Electric field effect in correlated oxide systems

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Semiconducting field-effect transistors are the workhorses of the modern electronics era. Recently, application of the field-effect approach to compounds other than semiconductors has created opportunities to electrostatically modulate types of correlated electron behaviour—including high-temperature superconductivity and colossal magnetoresistance—and potentially tune the phase transitions in such systems. Here we provide an overview of the achievements in this field and discuss the opportunities brought by the field-effect approach.

Our daily life is permeated by semiconducting field-effect transistors (FETs). About 10^{18} of these microscopic electronic switches are produced every year to run the tools that are indispensable to us, including our cars, computers, cellular phones and kitchen appliances. These devices work by modulating the electrical charge carrier density, and hence electrical resistance, of a thin semiconducting channel through the application of an electric field. This remarkably simple and very successful principle provides new opportunities for basic science and innovative device applications when applied to novel correlated electron systems whose properties depend strongly on the carrier concentration. Excited by the opportunities of the field effect in such materials—including organic conductors[1,2], high-temperature superconductors[3,4] and colossal magnetoresistance compounds[5]—dozens of groups have worked on this approach, yielding a panoply of striking results[6]. The burgeoning field of plastic electronics is a notable example that has reached technological fruition[2].

These opportunities probably served as a motivation for the research directions of the group at Bell Laboratories[7]. Several of this group's articles in *Nature* and *Science*, for example, addressed electric-field-induced superconductivity in new materials. The public debate about the scientific misconduct that underlies a large part of this group's claims[7], however, may unfortunately and wrongly give the impression that the electric field effect in such materials does not exist. Here we provide an overview of the electrostatic field effect in complex oxides, one of the newest classes of materials to which the field effect has been applied. We consider the role of electronic correlations, discuss examples in superconductivity and magnetism, and outline future directions, commenting on the technical challenges of this approach.

Strongly correlated systems

In materials ranging from ionic insulators and elemental semiconductors to simple metals, the band structure model and associated Fermi liquid description provide a deep understanding of their fundamental physical properties. In this framework, insulators and intrinsic semiconductors have only filled and empty bands, whereas metals have at least one partially filled band. These concepts reach their limits, however, as the interactions between electrons become strong or the dimensionality of the system is reduced. When the electrostatic Coulomb interactions become dominant, localization occurs for commensurate filling (for example, 1/4 or 1/2 filling), resulting in a special type of insulator, frequently called the Mott insulator[8]. One-dimensional (1D) GaAs quantum wires, organic

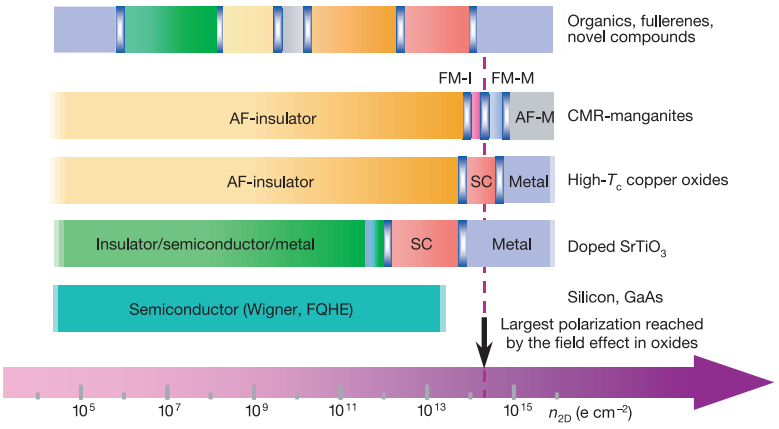

Figure 1 Illustration of the zero-temperature behaviour of various correlated materials as a function of sheet charge density (n_{2D}). Silicon is shown as a reference. The examples for high-T_c superconductors and for colossal magnetoresistive (CMR) manganites reflect $YBa_2Cu_3O_{7-\delta}$ and (La,Sr)MnO_3, respectively. The top bar has been drawn to illustrate schematically the richness of materials available for field-effect tuning and the spectrum of their phases. AF, antiferromagnetic; FM, ferromagnetic; I, insulator; M, metal; SC, superconductor; FQHE, fractional quantum Hall effect; Wigner, Wigner crystal.

