Complexity in Strongly Correlated Electronic Systems

Elbio Dagotto

A wide variety of experimental results and theoretical investigations in recent years have convincingly demonstrated that several transition metal oxides and other materials have dominant states that are not spatially homogeneous. This occurs in cases in which several physical interactions—spin, charge, lattice, and/or orbital—are simultaneously active. This phenomenon causes interesting effects, such as colossal magnetoresistance, and it also appears crucial to understand the high-temperature superconductors. The spontaneous emergence of electronic nanometer-scale structures in transition metal oxides, and the existence of many competing states, are properties often associated with complex matter where nonlinearities dominate, such as soft materials and biological systems. This electronic complexity could have potential consequences for applications of correlated electronic materials, because not only charge (semiconducting electronic), or charge and spin (spintronics) are of relevance, but in addition the lattice and orbital degrees of freedom are active, leading to giant responses to small perturbations. Moreover, several metallic and insulating phases compete, increasing the potential for novel behavior.

Materials in which the electrons are strongly correlated display a broad range of interesting phenomena, including colossal magnetoresistance (CMR), where enormous variations in resistance are produced by small magnetic field changes, and high-temperature superconductivity (HTSC). An important characteristic of these materials is the existence of several competing states, as exemplified by the complicated phase diagrams that transition metal oxides (TMOs) present (Fig. 1). The understanding of these oxides has dramatically challenged our view of solids. In fact, after one of the largest research efforts ever in physics, involving hundreds of scientists, even basic properties of the HTSC cuprates, such as the pairing mechanism, linear resistivity, and pseudogap phase, are still only poorly understood. In the early days of HTSC, it was expected that suitably modified theories of ordinary metals would explain the unusual properties of the cuprate's normal state. However, important experimental results gathered in recent years have revealed an unexpected property of oxides: Many TMOs are inhomogeneous at the nanoscale (and sometimes at even longer length scales). This explains why the early theories based on homogeneous systems were not successful and raises hopes that a novel avenue for progress has opened.

Department of Physics, University of Tennessee (UT), Knoxville, TN 37996–1200, USA. Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831–6393, USA.

What are the implications of these and other results reviewed below? It will be argued that the current status of correlated electrons investigations must be considered in the broader context of complexity. In his pioneering article (*1*), Anderson wrote that "the ability to reduce everything to simple fundamental laws does not imply the ability to start from those laws and reconstruct the universe." In complex systems (*2*), the properties of a few particles are not sufficient to understand large aggregates when these particles strongly interact. Rather, in such systems, which are not merely complicated, one expects emergence, namely the generation of properties that do not preexist in a system's constituents. This concept is contrary to the philosophy of reductionism, the traditional physics hallmark. Complex systems spontaneously tend to form structures (self-organization), and these structures vary widely in size and scales. Exceptional events are important, as when the last metallic link completes a percolative network. The average behavior is of no relevance for this phenomenon, and often only a few rare events dominate. Evidence is accumulating that TMOs and related materials have properties similar to standard complex systems, and several results must be reexamined in this broader framework.

Nanostructures in Manganites and Cuprates

Manganites. The Mn oxides called manganites (*3*–*9*), especially those displaying the CMR effect, are an important oxide family in which the presence of inhomogeneous states is widely accepted. A remarkable cross-fertilization between theory and experiments has led to considerable progress in unraveling the role of these inhomogeneities. Theoretical investigations (*4*) predicted that, in a broad region of parameter space, the ground state is actually a nanoscale mixture of phases, particularly in the presence of quenched disorder (*10*–*12*), namely, when random "frozen" deviations from the perfectly uniform system are incorporated in the study. Many experimental results are indeed in agreement with the basic notion that the relevant phases are not homogeneous; these results also provide information crucial to understanding the CMR effect (*4*, *5*, *13*, *14*). Some of the general theoretical ideas are summarized in the schematic phase diagram (Fig. 2A) (*10*), which has been experimentally confirmed (*15*, *16*) (Fig. 2B).

