Whither Oxide Electronics?

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Guest Editors

The theme of this issue on "Whither Oxide Electronics?" is an outgrowth of the 2007 David Turnbull Lecture, presented by Ramamoorthy Ramesh, on November 27, 2007, at the 2007 Materials Research Society Fall Meeting in Boston. The Lectureship is awarded annually by MRS to recognize the career of a scientist who has made outstanding contributions to understanding materials phenomena and properties through research, writing, and lecturing, as exemplified by David Turnbull of Harvard University.

Abstract

In this issue we have endeavored to answer the question, "Whither oxide electronics?" This issue provides a framework and perspective on the progress in the field of oxide electronics over the past several decades, as well as the challenges and opportunities in the years to come. Building on the foundations laid by the pioneers in the materials community and spurred by the discovery of high-temperature superconductivity, there has been both tremendous progress in understanding the complex science of oxide electronic materials and the discovery of other fascinating new phenomena, including colossal magnetoresistance, multiferrocity, and two-dimensional electron gases in correlated oxide systems. Thin-film heterostructures provide a pathway to create novel devices and combinations of physical phenomena. Indeed, the ability to synthesize and control oxide heterostructures using sophisticated deposition techniques has become a key enabler of the recent advances in this field. These oxides are beginning to enter mainstream products because of their higher performance, for example, ferroelectric memories and oxides with high dielectric constant for computers that run at higher speed and use less power.

A Brief History of Oxide Electronics

The field of complex oxides traces its lineage to the pioneering work of Sir Neville Mott[1] and co-workers, who originally identified the complex nature of the electronic behavior of oxides, even those as simple as NiO. Their work illuminated the role of electron correlations in determining the transport properties of such materials: correlated electron effects. In the 1940s, the wartime effort demanded new and better materials for a broad range of applications and stimulated Arthur von Hippel's research[2] on dielectric perovskites and related materials. In the 1950s, the work of Goodenough[3] and his generation led to some dramatic developments in understanding the crystal chemistry and transport physics of doped transition-metal perovskites. This work especially focused on the manganites, which, in essence, revisited the correlated behavior of electrons in the broad family of perovskites. Indeed, many of the "rules" of interactions in these transition-metal-based oxides were outlined in this period. In parallel, the work of Eric Cross and colleagues[4] on perovskite dielectrics both opened up the field of technologically viable ferroelectrics and piezoelectrics and established a strong fundamental scientific basis for these materials. During these decades, Ted Geballe[5] played a significant role in the discovery of advanced materials of all sorts, including oxides. Finally, the field of complex oxides would not be where it is now without the seminal contributions of K. Alex Müller,[6] Georg Bednorz,[6] and Bernard Raveau in the area of superconductivity. This "fast forwards" us to the 1980s and to high-temperature superconductivity.

The fascinating properties of complex oxides continue to infatuate and drive materials scientists to master both understanding and synthesis of the materials, with the goal of exploiting these properties in devices. In addition to superconductivity, the properties include ferromagnetism (materials where an applied magnetic field can cause the spontaneous magnetization of the material to flip) and ferroelectricity (materials where an applied electric field can cause the spontaneous polarization of the material to flip), and even materials that are both at the same time. The crystal structures of several of the key structural families of complex oxides are shown in Figure 1.[7]

The Impetus of High-Temperature Superconductivity

The advent of superconductivity in cuprates[6,9–16] at temperatures far higher than any previously known superconductors (making them "high-temperature" superconductors) has been, by far, the most significant development in the field of condensed matter sciences in the past several decades. It brought the field of complex oxides onto the main stage. The potential for applications of these materials in next-generation Josephson junctions for high-speed, low-power computing, sensitive magnetic field sensors, and high-frequency microwave filters stimulated the evolution and implementation of a variety of thin-film deposition techniques.