Bechgaard salts, and undoped two-dimensional (2D) high-temperature superconductors are notable examples of such insulators[1,9,10]. Disorder in these correlated electron systems also has an important role. At low densities, depending on the balance between disorder and the strength of the electronic correlations, one expects to have Anderson localization (in 1D or 2D) or a crystallization of the electrons into a Wigner crystal; both schemes lead to an insulating state[11,12]. Recent work on 2D, high-mobility Si field-effect structures proposes that extremely strong electron interactions may overcome the disorder that leads to localization, resulting in a metallic ground state[13].

Interactions combined with low dimensionalities thus lead to novel phases, which are strongly influenced by the carrier concentration[14]. Figure 1 illustrates several of the properties of the materials discussed above as a function of their sheet charge density; silicon is shown as a reference. Because the carrier concentration is a crucial parameter that governs the properties of these systems, the electrostatic field-effect approach is an ideal tool to investigate the physics of correlated electron systems, allowing controlled and reversible changes of the carrier concentration without altering the disorder. The fractional quantum Hall effect, exhibited in GaAs/AlGaAs heterostructures, is a striking example of correlations in a clean, 2D system, in which the field effect has been used effectively to tune the carrier density[15]. Other methods to change the carrier density without affecting the disorder include the application of pressure and photon irradiation[16]. One challenge that is apparent from Fig. 1, however, is that much of the novel behaviour beyond the realm of the fractional quantum Hall effect and Wigner crystallization occurs at relatively high carrier densities, requiring the use of extremely large polarization fields for the field effect, as described below.

Experimental parameters

Field-effect experiments are conceptually elegant, but pose significant challenges in practice because the underlying physics requires structures that are difficult to achieve. Applying an external electric field to a material attracts or repels charge carriers, creating a thin charge accumulation or depletion layer at the surface that modifies the electrical conductivity between a source (S) and a drain (D) contact (Fig. 2). The field is applied across a gate insulator using a gate electrode (G). The characteristic width of the accumulation or depletion layer is given by the electrostatic screening length λ_{el}, which in the semiclassical, metallic limit is the Thomas–Fermi length. In standard metallic systems, λ_{el} is extremely short, a fraction of an atomic diameter, and thus negligible field effects are found. In low-carrier-density systems, larger screening lengths and field effects are expected, which are reasons why standard transistors are made with semiconductors rather than metals.

Many of the interesting physical properties described in Fig. 1 occur at carrier densities in the range of 10^{19}–10^{22} carriers cm^{-3}, intermediate between those of conventional metals and semiconductors such as Si or GaAs. To achieve substantial carrier modulation at these densities, two approaches are followed. The first is to use ultrathin drain–source (DS) channels, which are only a few nanometres thick, so that the absolute number of carriers is small, $\sim 10^{14}$ cm^{-2}, allowing large relative changes in their total number. Because the carrier modulation occurs in an interfacial layer, it is also advantageous to use ultrathin channels to avoid shunting effects. The second means of achieving large modulation of the carrier density is to apply extremely large electric fields across insulators that are tailored for large breakdown field strengths (E_b) and dielectric constants (ϵ_r). The polarization (σ), or areal charge density (charge cm^{-2}), that can be induced at a given gate voltage V_G applied across an insulator of thickness t is given by $\sigma = \int_0^{V_G} \varepsilon_0 \varepsilon_r(V)/t \, dV$, where $V_G < E_b t$. To change the carrier density by 50% in a 1-nm-thick oxide superconductor with 4×10^{21} holes cm^{-3}, for example, requires sheet charge densities or polarizations of 30 μC cm^{-2}, an order of magnitude larger than what can be achieved in standard FETs.

