to the eye. Δ shows results of a simulation using a broad distribution of bonds ($\alpha=0.857$). (b) Resistivity vs T , at magnetic fields 0 (●) and 7 T (○) (Ref. 6). (c) Field dependence of the spin-wave stiffness at $T=40$ K (Ref. 7).

when two FM clusters with different orientations become in contact, they will align their moments. An insulating wall can prevent this process by drastically reducing the interaction between domains. However, when a relatively small magnetic field is switched on, these preformed large FM moments can easily rotate into the direction of that field. When this occurs, the insulating walls are no longer needed to prevent moment alignment, and they melt. This last process leads to a percolation, namely, as the FM clusters rotate their moments they also slightly increase their size to occupy the space left by the insulating walls, allowing for charge conduction to occur.⁵ Although this last step appears percolative, it is likely nonstandard and abrupt, since it involves large rounded clusters that are not very ramified (contrary to standard percolation), due to surface-tension effects that are of relevance in a regime of phase competition.

It is the main purpose of the present paper to describe, phenomenologically, this percolation of smooth objects, believed to describe CMR manganites. It will be argued that first-order characteristics can be found in percolative processes where surface-tension effects are relevant. It is important to clarify that the *full* description of the CMR process is far more involved, with the random distribution and subsequent field-induced alignment of FM moments playing a key role. The first steps in that direction were already described.⁵ Here our goal is the following: if the FM clusters are assumed to be already aligned (and in that case the spin degree of freedom can be dropped), how does the subsequent percolation occur? It will be argued that this transition can be fairly abrupt, even discontinuous. The paper is organized as follows. In Sec. II, a variety of nonstandard forms of percolation is described, searching for situations where first-order transitions are induced. The main conclusion of this section will be that achieving a discontinuous behavior is highly nontrivial and difficult. In Sec. III, an alternative solution is proposed, involving deterministic rules within a cellular automaton system. A discussion of results is presented in Sec. IV, while Sec. V contains the main conclusions.

II. FORMS OF PERCOLATION

Standard percolation (SP) is a successful statistical model for a variety of heterogeneous systems.⁸ It has been used to describe diffusion through porous media, conduction through random networks, and forest fires, among others. In spite of its successes, however, SP has several shortcomings, including the failure to account for correlation effects. Indeed, in SP each site (or bond) is occupied *independent* of its neighbors. This feature is responsible for the ramified structure of the usual percolation clusters. In Mn oxides the coexistence of large clusters has been reported,^{2,3} but the cluster distribution—while complex and involving many lengths scales—is not of the very ramified form characteristic of SP. A possible explanation lies in the long-range character of the elastic distortions induced by holes in Mn oxides,⁹ as well as surface-tension effects as described in the following section. These factors provide a natural tendency toward *smooth* surfaces of coexisting metal-insulator clusters. In addition, in physical systems with phase separation, there is a local ten-