Venkatesan and co-workers pioneered the synthesis of complex oxides by pulsed-laser deposition.[17] This is a novel approach to create stoichiometric films and layered stacks of different complex oxides (heterostructures). The dynamic interaction of the material with short ultraviolet laser pulses circumvents the differences in vapor pressures of the constituents. The ability to flash evaporate the surface of a target enables this single-target method to produce films with nearly the same composition as the multicomponent target from which they are made. Molecular-beam epitaxy, a key tool that enabled the growth of semiconductor films following the crystalline template of the single-crystal substrate beneath them (semiconductor heteroepitaxy) in the 1970s and 1980s[66], was also employed to grow these multicomponent oxides.[18–20] In parallel, electron-beam evaporation,[21,22] sputtering,[23–27] and ion-beam deposition[28] evolved as viable physical vapor deposition techniques for complex oxides, and a strong effort toward the discovery of new organometallic precursors for chemical vapor deposition was initiated.[29]

The Example of Ferroelectric Memories

While the high-temperature superconductor revolution was emerging, other

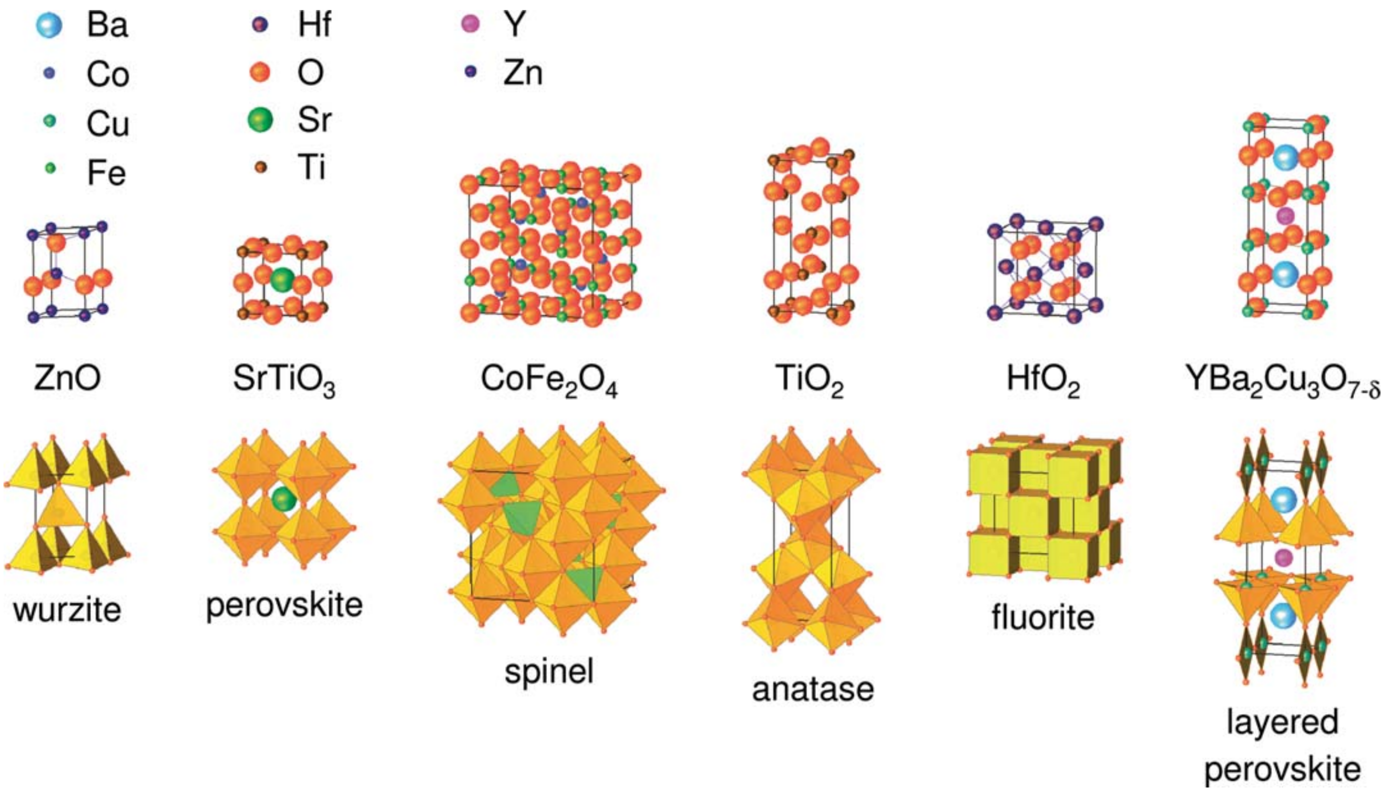

Figure 1. Examples of the crystal structures of complex oxides with striking properties that are the subject of the articles in this issue. Two equivalent representations of these crystal structures are shown: the atomic positions (top) and the coordination polyhedra (bottom). The oxygen atoms occupy the vertices of the coordination polyhedra. Color is used to distinguish the two types of oxygen coordination polyhedra in $CoFe_2O_4$, namely, octahedra (orange) and tetrahedra (green). The relative sizes of the atoms reflect their relative ionic radii as reported in Reference 7.