In the clean limit without quenched disorder, the two key competing states in manganites, ferromagnetic (FM) metallic and antiferromagnetic (AF) insulating (AFI), are known to be separated by a first-order transition (*4*, *5*). However, once the inevitable quenched disorder is included in the calculation, arising, for example, from the lattice-distorting chemical doping procedure, nonstatistical fluctuations of dopant density or strain fields, the region in which the two states are nearly degenerate (that is, they can coexist) is dramatically modified. In this regime, there is still a local tendency toward either FM or AFI short-distance correlations. However, globally neither of the two states dominates (Fig. 2C). A mixed glassy region is generated between the true critical temperatures, the Curie or Néel temperatures in this case, and a remnant of the clean-limit transition, T^*. In this regime, perturbations such as small magnetic fields can have dramatic consequences, because they only need to align the randomly oriented magnetic moments of preformed nanosize FM clusters to render the system globally ferromagnetic. A concomitant percolation induces metallicity in the compounds. The fragility of the state shown in Fig. 2C implies that several perturbations besides magnetic fields should induce dramatic changes, including pressure, strain, and electric fields (*4*, *5*). Moreover, the discussion centered on Fig. 2, A to C, is independent of the details of the competing states and should be valid for the AFI versus

superconducting (SC) state competition in cuprates (*17*) and many other cases (*18*).

Calculations that incorporate the effects of phase competition and quenched disorder have been able to reproduce the huge magnetoresistance observed experimentally (*10*, *11*); this suggests that the CMR effect would not occur without either competing states or quenched disorder and interactions necessary to nucleate clusters. This is in agreement with experiments for $Re_{0.5}Ba_{0.5}MnO_3$ (where Re is a rare earth element) (*16*), which can be prepared both in ordered and disordered forms for the Re-Ba distribution. Remarkably, only the latter was found to exhibit CMR (Fig. 2D). This suggests that when phases compete, the effect of (typically small amounts of) quenched disorder results in dramatic properties that are very different from those of a slightly impure material (*10*, *11*, *19*, *20*). Disorder in the regime of phase competition is not a mere perturbation; it alters qualitatively the properties of the material.

How strong should the disorder be to induce the inhomogeneous patterns discussed here? Are there other alternatives? Studies incorporating long-range effects, such as Coulombic forces (*21*) or cooperative oxygen octahedra distortions (*11*), suggest that very weak disorder, even infinitesimal disorder (*21*, *22*), may be sufficient to do the job. Calculations without explicit disorder incorporating strain effects (*9*), or within a phenomenological Ginzburg-Landau theory, also lead to inhomogeneous patterns (*23*). Although the discussion on the details of the origin of the inhomogeneities is still fluid, their crucial relevance to understanding the manganites, as originally predicted by theory (*4*, *5*), is by now widely accepted.

Cuprates. In the HTSC context, the $La_{2-x}Sr_xCuO_4$ (LSCO) phase diagram is usually considered the universal diagram for cuprates. However, some investigations suggest otherwise (*17*, *24*). For example, only