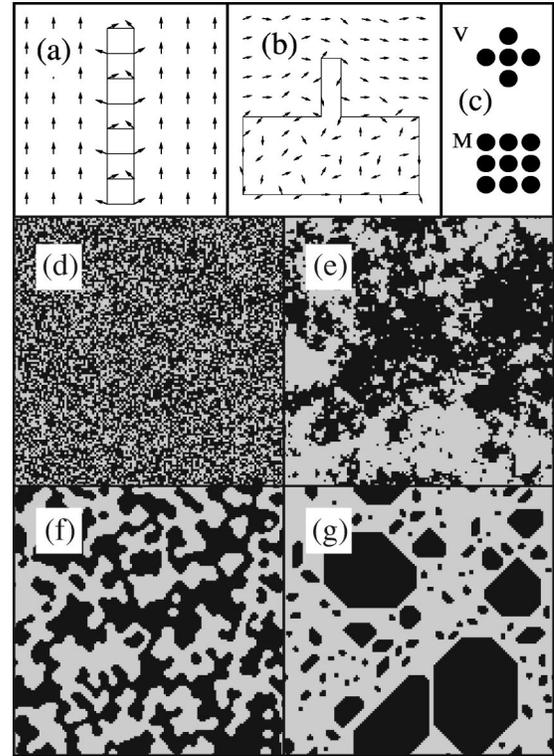


FIG. 2. (a) Typical snapshot of an 8×8 MC simulation of the one-orbital model (Hund coupling $J_H = \infty$, $T = 0.01t$, t the hopping amplitude, and density is 0.8). J_{AF} is 0 everywhere, but in the bonds of the ladder where they take the value $0.12t$. The average spin-spin correlation in the ladder is -0.4 (effective “double-exchange” hopping being 0.54), as opposed to -1.0 (effective hopping 0) when isolated from the FM region (effective hopping being 1). (b) Similar to (a) but for density 0.5. Here $J_{AF} = 0$ in the upper portion excluding the fingerlike region. In the rest, $J_{AF} = 0.15t$, favoring a flux state (Ref. 4). The effective hopping in the flux region is 0.7 , while in the finger it is as high as 0.9 . (c) The different neighborhoods in Vichniac’s notation (Ref. 16). (d) Typical configuration for standard percolation on a 128^2 lattice at (or just below) the critical density. (e) Same as (d) but for correlated disorder ($\gamma = 0.2$). (f) Same as (d) but for the annealing rule (MGE5). (g) Same as (d) but for the nucleating rule (MGE4). The latter is particularly promising since a small increase in p leads to a fully saturated final state in a first-order transition.

dency to homogenize the landscape, minimizing the interface tension.

While simulations of models considering the elastic effects through cooperative Jahn-Teller phonons are CPU time expensive, the cluster rounding effects appear even in simpler Hamiltonians, such as the one-orbital model⁴ (the actual form of this Hamiltonian has been extensively discussed in the previous literature, and it will not be reproduced here). Monte Carlo (MC) results in Figs. 2(a,b) illustrate this phenomenon. In the simulations, the Heisenberg coupling J_{AF} between localized t_{2g} spins is selected such that some regions of the studied lattice are in a FM state if they were isolated, while others present some competing spin arrangement [antiferromagnetic (AF) or flux⁴]. However, when the FM and non-FM regions are in contact, there is a consider-

able influence from one another, and the system tends to homogenize its transport properties. For example, the ladder in Fig. 2(a) would prefer to be in a perfectly AF spin state if isolated. However, by immersing the ladder into a ferromagnet, the spins cant appreciably, allowing for substantial charge hopping across the ladder. A thin AF region cannot survive the “pressure” of a FM environment, and it transforms into a fairly good conductor. Fig. 2(b) shows similar evidence, now using competing FM and flux states. A finger-like region emerging from the lower half (with coupling favoring the flux phase), also does not survive in its original form, but spins rotate to allow for conduction.¹⁰ Modeling percolative manganites with the metal/insulating character of a link independent of its neighbors can only be a crude approximation, and it may cause the present discrepancy between theory and experiment. If the clusters could be made more “rounded,” the transition would be more abrupt, and hopefully in better agreement with real manganites. The exponent t may depend on the short-scale, real-space metal-insulator arrangement, and changing the typical cluster shapes may alter t as well.