oxide-related fields were gaining prominence as well. The critical need for a high-density, solid-state memory technology was the key driver for researchers to explore ferroelectric thin films to store binary bits in the form of the two polarization states of a ferroelectric material.[30] Of course, the potential of ferroelectrics as memory elements had been recognized in the late 1950s.[31]

Three key issues limited the pervasive implementation of ferroelectrics in such technologies. First, these materials generally do not exhibit a well-defined switching threshold. Matrix-addressed memory architecture was becoming popular in the 1960s because of the emergence of semiconductor integrated circuit technologies, and in these devices, all of the bits would be subjected to at least one-half of the probing voltage. Under such conditions, the ferroelectrics would exhibit a finite probability of switching their polarization state, that is, the so-called "half-voltage disturb effect." This problem was solved by adding a transistor to each memory cell so that the write voltage could reach the ferroelectric memory element and alter its polarization state only when the transistor is activated. This pass-transistor architecture is now common in all solid-state memories.

The second issue was the switching voltage itself. Note that, in the 1960s, advanced thin-film deposition techniques for such complex materials were not available. Therefore, much of the work was carried out on bulk crystals, which needed switching voltages of ~100 V. Such high voltages were completely incompatible with the emerging memory technologies based on complementary metal oxide semiconductor (CMOS) materials, all of which used 5 V operating voltages. Once again, this issue was solved simply by changing to thin films.

By far, the most serious difficulty with implementing thin film-based ferroelectric memories was a fundamental materials issue related to the interactions of ferroelectric domains with the film microstructure, especially defects. This gave rise to two problems. The first of these was fatigue, which is a drop in the polarization with progressive switching. The second related issue was imprint, which is a preference of the ferroelectric storage element to remain in (or go back to) one of the two states. Both of these problems were related to how domains interacted with point defects as well as electronic defects, with the role of the electrode/ferroelectric interface being particularly critical.

In the early 1990s, a few groups began addressing these two issues. They explored conducting perovskites including cuprate superconductors,[32–34] $SrRuO_3$,[34,35] $(La,Sr)CoO_3$,[34,36] and $(La,Sr)MnO_3$.[37] In parallel, Kingon et al.,[38] as well as researchers at National Semiconductor, were exploring RuO_2, again a conducting oxide electrode material. In early 1991, both oxide-based approaches proved to be successful in solving the problem of fatigue. The key breakthrough was the recognition that charged oxygen vacancies (and, to a smaller extent, electronic defects) could segregate to the interface between the electrode (typically Pt) and the ferroelectric lead zirconate titanate (PZT) film, causing a space-charge layer that subsequently prevented switching; indeed, a similar defect-mediated mechanism is likely to be operative in causing imprint.

Are oxide electrodes the only approach to solving the problem of fatigue? This might not be the case, as the work of Scott

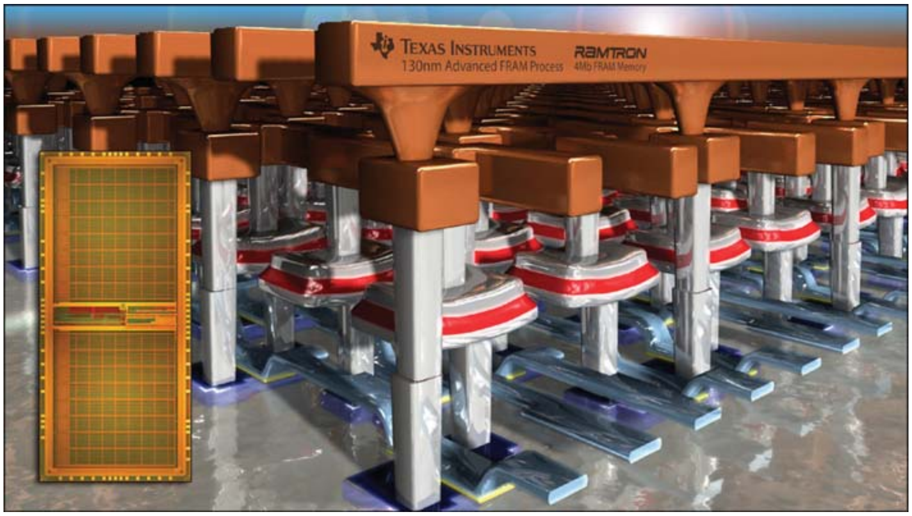

Figure 2. Schematic of the architecture of a 4-Mb ferroelectric random access memory device (from Texas Instruments/Ramtron).