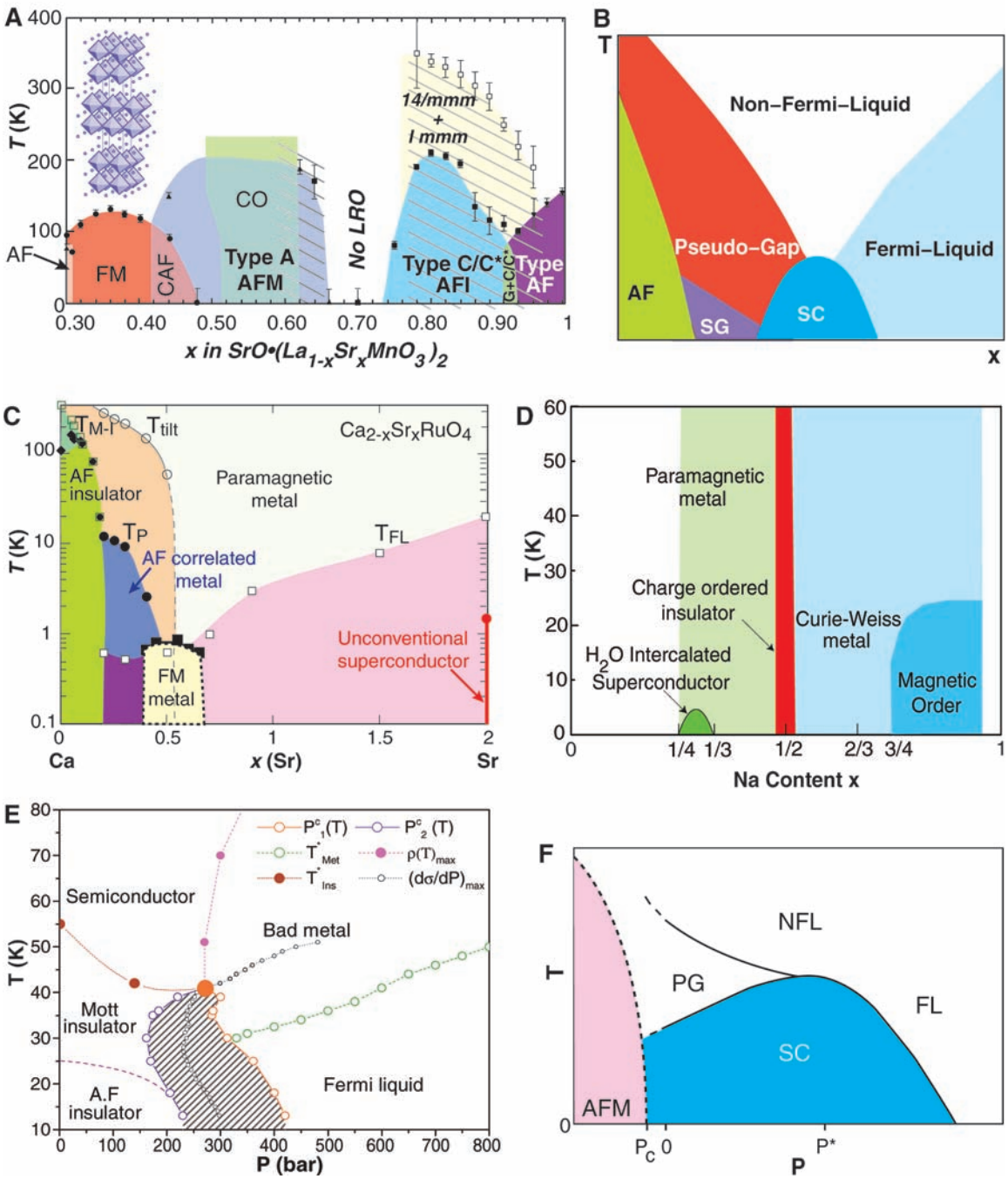

Fig. 1. Phase diagrams of representative materials of the strongly correlated electron family (notations are standard and details can be found in the original references). (**A**) Temperature versus hole density phase diagram of bilayer manganites (*74*), including several types of antiferromagnetic (AF) phases, a ferromagnetic (FM) phase, and even a globally disordered region at $x = 0.75$. (**B**) Generic phase diagram for HTSC. SG stands for spin glass. (**C**) Phase diagram of single layered ruthenates (*75*, *76*), evolving from a superconducting (SC) state at $x = 2$ to an AF insulator at $x = 0$ (x controls the bandwidth rather than the carrier density). Ruthenates are believed to be clean metals at least at large x, thus providing a family of oxides where competition and complexity can be studied with less quenched disorder than in Mn oxides. (**D**) Phase diagram of Co oxides (*77*), with SC, charge-ordered (CO), and magnetic regimes. (**E**) Phase diagram of the organic κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl salt (*57*). The hatched region denotes the coexistence of metal and insulator phases. (**F**) Schematic phase diagram of the Ce-based heavy fermion materials (*51*).

after Ca is added to $YBa_2Cu_3O_{6+\delta}$ (where δ is the excess of oxygen, and it ranges between 0 and 1) does its phase diagram resemble that of LSCO (*25*, *26*). Moreover, organic superconductors do not have a glassy phase between the AFI and superconducting states, and they are believed to be cleaner than the cuprates (*27*). This suggests that quenched disorder (or strain, etc.) in cuprates may play a role as important as that in the manganites, and the exotic underdoped regime and T^* may emerge as a consequence of its influence (*17*). If so, then it is not sufficient to consider phase diagrams involving only temperature and hole-doping x. A disorder strength axis should be incorporated into the phase diagram of these materials as well.

