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Electron glasses

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Summary. Electron glasses are disordered insulators with long-range interactions that exhibit slow non-equilibrium electronic transport effects. After introducing the basic physics of disordered insulators and hopping conduction, I briefly review the experimental evidence of glassy dynamics in these systems and some of the theoretical work aimed at understanding its origin. Similarities and differences with structural glasses and with another glass of electronic origin, the spin glass, are pointed out. [**Contrib Sci** 11(2): 163-171 (2015)]

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Introduction

In condensed matter physics, the term *glass* denotes a variety of systems that, due to their very slow dynamics, fail to reach thermal equilibrium on any reasonable experimental time scale. *Structural glasses* (such as the silica glass in windows), produced by quickly cooling a liquid so that it cannot crystallize, its molecules becoming trapped in an amorphous configuration, are the best known example.

This article is a non-technical introduction to a different type of glass in which electrons, rather than molecules, display a slow collective dynamics. Because of their light mass, one usually thinks of electrons as fast particles. However, experimental studies of electronic transport in various kinds of disordered insulators have shown that their electrical conductivity can relax over huge time scales, up to 20 orders of magnitude larger than the microscopic relaxation time [1,6,12,13,22,24,30,33]. Understanding the origin of the slow

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dynamics in these materials, collectively known as *electron glasses*[1] [1,8,30] or as *Coulomb glasses*, remains an outstanding challenge.

Below I briefly recall the main predictions of the band theory of solids, and then discuss how these are modified by disorder and by the interactions between electrons. I then briefly review the hopping conduction mechanism in disordered insulators, some of the experimental evidence of glassy relaxation, and recent theoretical work aimed ad understanding it.

Normal metals and band insulators

The band theory of solids predicts whether a crystalline solid is a metal or an insulator, based on its chemical composition. In a perfect crystal, the allowed energy levels of non-interacting electrons form *bands* separated by forbidden gaps; at an absolute temperature of zero, the electrons fill the levels from the bottom up. If some bands are filled only partially, the electrons near the Fermi energy E_F (the energy of the highest occupied level) move freely through the crystal as waves. This defines metallic behavior. At temperature $T > 0$, crystal vibrations (phonons) scatter the electron waves in random directions, thus reducing the electrical conductivity $\sigma(T)$ as the temperature is increased.

Partially filled bands arise if each crystal unit cell contains an odd number of electrons, or if two bands overlap. If neither of these conditions is met, then all bands are either empty or full and, since quantum mechanics forbids a net displacement of electrons in a full band, $\sigma(T)$ vanishes as $T \to 0$, which is the defining property of an insulator. As the temperature is increased, more and more electrons in the valence band (the full band of highest energy) receive enough energy from the crystal vibrations to overcome the band gap E_g, allowing them to populate the empty conduction band above it (Fig.1). This gives rise to a strong increase of the conductivity with temperature, $\sigma(T) \propto \exp(-E_g / 2k_B T)$, where k_B is the Boltzmann constant. Nervertheless, the conductivity remains negligible compared to metals (for example, at room temperature it is 20 orders of magnitude smaller in diamond than in copper).

Disorder and interactions: the metal-insulator transition

Band theory is remarkably successful even though it neglects two key features of real solids: disorder, which breaks the translational symmetry of perfect crystals, and the repulsive Coulomb interaction between electrons.

In metals, the average interaction energy between conduction electrons is not negligible compared to their kinetic energy. Nevertheless, Landau's theory of *Fermi liquids* showed that, in normal metals, the correlations between electrons are sufficiently weak that they do not destroy metallic conductivity, thanks to efficient screening[2] and to the Pauli exclusion principle. The main effect of interactions is to cause electrons to scatter each other, reducing $\sigma(T)$ at low temperature.

Certain materials, however, do not behave as Fermi liquids, an example being high-critical-temperature cuprate superconductors. Another notable example is represented by Mott insulators, which should be metallic according to band theory but behave as insulators because the Coulomb repulsion prevents an electron from traveling to a site occupied by another electron, thus splitting the metallic band into a lower full band and an upper empty band. A sharp transition from insulator to metal, the Mott transition [18], occurs when the width of both bands becomes large compared with the intra-site Coulomb energy and thus the bands merge. The transition can be induced by applying external pressure to affect a stronger overlap, thus broadening the bands.

A certain amount of disorder, in the form of impurities or crystal defects, is unavoidable in crystals. Furthermore, inherently disordered, non-crystalline materials are found in many technological applications. In these systems, disorder creates a random potential that scatters the electron waves. If the random potential is small compared with the electron kinetic energy, then metallic behavior is preserved: the conductivity, albeit reduced by disorder, remains finite down to zero temperature. On the other hand, Anderson [2] predicted that for strong enough disorder the electrons become trapped in small regions of typical size ξ (the localization length). The system then behaves as an *Anderson insulator*:

1 Electron glasses were a major topic of the 15th International Conference on Transport in Interacting Disordered Systems (TIDS15), organized by the author in Sant Feliu de Guíxols (Girona) in September 2013. The proceedings are published in Ref. [25].