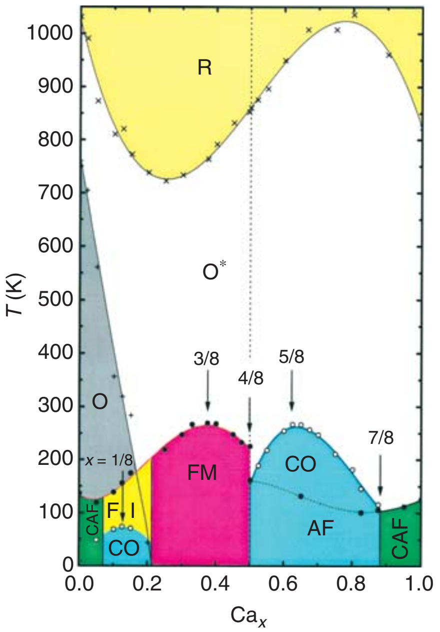

Figure 3. Phase diagram of hole-doped manganites. The parent structure is an antiferromagnetic insulator, but with doping, it becomes ferromagnetic and exhibits the colossal magnetoresistance effect. Analogous phase diagrams occur in high-T_c superconductor and relaxor ferroelectric systems. *Note:* AF is antiferromagnetic, FM is ferromagnetic, CAF is canted antiferromagnetic, CO is charge-ordered, FI is ferromagnetic insulating, R is rhombohedral, O is orthorhombic (Jahn–Teller-distorted), and O* is orthorhombic (octahedron rotated). (Reprinted from Reference 67 with permission).

and Araujo showed the possibility of using an alternative ferroelectric, $SrBi_2Ta_2O_9$, with Pt electrodes without any significant fatigue.[39] Thus, the electrode/ferroelectric interface is important, but the domain dynamics within the framework of defect interactions is also critical.

Translating these basic materials science discoveries into a manufacturable, commercial technology requires several steps and materials innovations. For example, these ferroelectric oxide capacitor stacks have to be integrated with a conducting plug directly on top of the pass transistor in the one-transistor–one-capacitor (1T–1C) cell of advanced ferroelectric memories.[40] Such a cell, which is currently being manufactured by Texas Instruments,[40] is shown in the image in Figure 2. The ferroelectric capacitor stack must be deposited by a scalable process suited to manufacturing, such as chemical vapor deposition or sputtering.

The capacitors also have to be defined through a dry etch process, another critical step. After the process is completed, the memory cell must be treated with forming gas (H_2/N_2 mixture) to clean up traps at the transistor interface. This step can destroy the ferroelectric oxide, and therefore, a robust passivation layer is needed. It is satisfying to note that the field has successfully solved all of these integration and reliability problems, thus paving the way to real, high-density memories. Several companies (such as Texas Instruments, Fujitsu, Toshiba, Panasonic, and Sony) are in the advanced stages of product development of high-density, embedded ferroelectric memories. This could be the emergence of the so-called "universal memory."

Renaissance of Correlated Electron Materials

The early 1990s witnessed the "rebirth" of correlated electron oxides, in this case through the manganites. High-temperature superconductors had already set the stage for this rebirth by providing both the intellectual framework and the experimental and theoretical tools to explore related systems. The work of Chahara et al.[41] and von Helmolt et al.[42] triggered the work of Jin and co-workers[43] in the pursuit of materials that exhibit dramatically large (dubbed "colossal") changes in transport properties upon the application of a magnetic field.[16] These early works attracted much attention around the world, setting the stage for an explosion of research activity that is yet to abate.

A model system of correlated oxides is the antiferromagnetic insulator $LaMnO_3$. Upon doping with holes (e.g., Sr substitution in the La site), the system becomes electronically inhomogeneous and is highly susceptible to perturbations and external manipulation.[44–46] As a consequence, the phase diagram of such a system (Figure 3) exhibits multiple phases that are susceptible to change through temperature; chemical doping; and electric, magnetic, and mechanical fields. The article by Dagotto and Tokura in this issue details the properties of these extraordinary materials.