Considerable discussion concerning the existence of inhomogeneous states in cuprates started several years ago when stripes were reported in studies carried out with neutron-scattering techniques (*28*). These states had been predicted theoretically (*29*, *30*). The nontrivial real-space structure of stripes emerges from Hamiltonians that do not break translational invariance, which is a remarkable result. However, because approximations were made in the calculations, it is still controversial whether stripes do exist in Hubbard Hamiltonians (*31*–*34*). Experimentally, the presence of stripes is also a matter of debate. Recent neutron studies of HTSC materials have been interpreted as caused by a phase that contains stripes separated by two-leg ladders (Fig. 3A) (*35*, *36*), with spin-gapped properties that could be important for pairing (*37*). In addition, doped Ni oxides and Nd-doped LSCOs are widely believed to have stripes (*29*).

While the stripe debate continues, scanning tunneling microscopy (STM) investigations have recently provided additional important information on the cuprates, unveiling a variety of other inhomogeneous states. Figure 3B shows a real-space distribution of *d*-wave SC gaps in $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212). The many colors illustrate the inhomogeneous nature of the state (*38*) with randomly distributed nanoscale patches. These patterns could be caused by phase competition or by a random oxygen distribution. Other recently synthesized cuprate-based compounds also have inhomogeneous states (*39*), and additionally, a new charge-ordered "checkerboard" state has been observed (Fig. 3C) (*40*). This state also exists in Bi-2212 (*41*) and appears to compete with superconductivity. Understanding these novel states remains a challenge, but for our purposes two issues are crucial: (i) When scrutinized with powerful microscopic techniques, doped HTSC systems reveal inhomogeneous states. Supporting this statement, a novel scaling law for the cuprates was interpreted as produced by a Bardeen, Cooper, and Schrieffer system in the dirty limit (*42*, *43*). (ii) The intermediate states between the AFI and SC states do not seem universal (they could have stripes, a