In addition to this stringent requirement on the sustainable polarization, leakage currents through the insulator must be much smaller than the DS current, and the density of localized interface states must be small compared with the modulated carrier density. In conventional Si FETs, the universal gate dielectric is SiO_2, an excellent insulator with low leakage that is chemically stable and virtually free of interface states. These outstanding properties of SiO_2 are a primary reason for the success of Si-based electronics. For oxide FETs, however, SiO_2 is unsuitable because its breakdown field $E_b = 10$ MV cm^{-1} and dielectric constant $\epsilon_r = 3.9$ lead to polarizations of 3 μC cm^{-2} (2×10^{13} charges cm^{-2}) at best. In the case of plastic electronics, this relatively small polarization does not preclude the use of SiO_2, although issues related to traps, surface states, degradation of the channel, and leakage had to be mastered. An additional difficulty for organic FETs is that the design and fabrication of the contacts to the DS channels are non-trivial.

Because of these technical challenges, chemical doping, which can achieve metallic carrier densities, is by far the most commonly used method to manipulate the carrier densities of complex oxides. Largely owing to advances in physical vapour deposition techniques, it is only in recent years that doping of complex oxides by the electric field effect has become possible. It is now routine to fabricate epitaxial high-ϵ_r complex oxide dielectrics, such as $SrTiO_3$, and ferroelectric oxides, such as $Pb(Zr,Ti)O_3$ (PZT), that can achieve polarizations in the range of 10–40 μC cm^{-2}. *In situ* multilayer growth of the oxide dielectric and DS channels has led to improved structural and electronic properties of the DS channel with acceptable interface trap densities[6,17,18].

Interestingly, these advances in complex oxide gate dielectrics have led the semiconductor industry to consider replacing SiO_2 with high-ϵ_r complex oxide materials. As the thickness of the SiO_2 layer approaches one nanometre, below which leakage from tunnel currents becomes unacceptably large, alternative gate dielectrics will become a necessity[19]. The possibility of using thicker gate dielectrics with larger values of ϵ_r is driving the exploration of complex oxide gate dielectrics, as well as the development of the methodologies to deposit these materials directly on Si. As with complex oxide FETs, challenges related to the density of interface states and the chemical and thermodynamic compatibility of the gate insulator with the DS-channel material are being addressed[20–22].

Despite the technical challenges of fabricating complex oxide FETs, the notion of switching exotic behaviour (such as superconductivity) on and off just by turning the knob on a power supply is so tempting that dozens of groups have addressed these obstacles, with experiments starting as early as 1960 (for example, refs 23–34; for an extended list, see http://www.physik.uni-augsburg.de/exp6/field-effect/). It was in 1991, working with high superconducting transition temperature (high T_c) copper oxide superconductors, that Mannhart and colleagues at the IBM Research Laboratory in Rüschlikon observed electric-field-induced shifts of T_c of several K in epitaxial heterostructures (Fig. 3a)[25]. Subsequent work using the ferroelectric field effect has also allowed reversible modulation of superconductivity to be induced in these materials[18] (Fig. 3b, c).

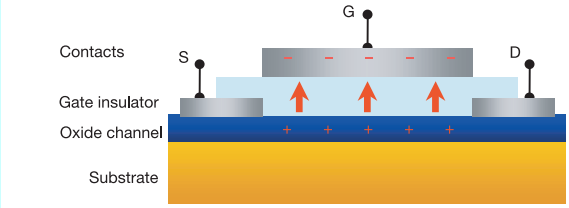

Figure 2 Cross-section of a typical sample geometry used for field-effect studies. S, source; G, gate; D, drain.