A key aspect to keep in mind is the energy scale involved in creating these changes, as it directly impacts the ability to translate these discoveries into commercial technologies. For example, the colossal magnetoresistance (CMR) effect, the change in resistance accompanying the application of a magnetic field, is typically observed with a magnetic field of a few Tesla (1–10 T). This translates to a temperature scale of ~10 K, which is certainly a small value compared to the temperatures (200–380 K) at which the CMR effect peaks. On the other hand, creating such a magnetic field, say, 6 T, requires a current of several amperes running through a wire. Clearly, this is technologically prohibitive and is the main reason that products, such as magnetic sensors and read heads, involving CMR materials have not emerged.

This difficulty also provides some insight into exploring new materials in the future: If magnetic fields are to be used, then they have to be competitive with current technologies, such as giant magnetoresistance (GMR)-based magnetic tunnel junctions used in the read heads of hard disks.[47] On the other hand, if electric fields are used, then the large magnetoresistance effects observed at a magnetic field of 6 T can be obtained at fields on the order of a few hundred kilovolts per centimeter, which translates to a potential of a few volts across a 100-nm-thick film. A key conclusion emerges from this analysis: Electric-field-based modulation of properties is much more feasible if thin films and thin film heterostructures are used. This naturally leads us to the field of multiferroics and magnetoelectrics, which is described in more detail in the article by Spaldin and Ramesh in this issue.

Atomic-Scale Engineering: A Key Enabler of Oxide Electronics

Having started with vacuum deposition tools that have achieved unparalleled control in the integration of semiconductors at the nanometer level, it should come as no surprise that these same tools, once properly modified for the challenges of oxides, are now producing customized oxide heterostructures with control at the atomic level.[48–53] These epitaxial techniques allow for the preparation of ultrathin oxides that can be used for a range of purposes: studying the thickness at which ferroelectricity and other functional properties emerge,[54,55] determining ultimate scaling limits,[54–56] orienting oxides in directions that maximize a desired property, creating metastable oxides, integrating complex oxides with conventional semiconductors,[57] synthesizing oxide heterostructures containing electrodes and multifunctional active layers all in an epitaxial stack, and making oxide superlattices.[48–52] Even single interfaces can have surprising properties, for example, the conductive (and even superconducting) interface between the two insulators $SrTiO_3$ and $LaAlO_3$. See the article by Mannhart, Blank, Hwang, Millis, and Triscone in this issue for a full discussion of two-dimensional electron gases at interfaces.

An example of an oxide superlattice is shown in Figure 4a.[58] The perovskites $SrTiO_3$ and $BaTiO_3$ are alternately deposited to build up a 2–2 epitaxial composite, in which both constituents of the composite are connected in two dimensions.[59] Although $SrTiO_3$ and $BaTiO_3$ are fully miscible and form a complete solid solution in the bulk,[60] bulk diffusion is minimal at the low substrate temperature involved in oxide epitaxy (typically 700°C), and such atomically abrupt metastable heterostructures are readily grown. Such superlattices not only are relevant to fundamental science involving size, strain, and the coupling of order parameters between constituent materials, but are also being investigated for novel applications including acoustic cavities for terahertz modulators or phonon lasers.[61] 1–3 epitaxial composites,[59] where one composite constituent is connected in just one dimension and the other is connected in three dimensions, are also possible, and such composites with pillar dimensions as small as 20 nm have been demonstrated,[62] as shown in Figure 4b.