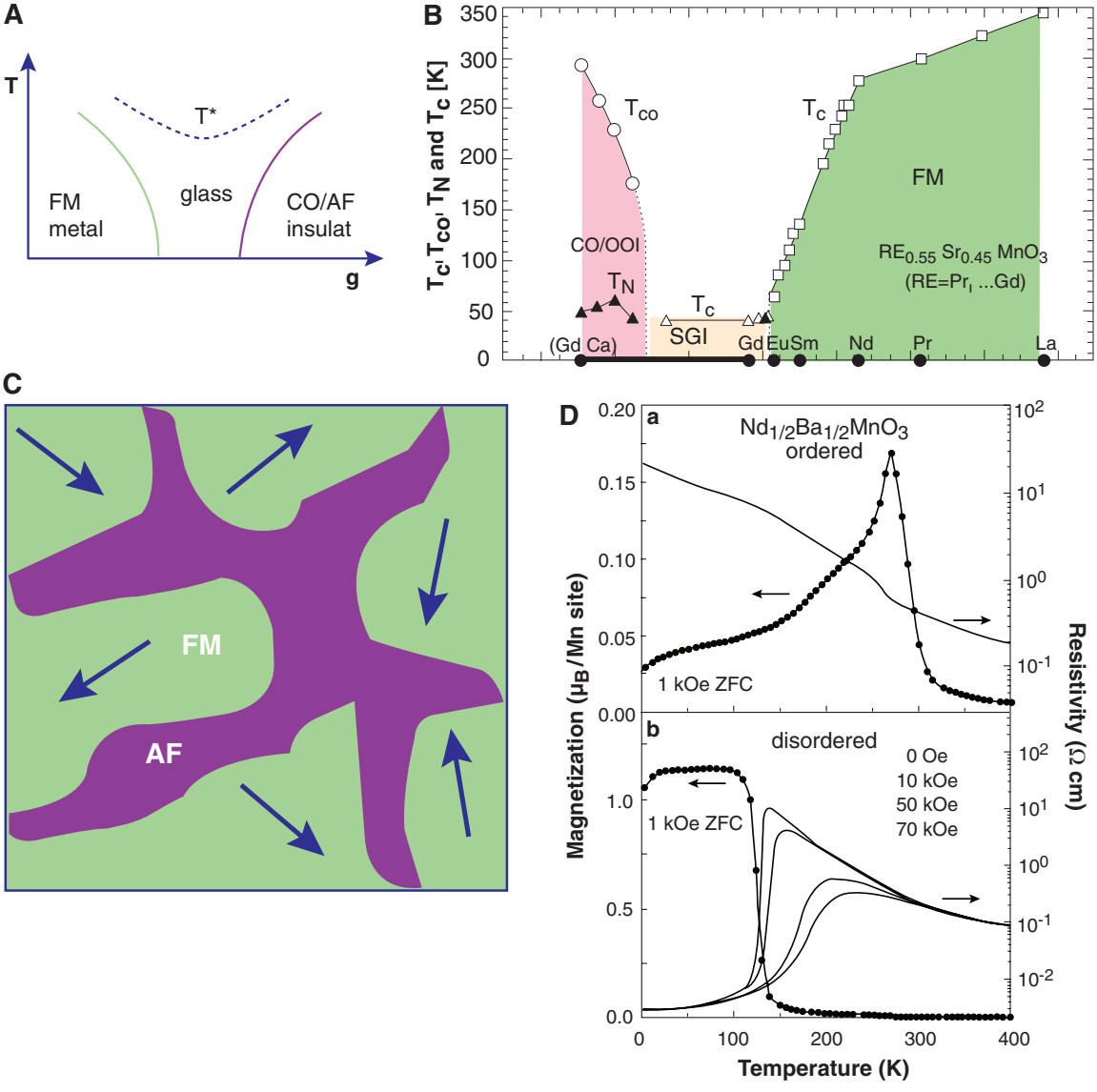

Fig. 2. (**A**) Generic phase diagram of two competing states [here FM metal versus charge-ordered antiferromagnetic (CO/AF) insulator] in the presence of quenched disorder (*4*, *5*). g is a generic variable to move from one phase to the other (e.g., electronic density or bandwidth). A glassy mixed-phase state is created and a T^* scale appears. (**B**) Experimental phase diagram of manganites with large disorder (*15*, *16*). Note the disorder-induced suppression of the ordering temperatures and the appearance of a glass state, as predicted by theory (A). Details and the phase diagram with weak disorder can be found in (*15*). (**C**) Sketch of the proposed CMR state for the manganites containing FM clusters with randomly oriented moments separated by regions where a competing CO/AF phase is stabilized (*4*, *5*, *13*). (**D**) Resistivity and magnetization versus temperature for the ordered and disordered structures of $Nd_{0.5}Ba_{0.5}MnO_3$ (*16*). Only the disordered crystal has the CMR effect (*16*).

charge checkerboard, or glassy patterns). All these characteristics are hallmarks of complex systems, showing sensitivity to details as they occur in nonlinear chaotic systems.

Some additional issues should be remarked upon: (i) Although the most complex behavior in cuprates appears in the underdoped regime, dynamic electronic inhomogeneity and competition among the many degrees of freedom could also underlie the superconductivity even at optimal doping. Are the inhomogeneities and complexity at the root of the superconducting phase, or are they unrelated? The discussion continues. (ii) Interactions can also generate inhomogeneous patterns (*17*, *29*, *44*), and the combination of these interactions with quenched disorder may be at the heart of the complexity in cuprates.

Another unexpected property of the cuprates is the giant proximity effect. This phenomenon has a long story, but recently it has been very carefully studied by using atomically smooth films made of HTSC compounds in S-N-S trilayer junctions (S is for superconductor and N equals normal metal) (*45*). The big paradox here is that the trilayer behaves as a Josephson junction for N barriers a hundred times thicker that the coherence length, ξ. Then, the normal state cannot be featureless, it must already contain a tendency toward superconductivity, which could be in the form of nanoscale SC islands (*17*) or phase-fluctuating homogeneous states (*46*). This proposal leads to an exciting prediction: Under the proper perturbation, the state with preformed SC clusters should present a gigantic susceptibility toward superconductivity (*17*). This is the analog of CMR in Mn oxides but translated into Cu-oxide language. In general, theory predicts that giant responses to external perturbations should be far more common than previously anticipated.