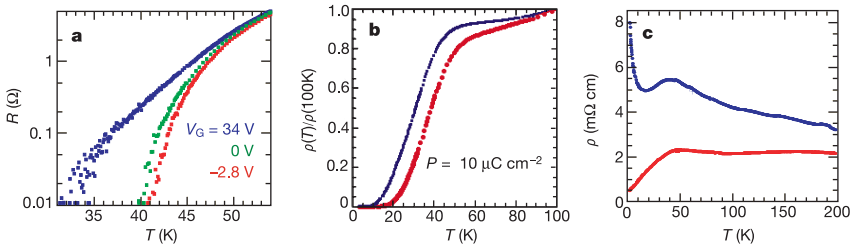

Figure 3 Field effects in superconducting films. In each case, the blue curve corresponds to depletion of the carrier density, and the red curve corresponds to enhancement of the carrier density in the DS (drain–source) channel. **a**, Change of the DS resistance of an ~8-nm-thick $YBa_2Cu_3O_{7-\delta}$ channel with a ~300-nm-thick $Ba_{0.15}Sr_{0.85}TiO_3$ gate insulator. The scatter in the data results from the noise of the measurement system (from ref. 6). **b**, Resistance change of a ~2-nm-thick $GdBa_2Cu_3O_{7-\delta}$ film induced by a 300-nm-thick PZT layer acting as ferroelectric gate. The two curves have been normalized in the normal state. **c**, Resistance change of a ~2-nm-thick $GdBa_2Cu_3O_{7-\delta}$ film whose doping level has been chosen to be close to the superconductor–insulator transition, induced by a 300-nm-thick PZT layer acting as ferroelectric gate. These data have been measured at 1 T (from ref. 18).

Correlated oxide superconductivity

We now turn to the role of charge in the physics of the high-T_c copper oxide superconductors. In transition metal oxides with a partially filled *d*-electron band, the electronic correlations lead to the opening of a direct gap in the 3*d* band (Mott–Hubbard insulators) or an indirect gap between the oxygen 2*p* band and the upper Hubbard band (charge transfer insulators); both cases result in an insulating state at half filling. The high-T_c superconductors are doped charge transfer insulators, and the role of carriers is depicted in the temperature–doping (T–x) phase diagram of Fig. 4. The parameter x stands for the doping level, which is usually changed by non-isovalent chemical substitution of the insulating $x = 0$ parent compound, and is roughly proportional to the number of 'free' electrical carriers in the system. At low doping levels, the system is an antiferromagnetic insulator owing to strong electronic correlations. As the doping level is increased, superconductivity appears, with the transition temperature T_c following a roughly parabolic dependence on x.

At low doping levels, near the superconductor–insulator phase boundary, field-effect tuning of the carrier density should induce a reversible insulator–superconductor quantum phase transition, an active area of research in condensed matter physics[35,36]. Such superconducting switching behaviour is appealing from both a fundamental perspective and for potential applications. Figure 3c shows ferroelectric field-effect experiments on underdoped, ultrathin (20 Å) $GdBa_2Cu_3O_7$ films, in which non-volatile, reversible switching between superconducting and insulating behaviour could be achieved by modulating the polarization state of the ferroelectric gate[18]. This switching occurs at a very large value of the sheet resistance (about 40 kΩ per square), in agreement with values obtained by chemical doping and disorder-induced superconductor–insulator transitions in high-temperature superconductors, but significantly larger than the values observed in low-temperature superconductors. On the insulating side, analysis of the transport properties reveals a resistivity described by $\rho \propto \ln(1/T)$. This peculiar temperature dependence was also observed in very-high-magnetic-field (60 T) experiments that allowed superconductivity to be suppressed in underdoped $La_{2-x}Sr_xCuO_4$ and other compounds[37]. These results suggest that the physics of the insulating state reached by high electric or magnetic fields is different from that obtained by chemical doping, which may be related to the fact that even as the superfluid density is modulated by the applied electric or magnetic field, the microstructure and disorder remain fixed.

In addition, in the underdoped region of the T–x phase diagram, the copper oxide superconductors have a relatively low carrier density, resulting in a low superfluid density and weak phase stiffness. It has thus been proposed that the T_c in this region is controlled by phase fluctuations[38,39]. As a result, T_c only reflects the establishment of long-range phase coherence and should be proportional to the superfluid density n_s. This relationship has been observed experimentally in muon spin resonance experiments on chemically doped bulk samples[40]. Recent ferroelectric field-effect experiments on underdoped thin films of $NdBa_2Cu_3O_{7-\delta}$ have also correlated quantitatively changes in T_c with changes in the carrier concentration produced by the field effect, in agreement with the phase fluctuation model[41]. In contrast to chemical doping, the field-effect experiments only modify the charge, revealing directly the relationship between the carrier density and T_c.