The physics of manganese oxides, in particular the abrupt change in resistivity as a function of both field and temperature, is supposed to emerge from the realistic one- and two-orbital models that are widely studied⁴ (supplemented by the addition of quenched disorder). However, simulations using realistic Hamiltonians for manganites—containing the rounding effect spontaneously—are simply prohibitive in the percolative regime if critical exponents are needed, and one must rely on semiphenomenological approaches, as in the present paper. Even if we accept this simplification, there is, *a priori*, no obvious way to introduce the proposed local cluster compactification in percolative scenarios. In alternatives to standard percolation such as “bootstrap” percolation,¹¹ one simply removes metallic sites or links if they do not have enough metallic neighbors. This is repeated until no more sites can be removed. However, numerical evaluation of the cluster number exponent τ suggests no new universal behavior.¹² To generate compact configurations, long-range “correlated disorder” was also investigated here.¹³ The disorder is selected to follow the real-space correlation function $C(l) = (1 + l^2)^{-\gamma/2}$, which falls off like $l^{-\gamma}$ for large distances l but is well behaved at $l=0$. Typical configurations confirm the presence of large clusters [Fig. 2(e)]. In addition, our conductivity studies for $\gamma=0.2$ (Fig. 3) are compatible with a new universality class and $t > 1.31 = t_{sp}$, as predicted before.¹⁴ However, to reproduce the large exponent $t \sim 7$ found in experiments,² anomalously small values of γ are needed. The reason is that even with correlated disorder, the weakest links still govern the critical behavior [Fig. 2(e) shows weak links separating large clusters]. Correlated disorder, with its statistical nature that allows for those links, is not sufficient to induce the smooth clusters needed to describe manganites. Naively, these already known results appear to rule out the possibility of finding an abrupt percolative transition to mimic the behavior of manganites.

Another alternative is the Swiss-cheese model.¹⁵ Halperin *et al.*¹⁵ have shown that this model is equivalent to a random

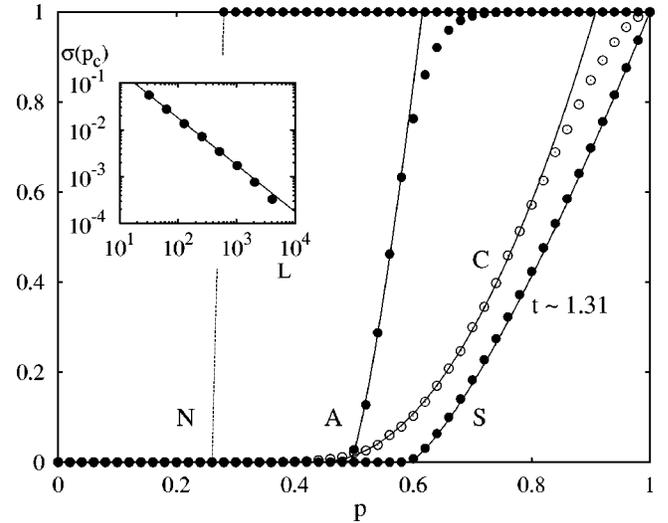


FIG. 3. Conductivity of a two-dimensional resistor network vs initial metallic fraction, for several representative cases. $N, A, C,$ and S stand for nucleated (MGE4), annealed (MGE5), correlated, and standard, respectively. Both the A and S lines are proportional to $(p - p_c)^t$, with $t \sim 1.31$. The inset shows finite-size scaling data for A to support universality. L is the linear size. Line C has a correlation power $\gamma = 0.2$ and exponent $t \sim 2.1$.

resistor network with a broad distribution of conductances g given by

$$P(g) = (1 - p)\delta(g) + p(1 - \alpha)g^{-\alpha}. \quad (1)$$

Here P denotes the probability that a conductance has values between g and $g + \delta g$, p is the metallic fraction as before, and $P(g)$ is characterized by an additional parameter α . The weakest bond dominates in the effective resistance of the backbone. Extensive numerical tests¹⁶ support the conjecture that $t = 1/(1 - \alpha)$ for $0.25 \leq \alpha < 1$. The previous equation becomes an equality for α large enough. Hence, since the data in Fig. 1(b) follow a straight line with slope -7 , it can be fitted with results of a simulation at $\alpha \approx 0.857$. Although this numerical success is encouraging, it is still intuitively unsatisfactory and does not address the first-order percolative transitions of Figs. 1(b,c). In fact, the rapid change in ρ_{dc} in Fig. 1(a) may also be indicative of a first-order transition hidden by imperfections in the samples used.

III. FIRST-ORDER PERCOLATION

The above examples clearly show that it is highly non-trivial to generalize percolation to yield a first-order transition, although it is possible to change the critical exponents. The existence of experiments [Figs. 1(b,c)] with discontinuous transitions indicate that there must exist yet another class of percolative transitions escaping our attention. We believe that the key issue missing in standard percolation and the related variations described above is similar to the effect of *surface tension*, which naturally arises when two phases

compete. This naturally penalizes the roughness of a surface and avoids the weak links ubiquitous in continuous percolation.