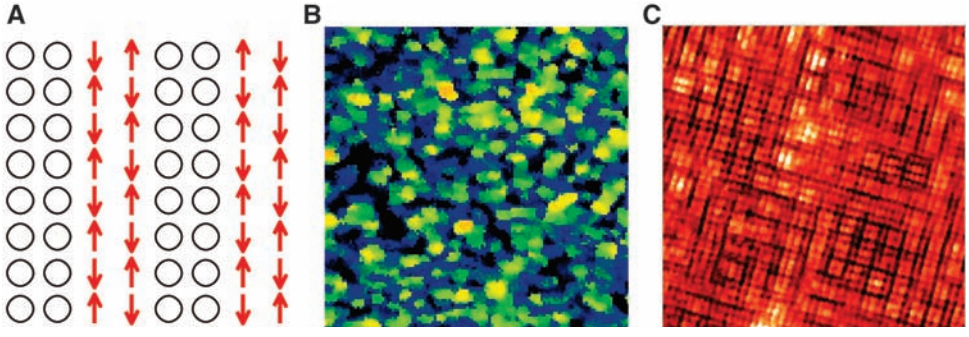

Fig. 3. Examples of inhomogeneous states in HTSC materials. (**A**) Schematic perfect stripes (*35*) (circles are holes; arrows, spins). Real systems may present more dynamical stripes (*29*). (**B**) *d*-wave SC gap real-space distribution obtained by using STM techniques (*37*). Inhomogeneities at the nanoscale are observed (patches). The entire frame is 560 Å by 560 Å. (**C**) Recently unveiled charge-order state (checkerboard) in Na-doped cuprates (*40*, *41*).

The Case for Complexity in Correlated Electron Systems

Are TMOs examples of complex matter? Considering the general properties of complexity briefly reviewed in the introduction as well as the oxide results discussed in the previous section, it is natural to wonder whether these systems can be considered as special cases of complex matter. Although complexity is natural when associated with soft matter (literally soft, for example, polymers and liquid crystals), it seems out of place in the context of hard materials. But the several simultaneously active degrees of freedom may conspire to provide a soft electronic component to transition metal oxide compounds, soft not in the physical hardness sense but denoting the existence of a multiplicity of nearly degenerate conformations of the electronic component that can be easily modified by external perturbations. Conventional soft matter is classical [$(^{h}/_{2\pi}) = 0$], but in the electronic systems described here quantum effects are important.

TMOs are soft in the sense described above as already proposed in the HTSC context (*44*). They are also complex, because several effects become simultaneously important and prevent a simple physical description. More specifically, consider one of the popular definitions of complexity recently discussed in (*47*): "… randomness and determinism are both relevant to the system's overall behavior. Such [complex] systems exist on the edge of chaos—they may exhibit almost regular behavior, but also can change dramatically and stochastically in time and/or space as a result of small changes in conditions." This definition is satisfied by manganites, in which a small magnetic field produces a drastic change in transport properties, and it may apply to underdoped cuprates as well (*17*, *45*). When phases compete, general arguments suggest that large responses to weak perturbations should be far more common than previously believed (*4*, *5*, *17*). Although the basic rules for electrons (i.e., the Hamiltonians) are deceptively simple (nearest-neighbor carrier hopping, Coulomb or phononic interactions, etc.), the outcomes are highly nontrivial when phases compete and percolative physics, as when only a narrow channel exists for electrical conductivity through a material, is at work.

Another argument can be found in the known properties of traditional soft condensed matter, which is a phase of matter between a simple fluid and a regular solid crystal. In soft matter, large groups of atoms form regular patterns as in a solid, but when several of these large groups are considered together a complex fluid behavior emerges. Typical examples are polymers: in each large molecule there is atomic regularity, but an ensemble of them has a variety of fluid phases (*48*). This variable behavior is also present in some TMOs: in manganites, several experimental investigations have found evidence for Jahn-Teller ordered small regions (i.e., with a particular form of lattice distortions) in the state above the FM ordering temperature (*4*, *5*). As a system, these small Jahn-Teller clusters, along with the magnetic clusters present in the same phase region, generate a collective behavior that is different from the behavior of the system's individual parts, and in this temperature range colossal magnetoresistance occurs (*4*, *5*). Also cuprates may behave as electron liquid crystals, intermediate between electron liquid and electron crystal (*44*). Softness in the manganite context has also been recently discussed (*23*). Once these concepts are accepted, then the long history of soft-matter investigations suggests that it is natural to expect new kinds of organized behavior. In complex systems, randomness and determinism are simultaneously relevant, and these are ideas compatible with recent correlated electrons investigations (*10*, *11*, *15*, *19*, *20*).