Electrostatic modulation of the carrier density has also been used to study the normal state of the copper oxide superconductors above T_c. Besides the opening of a gap in the electronic spectrum (called the pseudogap) at a temperature T^* (Fig. 4), it was recognized early on that the normal-state transport properties are anomalous, in particular the temperature dependence of the resistivity and Hall constant R_H. In ferroelectric/superconducting heterostructures, analyses of the normal-state temperature dependence of the resistivity and Hall constant as a function of the ferroelectric polarization show that both these quantities can be rescaled over the entire measured temperature range[42]. These results point to a temperature-indepen-

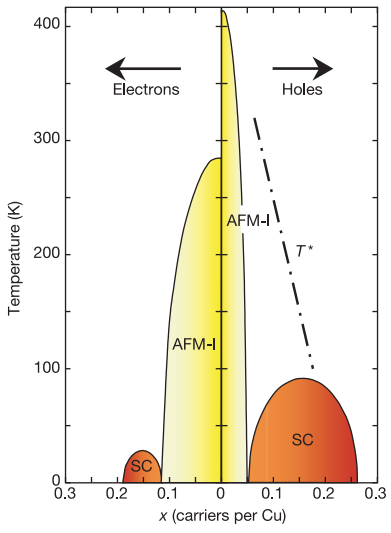

Figure 4 Phase diagram of the high-T_c superconductors. The electron-doped material represents $Nd_{2-x}Ce_xCuO_{4-y}$, and the hole-doped system represents $YBa_2Cu_3O_{7-\delta}$. The insulating, antiferromagnetic phases (AFM-I) are drawn in yellow, and the superconducting phases (SC) are in orange.

dent carrier concentration, with the observed temperature dependence of the Hall constant possibly being related to the temperature dependences of the scattering times and effective masses.

New approaches

Although most field-effect work on complex oxides has focused on superconductors, the approach has been extended recently to other materials. Manganites exhibiting colossal magnetoresistance (CMR) reveal large changes of their magnetic and transport properties induced by electric fields[43–45]. These experiments show that charge modulation at fixed disorder influences the CMR effect, whereas existing theoretical models have highlighted the role of disorder. One can also consider the possibility of achieving electric-field-tuned metal–insulator phase transitions, an idea that is being explored for applications based on oxide channel FETs operating at room temperature[46,47]. In addition, the field effect in perovskite-based organic–inorganic hybrid materials is being examined to further improve the performance of low-cost thin-film transistors[2]. In these approaches, materials preparation remains a key challenge. As with the development of semiconductor heterostructures, which took many years but has provided spectacular dividends, it will take a sustained effort—including the development of new fabrication techniques and advanced nanoscale characterization tools—to spur further advances in the observation of novel field effects in correlated oxide systems.

What are the future prospects for the field? The diversity of behaviour exhibited by new materials with correlated electrons is so rich that the field effect offers many possibilities. In particular, the polarization fields that are now routinely achievable using ferroelectrics and high-ϵ_r complex oxides provide access to largely uncharted territory, where the interplay between charge at high carrier densities in clean systems and strong Coulomb repulsion becomes important. Nanoscale control of electronic properties using field effects is also an emerging area. Recently, scanning probe microscopy has been used to induce localized field effects in magnetic and superconducting oxides[42,48]. The possibility of controlling the electronic properties of materials with nanometre resolution will allow the examination of fundamental issues underlying the behaviour of correlated oxides, such as nanoscale electronic phase separation[49]. It would also facilitate the optimization of physical properties through reversible nanostructuring. In future superconducting devices, nanoscale pinning sites could be introduced, and Josephson junctions and arrays of junctions could be written into superconductors. Combined with engineering breakthroughs taking place in nanotechnology, such as the fabrication of high-density arrays of scanning probes[50], novel device and memory structures could be fabricated. The field effect, which has been so spectacularly successful in its application to semiconductors, is only in its infancy in exotic correlated systems; the combination of advanced twenty-first-century techniques with this new materials approach offers broad perspectives for basic science and technology. □

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