The simplest way to model the gregarious tendency of the two competing phases of manganites is to consider the nature of the cells in the neighborhood of the cell under inspection. If there are enough “friendly faces” around, the cell will not change, otherwise it will flip to the competing phase. In a magnetic system, the neighborhood induces a local field that influences the center spin. In addition, including oxygen movement, it is impossible to have an undistorted octahedron in the middle of a distorted Jahn-Teller region. The phenomenological approach followed here is based on Vichniac’s majority voting cellular automata.¹⁷ Loosely speaking, cellular automata are discrete dynamical systems whose evolution is described by a local deterministic rule: the fate of every cell at time $t + 1$ is determined by a function of the configuration of a few of its neighbors at time t . Cellular automata describe an approach to equilibrium in a system dominated by strong, short-range interactions. Vichniac¹⁷ defines four different neighborhoods: Q contains only the nearest neighbors, V consists of Q plus the center cell, H consists of Q plus the second-nearest neighbors, and M consists of H plus the center cell [see Fig. 2(c)]. Unlike in bootstrap percolation,¹¹ the center cell is treated on the same footing as its neighbors in V and M . This turns out to be a *key* property of the percolative process discussed here, making clusters compact and avoiding weak links. This is quite different from bootstrap percolation where the center cell is singled out. After the neighborhoods are defined, the update rules are as follows. Given a site i at a time t , let S be the number of occupied sites in the neighborhood N of i . S can be any number between 0 and the number of sites in the neighborhood. If S is greater than or equal to some threshold T , site i will be occupied at time $t + 1$. Otherwise, it will be empty. The procedure stops when no more changes occur.¹⁸ A rule is specified by a unique N and T . This is referred to as NGET.¹⁷ For the four neighborhoods defined above, only eight rules (with T close to half of the number of sites in N) are nontrivial, as explained below.

The starting point for the calculations are always standard percolation configurations, characterized by an occupation probability p . At time $t = 0$, p is a good measure of the occupied fraction, within statistical fluctuations. Under the “time evolution” provided by the cellular automaton, the occupied fraction needs no longer be equal to p . Rules with large (small) T tend to (increase) decrease it. For initial states with $p < p_c$, the final state does not percolate. Two examples, to be discussed in more detail in the next paragraph, are showed in Figs. 2(f,g). Note the clear visual difference from the more canonical cases, Figs. 2(d,e). The clusters are now more rounded, and our intuitive goal of phenomenologically mimicking the effect of surface tension has been reached. After the time evolution stops, the resistivity is measured using a numerically exact algorithm.¹⁹

The increase in conductivity σ immediately after p_c ($=0.5$) is clearly faster in the annealed [MGE5, i.e., each site remains or becomes metallic if five (or more) of its nine neighbors –including itself—are metallic] case than in stan-

dard percolation, as Fig. 3 indicates. This is in agreement with our intuition that rounding effects would speed up percolation, mimicking better the experimental situation after the randomly oriented FM clusters align in a magnetic field. However, it is surprising that this does not affect the value of t , as the inset shows. Nevertheless, this rapid increase could be experimentally relevant. On the other hand, the *nucleating rule* (MGE4, i.e., each site remains or becomes metallic if four or more of its nine neighbors—including itself—are metallic) leads to different results. Its resistivity jumps from 0 below p_c , where there is no connection across the system [see Fig. 2(g)], to the fully saturated value above p_c . To understand this, note that the clusters in Fig. 2(g) have very clean boundaries. Adding a single metallic site immediately outside one of the clusters will flip all the sites touched by the boundary. At p close to p_c , it is likely that this apparently small perturbation will cause the cluster to coalesce with a neighboring cluster. In practice, this starts a chain reaction (avalanche) that only stops when the entire lattice turns metallic. *The nucleating procedure indeed provides a percolative transition that of first order.*