2 Self-screening of the Coulomb interaction is based on the fact that electrons push away other mobile electrons and are thus surrounded by a net positive charge. In metals, due to the high mobility of electrons, the electrostatic potential created by a point charge decays exponentially with distance as $V(r) \propto \exp(-r / l)$, where *l* is the screening length, rather than as the inverse of the distance as in the unscreened case.

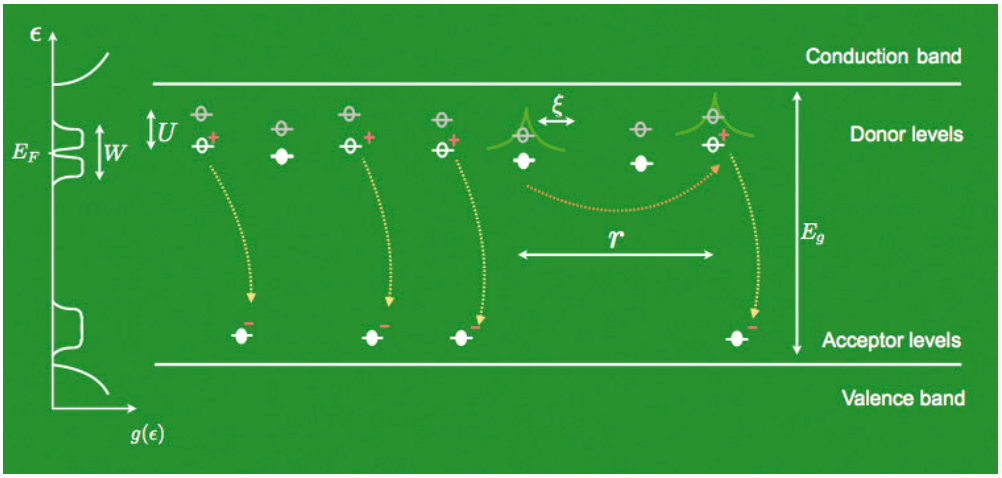

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Fig. 1. On the right, a pictorial representation of the energy levels in a lightly doped, n-type compensated semiconductor in the Anderson insulating regime. The abscissa represents position (drawn in one dimension) and the ordinate represents energy. Full circles represent neutral donors and charged acceptors. Empty circles represent donors that have lost an electron to a nearby acceptor (dotted yellow line). E_g is the energy gap between the valence band and the conduction band. The acceptors act as "spectators" and do not participate in conduction, but they create a random potential of width W. When W is large compared to the quantum mechanical bandwidth, the states in the impurity band are localized and the electronic wave functions decay exponentially over a characteristic distance ξ. Electrons can tunnel from occupied to empty donors at distance r and energy difference ΔE. Also shown is the upper donor impurity band, separated by the lower band by the intra-site Coulomb energy U. The density of electronic levels $g(\varepsilon)$ is represented schematically on the left: the depletion near the Fermi energy E_F is the Coulomb gap.

the conductivity vanishes at $T=0$ because localized electrons cannot diffuse. A sharp transition from metal to insulator, the Anderson transition, occurs upon reducing the electron concentration (thus decreasing the kinetic energy) or upon increasing the disorder [18]. Loosely speaking, the transition occurs when the mean free path of the conducting electrons (the average distance they travel between two scattering events) becomes comparable to their wavelength.

The Mott and Anderson transitions, driven by interactions and disorder, respectively, are examples of zero-temperature quantum phase transitions.

Hopping conduction in disordered insulators

At finite temperature, Anderson insulators conduct electricity via quantum-mechanical tunneling of the electrons between localization sites. This conduction mechanism, called *hopping*, is made possible by crystal vibrations that emit or absorb the energy difference (ΔE) between the initial and final electronic states. The transition rate of a single electron tunneling between two sites separated by distance r is proportional to $\exp(-2r/\xi-\Delta E/k_BT)$ for $\Delta E>0$, where the exponential dependence on r arises from the overlap between the wave functions of the two states. Mott [19] predicted that as T is lowered, the most probable hops take place between sites with increasingly small ΔE and increasingly large r, a mechanism called *variable-range hopping* [18,32]. Consequently, when the long-range Coulomb interaction can be neglected, the conductivity follows the Mott law $\sigma(T)\propto\exp[-(T_M/T)^{1/(d+1)}]$, where d is the system dimensionality and T_M is a characteristic temperature that depends on ξ and on the density of electronic levels at the Fermi energy (provided the density does not deviate much from this value throughout the hopping energy range).

Due to the low mobility of electrons, the Coulomb interaction is poorly screened at low temperature and thus, unlike in metals, it retains its long-range character, inducing strong correlations in the motion of the electrons. The best known correlation effect is the Coulomb gap, a reduction of the density of levels near the Fermi energy [9,29], which in turn reduces the conductivity (Fig. 1).