Each complex situation in correlated electrons may lead to a unique state. Some materials may have stripes, others may have patches, some may have phase separation at nanoscales, and others may have mesoscale phase separation; the number of states in competition and their nature can lead to enormous possibilities. This is exciting for applications but frustrating for those with a reductionist soul. What is likely is that new general concepts and paradigms will emerge as guiding qualitative principles in the study of complex oxides. It will be difficult to predict the precise shape of the nanopatterns and the phases in competition unless detailed calculations are performed. But the existence of some patterns, as well as giant responses to selected external perturbations, will be predictable. Certainly the highest degree of complexity is expected when many degrees of freedom are active simultaneously and when many phases with different properties are in competition.

Theory, phenomenology, computer simulations. How can we make further progress in this context? Investigations involving the

fundamental Hubbard and *t-J* models are reaching the limits of our current many-body techniques. It appears unlikely that the large length scales needed to fully capture the complex behavior of oxides, where percolation is probably very relevant, will be reached via this path, and we must focus on the right level of description. As Laughlin and Pines (*49*) wrote, ''Deduction from microscopics has not explained, and probably cannot explain as a matter of principle, the wealth of crossover behavior discovered in the normal state of the underdoped cuprates.'' It is still reasonable that key issues such as the pairing mechanism and short-distance nature of the dominant states can still be analyzed in the context of Hubbard-based approaches, perhaps supplemented by long-range Coulomb and/or electron-lattice interactions. However, the complexity of the resulting states, with emerging self-organization and giant responses, can only be addressed with simpler phenomenological models that assume competition between a few selected states and analyze its consequences. For example, the famous linear resistivity and puzzling underdoped behavior of the cuprates and the CMR effect in the manganites may only be explainable with use of this coarse-grain approach.

The logical chain starts with ab initio calculations to evaluate the main parameters and couplings, followed by Hubbard modeling to obtain the dominant short-distance correlations, and ends with the use of more phenomenological models (*17*) to handle the long length scales of relevance in an electronic complex fluid. The inclusion of both symmetry and spontaneous symmetry breaking will be important to achieving these objectives, as will the inclusion of the effects of disorder and lattice distortions. Essential for the success of the present flurry of research in complex systems is the ability to use high-speed computers to perform unbiased calculations. By simulating a system made of many small units, the behavior of the whole ensemble can be understood and manipulated much better than with other techniques, providing new ways of learning and visualizing in this context.

Other systems with similar complex behavior. There are many other materials that behave similarly as the TMOs emphasized in this review. For example, in the area of heavy fermions (metals where the effective mass of electrons is much larger than the bare mass) the presence of ''electronic Griffiths phases,'' inhomogeneous states at zero temperature, has been described (*50*), and strong similarities with the cuprate's phase diagram were unveiled (*51*) (Fig. 1F). In general, glassy behavior is expected near a metal-insulator transition at low temperatures (*50*, *52*, *53*), establishing an interesting connection with the area of investigations known as ''quantum critical phenomena'' (*54*). Glassy dynamics is also observed in other two-dimensional electronic systems (*55*). Cobalt oxides (*56*), organic materials (*57*, *58*) (Fig. 1E), and Ca-doped ruthenates (Fig. 1D) are other examples. Materials where charge density waves and superconductivity compete provide other cases of complex behavior (*59*). The area of complexity in correlated electrons is far wider than the two TMOs chosen in this article to focus on.