How common are the first-order characteristics expected to be in the cellular automata models described here? Using the four (Q, V, H, M) neighborhoods defined above on the square lattice, there are eight nontrivial rules (QGE2, QGE3, VGE3, HGE4, HGE5, MGE4, MGE5, MGE6). By nontrivial we mean rules whose transition occur at a finite value of p (i.e., neither at 0 nor at 1) in the infinite-system limit. In all cases, a careful size extrapolation was carried out numerically to determine the critical concentration. They are, respectively, (0.16, 0.83, 0.5, 0.33, 0.66, 0.25, 0.5, 0.75). Although these values are close to the fractions (1/6, 5/6, 1/2, 1/3, 2/3, 1/4, 1/2, 3/4), we know of no rigorous result that they are equal to them. In two cases not listed above (VGE2 and VGE4), the numerical uncertainty prevented us from determining for sure whether the rule is trivial or not. To be specific, the critical p for VGE2 is very small, and it becomes smaller still as we increase the size of the system used in the simulation. We could not rule out the fact that p may extrapolate to zero in this case. As a consequence, we chose to leave those two cases out of the present discussion. Of the eight rules, the conductivity of three of them (QGE2, HGE4, and MGE4) was found to vary discontinuously at the transition. The conductivity of MGE4 is shown in Fig. 3 where the reader can convince oneself that the transition is discontinuous. QGE2 and HGE4 look identical apart for a shift in the location of p_c . Phenomenologically, the three cases behave similarly in the sense that states very close to the transition are unstable against flipping even just one site. Hence, we conclude that first-order tendencies are certainly not a fragile feature among the set of possible voting cellular automata. However, far more work is needed to fully understand the behavior of these systems, with the present paper hopefully providing the first steps in that direction. The detailed analysis of the models introduced here may be as rewarding as the previous studies of standard percolation.

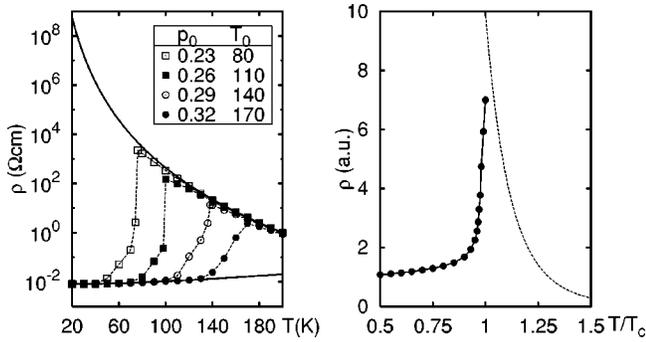


FIG. 4. Left panel: Resistivity ρ_{dc} vs T using the nucleating rule. The values of ρ_{dc} for metallic and insulating bonds are taken from experiments (Ref. 2), as in Ref. 20. Following Ref. 20, the metallic fraction varies linearly with temperature, $p - p_0 = \alpha(T_0 - T)$. We consider $\alpha = 0.1\%$, $T_0 = 100$ K, and label the curves by p_0 , their metallic fraction at $T = T_0$. Right panel: ρ_{dc} vs T of the $Q = 20$ Potts model (for details, see text). A cluster algorithm (Ref. 21) was used on a 64^2 lattice. Long simulations (several thousand sweeps) were required to sample both peaks in the probability distribution function satisfactorily. Above T_c there is no percolation ($\rho_{dc} = \infty$), but this can be solved phenomenologically by allowing for conductivity among the several Q clusters, as in the dashed line.