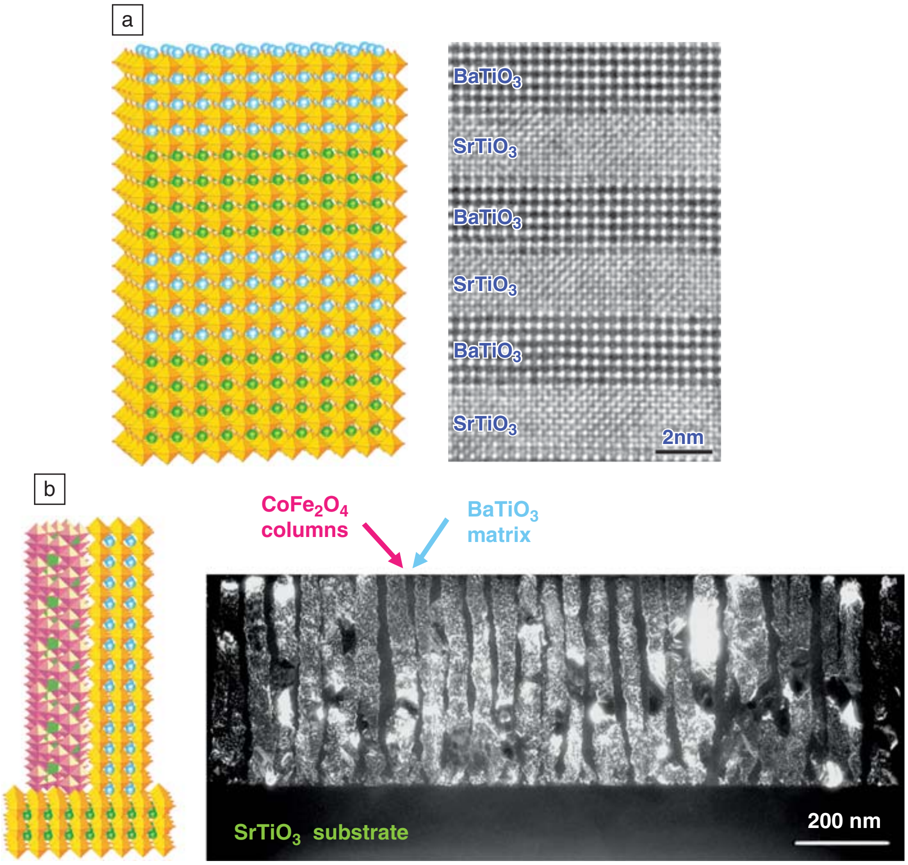

Figure 4. Examples of (a) layered epitaxial heterostructures (2–2 composites) and (b) epitaxial 1–3 nanocomposites produced by modern complex oxide growth methods. A transmission electron microscopy (TEM) image of each type is shown together with a schematic. (Figure 4a is reprinted from Reference 58 with permission. Copyright 2006, American Institute of Physics. Figure 4b is reprinted from Reference 62 with permission. Copyright 2004 American Institute of Physics.)

Because epitaxy is involved, huge (percent-level) strains can be imposed on oxide films. With sufficiently defect-free substrates, the crystalline perfection of heteroepitaxial films of complex oxides can rival that of single crystals of the same composition.[63] Finally, these same epitaxial oxides can be doped to create new functionalities. The article by Kamiya and Kawasaki in this issue addresses the topic of doping ZnO to make it magnetic and optically active, and the article by Coey and Chambers in this issue explores the controversial topic of magnetism in doped TiO_2 films.

Nonepitaxial heterostructures are also of interest, with a key recent technological breakthrough involving the replacement of the SiO_2-based gate dielectric in computer chips with a HfO_2-based alternative. Here, the heterostructures are amorphous and polycrystalline. The higher dielectric constant of this alternative gate dielectric, while maintaining the excellent electrical characteristics that made the SiO_2/Si interface the material of choice for metal oxide semiconductor field-effect transistors, provides for higher performance computers that operate at higher speeds and lower power. Its development is enabling transistor scaling (Moore's law) to continue for at least a few more years. Achieving this materials breakthrough involved overcoming a multitude of materials issues as described in the article by Schlom, Guha, and Datta in this issue.

The Future

So, what does the future of oxide electronics hold? We believe we are at the cusp of some dramatic developments in the science of complex oxides, especially in the form of thin-film heterostructures. At the fundamental level, the novel electronic phenomena that exist in superconducting cuprates and CMR manganites can be summarized into a rather simple theme: It is doping a well-behaved antiferromagnetic insulator with carriers (holes or electrons) or creating local inhomogeneities in charge distribution that leads to such physically interesting behavior as a consequence of a two-phase coexistence.

The next question is this: Can we employ this approach as a generic algorithm to explore other novel phenomena that occur at the phase boundaries and tap into phenomena that involve interactions between electron spin, electron charge, electron orbitals, and the underlying atomic structure of a material? For example, what happens if one dopes a multiferroic (which has both an antiferromagnetic and another order parameter)? What are the appropriate length and energy scales for doing so? Clearly, significant improvements in synthesis, especially in the control of chemical stoichiometry through the implementation of complementary surface analytical tools (e.g., electron diffraction coupled with simultaneous surface composition measurement through analysis of emitted characteristic x-rays[64]), will play a critical role in enabling us to probe such novel phenomena.