Complexity in pure states. The emphasis of this review has been on self-organization and the complexity in the electronic sector associated with the existence of several competing states. This corresponds to the physics of the HTSC cuprates in the underdoped regime and the manganites in the CMR regime. However, complexity in strongly correlated electrons also exists in the fascinating ground states observed in the clean limit or far from the region of phase competition if quenched disorder is present. For example, superconducting ground states with zero electrical resistance, a Meissner effect, and unconventional properties [*d* wave in the cuprates or spin-triplet pairing in the ruthenates (*60*)] emerge from simple interactions among electrons and lattice vibrations. In the manganites, a CE phase exists with simultaneous spin, lattice, orbital, and charge order (*4*). The list of exotic phases observed in the clean limit is enormous, and they all represent emergent phenomena in the sense that their properties cannot be predicted easily from the Hamiltonian. The collective behavior of electrons in these phases is relatively simple, and it can be described with a handful of concepts and parameters. The emergence of simplicity is part of the complex behavior of electrons (*49*). Whereas in the case of Mn oxides the inhomogeneities are crucial to understanding the CMR effect and in cuprates the analogous inhomogeneities are important to rationalize the curious underdoped regime, they do not provide an obvious means to comprehend the origin of all the many exotic ground states. Thus, with or without quenched disorder, in homogeneous or inhomogeneous forms, it is clear that systems of strongly correlated electrons are surprising and that the list of their many possible ground states is far from fully classified. Research producing highly pure samples is as important as those focusing on the region of inhomogeneities and pattern formations, leading to complementary insight. Clearly, these compounds are complex in more than one sense.

Applications? It is too early to decide if the complex properties of correlated oxides could be important for applications, but several results already provide interesting clues. To name a few, the resistance of some oxide films was unexpectedly found to switch between low and high values upon the application of voltage pulses (*61*, *62*). Also, gigantic magnetoelectric effects were reported (*63*), interfaces of magnetic oxides have been engineered (*64*), manganites with sharp magnetization steps exist (*65*), and manganite nanotubes were prepared (*66*, *67*). Creating ultra-smooth thin films and artificial superstructures is part of the avenue toward applications. Because complexity appears to be the reason behind the CMR effect, complex behavior is conducive to functionality. Relaxor ferroelectrics are also oxides with nanodomains with potential applications (*68*). Field-effect transistors made from TMOs are another exciting area of research (*69*): correlated electron materials could present phase transitions in the presence of electric fields because these fields can alter the carrier concentration. It is the diversity of behavior, namely the many possible metallic, insulating, magnetic, superconducting, and ferroelectric phases of strongly correlated systems, that makes these types of investigations so exciting.

Conclusions

TMOs are certainly not as simple as standard metals. The many active degrees of freedom—spin, charge, lattice, and orbital—interact in a nonlinear, synergetic manner, leading to an intrinsic complexity. STM, neutron and x-ray scattering, and other microscopic techniques are crucial to unveiling the subtle nanoscale phase separation tendencies that induce a variety of real-space patterns. Charge transport in oxides is quite different from the free flow in simple metals: an isolated charge strongly perturbs its environment, inducing a polaron, which often attracts other polarons to form larger structures. To capture this physics, it is important to incorporate several ingredients, including powerful nonperturbative many-body techniques, phenomenological approaches, and the effects of lattice distortions, strain, and quenched disorder. All these ingredients appear equally important. Phase competition rules the behavior of these compounds: Although the energies characterizing each phase (such as gaps) can be fairly large, at particular carrier densities or bandwidths the energetic proximity of two phases introduces a lower hidden energy scale and small perturbations cause huge responses, not via the melting of the analyzed state but by its replacement by a very different one.

Establishing electronic complexity in hard materials as a fundamental area of research will create scientific relations with other popular fields of investigations. For instance, the existence of complexity in biological systems is clear, and analogies between proteins and spin glasses, both of which have a distribution of barrier heights among competing nearly degenerate states, have often been remarked on (*70*). In fact, most correlated electronic systems exhibit exotic glassy behavior with notoriously slow dynamics (*71*, *72*), establishing one of the prime connections between traditional biological or soft systems and the complex states described here. Biological physics is one of the

major frontiers for physics in the new millennium and complexity certainly arises in macromolecules and complex fluids. A common language can also be established with other broad fields: for instance, in nuclear matter the self-generation of structures is under much discussion as well (*73*).

A novel paradigm involving "complexity in correlated electron materials" will help to focus on the right level of description, on the expected emergence of patterns, and on separating the physics of the individual phases from properties that arise from phase competition. Controlling the spontaneous tendencies toward complex pattern formation may open the way to achieving emergent functionalities in correlated electrons systems. The enormous diversity of phases in oxides provides a wide range of combinations to explore. Complexity and functionality are rapidly developing into the most exciting frontiers in the active area of strongly correlated electrons.

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