IV. DISCUSSION

The discontinuous percolative transition described here can be applied phenomenologically to the Mn oxides. In a previous related effort, Mayr *et al.*²⁰ constructed random resistor networks using standard percolation and provided a tentative phenomenological description of manganites. The curves ρ_{dc} vs T mimicked properly the results corresponding to some manganites, but were too smooth to accommodate the materials of Fig. 1 as special cases. The nucleating-rule procedure to generate metal and insulator clusters described here may solve this problem. The left panel of Fig. 4 contains the effective resistivity (obtained solving the Kirchoff equations) vs T for configurations generated by the nucleating rule, where the light (dark) regions in Fig. 2(g) are insulating (metallic). The individual resistivity of those regions is taken from experiments, and they carry a T dependence, similar to the work of Mayr *et al.*²⁰ The new curves are in *excellent* agreement with experiments [see, e.g., Fig. 1(b)], including the discontinuity, and the fact that there is a common value of ρ_{dc} at both low and high T .

Note that the family of percolative processes presented here naively seems triggered only by the occupation fraction p , not by temperature, as opposed to some of the results of Fig. 1. However, the ferromagnetic fraction in real manganites is a function of temperature: clusters are expected to be formed at T^* (although they are not sufficiently large in size to create a global FM state), their number grows with decreasing temperature (i.e., p increases in the language of this paper), until the Curie temperature is reached. It would be incorrect to believe that our results are only valid for cases where the temperature is held fixed while concentrations are changed.

Another clarification is important. Why do all manganites not have either a first- or second-order transition? Current

experimental evidence suggests that both cases can be found, although the latter tends to be fairly abrupt.⁴ Several aspects may influence these results. First, it may happen that the single crystals available are affected by extrinsic effects in their preparation. From this perspective, the current experimental results may vary in the future as cleaner samples are prepared. But there are other issues to consider. Clearly, different manganites have different degrees of intrinsic disorder, such as the chemical disorder emerging from having different ionic sizes at the A site of the perovskite. This disorder will likely influence the clusters shape, and both configurations Figs. 2(f) and 2(g), may emerge at different compositions, leading to either rapid but continuous or first-order percolative processes, respectively. It is clear that the issue of universality of the first-order transition in manganites is far from clear both experimentally and theoretically, and more work should be devoted to its analysis. Here, we have simply provided a plausible rationalization of some steps involved in the CMR transition of the materials mentioned in Fig. 1. Whether all CMR materials have the same kind of first-order transitions or not will hopefully be clarified by future experiments.

Finally, although the discussion here is based on a cellular automaton, we found it reassuring that first-order percolative processes can also arise from well-defined Hamiltonians. In particular, consider the Q -state Potts model known to have a first-order transition for $Q > 4$.²² Simulating the model ($Q = 20$) with MC techniques and using the Fortuin-Kasteleyn mapping²³ to construct clusters for the various Q 's, their percolative properties were analyzed.²⁴ Clusters for each Q are individually assumed metallic, and the network resistance is calculated. Results [Fig. 4 (right)] indeed have a ρ_{dc} discontinuity (above $\rho_{dc} \approx 7$) at the critical temperature T_c . At $T > T_c$, no cluster percolates leading to infinite ρ_{dc} , but this can be easily fixed assuming a finite conductivity among the many Q clusters. Of course, it is not at all claimed here that real manganites have the Potts symmetry, this is just an illustration of concrete model Hamiltonians that presents the first-order transition described in the preceding section by cellular automata rules (rather than emerging from a Hamiltonian approach).

V. CONCLUSIONS

In summary, a phenomenological approach is proposed to include correlation and surface-tension effects in percolation. The rounding of clusters induces an abrupt first-order transition in the model, consistent with results of some experiments Fig. 1(b). The present calculation suggests that many manganites do not belong to the standard percolation class, as believed by many before, but correlation effects change the universality class inducing a faster percolative process that may present hysteretic features.²⁵ Note that the fairly “random looking” configuration [Fig. 2(g)] is *self-generated*²⁶ by our approach, its phenomenological origin lies in phase competition, and is *not* pinned by explicit disorder. Thus, the first-order percolative transition is predicted to appear particularly in nominally “clean samples”

and for low intrinsic chemical disorder. If explicit disorder is added to the system, Fig. 2(g), then the transition is expected to become continuous. More work is needed to fully clarify the fascinating physics of CMR manganites.

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