A major challenge for the field is control over oxygen stoichiometry. Indeed, this could be one of the most significant opportunities for the future. How does one control oxygen in the way that our semiconductor colleagues control dopants and impurities in Si and GaAs? Can one quantitatively determine and control the surface termination of oxide heterostructures during growth?

Harnessing Novel Properties for Potential Devices

An important future direction that will be particularly stimulated by the multiple functionalities in complex oxides is cross-coupling or decoupling of fundamental phenomena. Magnetoelectric and multiferroic effects provide one such example that deals with the coupling between electric and magnetic degrees of freedom. This is but the beginning. As we forge ahead, we need to keep in mind the fact that both science and technology must work together, as illustrated in Figure 5. We need to be cognizant of both the competition (at the technology and market level) and the critical issues that can limit the transition of basic science into technology.

Complex oxides are poised to play a key role in the rapidly emerging area of materials for energy technologies. One can envision creating oxide heterostructures and nanostructures that will enable the decoupling of electron and phonon transport, of particular relevance to the design of next-generation thermoelectrics.

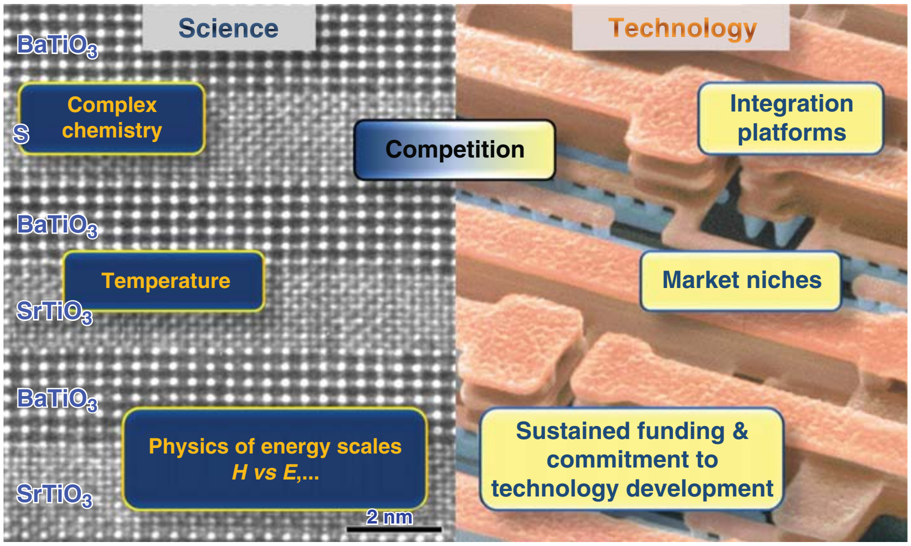

Figure 5. Illustration of the interplay between basic science and technology development. On the left side are shown some of the critical issues that need to be addressed at the fundamental level. On the right side are shown the factors that determine technology creation and commercialization. It is also clear that sustained resource commitment to both components is crucial for successful introduction of new fundamental discoveries into the technology stream. (The background of the left panel is reprinted from Reference 58 with permission. Copyright 2006 American Institute of Physics. The background of the right panel is courtesy of International Business Machines Corporation. Unauthorized use is not permitted.)

The ability to engineer artificial heterostructures (e.g., $SrTiO_3/LaAlO_3$) with tunable dielectric contrast as well as phonon characteristics is an attractive approach to creating phonon confinement and eventually phonon lasers. Indeed, ongoing work has already demonstrated the rudiments of these possibilities.[61,65] Another area of research is the conversion of photons to electrons, critical to photovoltaic solar energy conversion and photocatalysis for water splitting and CO_2 conversion to usable fuel. The multitude of exciting opportunities for oxides and especially oxide heterostructures will continue to inspire future generations of oxide electricians for decades to come.

Acknowledgments

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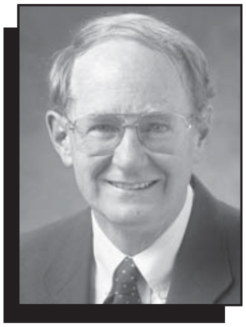
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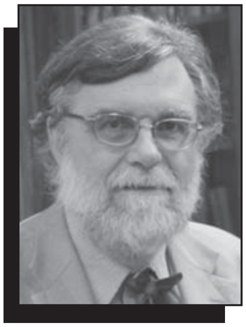
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