Efros and Shklovskii [9] argued that at $T=0$ the single-particle density of levels $g(\varepsilon)$ must vanish proportionally to $|\varepsilon-E_F|^{d-1}$ or faster as $\varepsilon\to E_F$, and predicted that if this bound is saturated, the conductivity follows $\sigma(T)\propto\exp[-(T_{ES}/T)^{1/2}]$, where T_{ES} is a universal tempera-

ture that depends only on ξ. A change from the Mott law to the Efros-Shklovskii law has been since been confirmed experimentally for a wide variety of materials. The original argument of Efros and Shklovskii [9] considered only single-electron hops that leave the rest of the system unperturbed. However, a full understanding of hopping conduction requires the consideration of multi-electron hops (i.e., the simultaneus tunneling of more than one electron), and of sequential hops induced by the Coulomb interaction after a hop [14,29]. In particular, it has been argued that sequential hops cause the density of levels to vanish faster than quadratically in three dimensions [5], and that many-electron transitions are responsible for glassy non-equilibrium relaxation [1,15,24].

Next, I describe several types of Anderson insulators exhibiting hopping conductivity.

Doped crystalline semiconductors. In a semiconductor (a band insulator with a relatively small band gap) doped with donor impurities[3], the ground-state energy levels of electrons bound to the impurities form a narrow "impurity band" below the conduction band [18,32] (Fig. 1). At room temperature, most impurities are ionized by thermal fluctuations, thus populating levels in the conduction band. These levels being delocalized, conduction occurs in the same way as in metals. At temperatures so low that ionization is negligible, conduction occurs only within the impurity band, which is half-filled since each impurity contributes one electron and two spin-degenerate levels. (An identical picture holds if the impurities are acceptors, except that the impurity band is near the valence band and charge is carried by holes instead of electrons.)

The width of the impurity band arises from two facts: i) quantum broadening spreads the levels by an amount I_b that increases upon reducing the average distance between impurities; ii) because of the random spatial distribution, the impurities experience a random potential which spreads the levels by an amount W. In the impurity conduction regime, the only relevant energy scales are I_b, W, the thermal energy $(k_B T)$, and the intra-site Coulomb repulsion energy (U) between two electrons bound to the same impurity.

If only impurities of one type (e.g., donors) are present, they remain electrically neutral and thus the random potential is typically small, i.e., $W << U$. For $I_b >> U$ the impurity

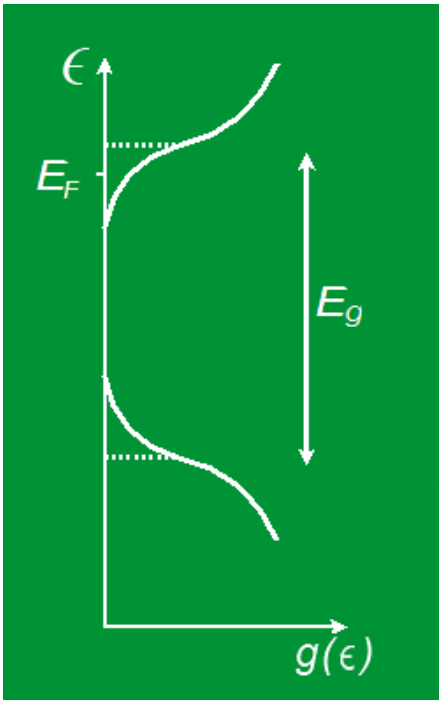

Fig. 2. Pictorial representation of the density of levels in an amorphous semiconductor. The dashed lines represent the mobility edges in the tails of the conduction (valence) band, below (above) which states are localized.

states are delocalized; thus, conductivity is metallic in the sense that $\sigma(T=0)>0$. By decreasing the impurity concentration, a Mott transition occurs as the ratio I_b / U falls below a certain threshold. In the Mott insulating regime, hopping conduction is not possible since there are no empty donor sites [18].

On the other hand, if both donors and acceptors are present, for example with a slightly higher concentration of donors (n-type compensated semiconductor), then each acceptor captures an electron from a nearby donor and becomes charged (Fig. 1). The charged impurities create a strong random potential, and typically $W >> U$. If the impurity concentration is so high that $I_b >> W$, the states are delocalized and conduction is metallic. Upon reducing the concentration, an Anderson localization transition occurs at a certain threshold for the ratio I_b / W. In the Anderson insulating regime, pho-

3 Donors (resp. acceptors) are atoms with one more (less) valence electron than the semiconductor atoms; thus, they "donate" an electron (hole) that, if the impurity is isolated, remains weakly bound to the impurity core, forming a hydrogen-like "atom" whose radius can be much larger than the crystal lattice spacing. Examples are phosphorous (boron) in a silicon crystal.

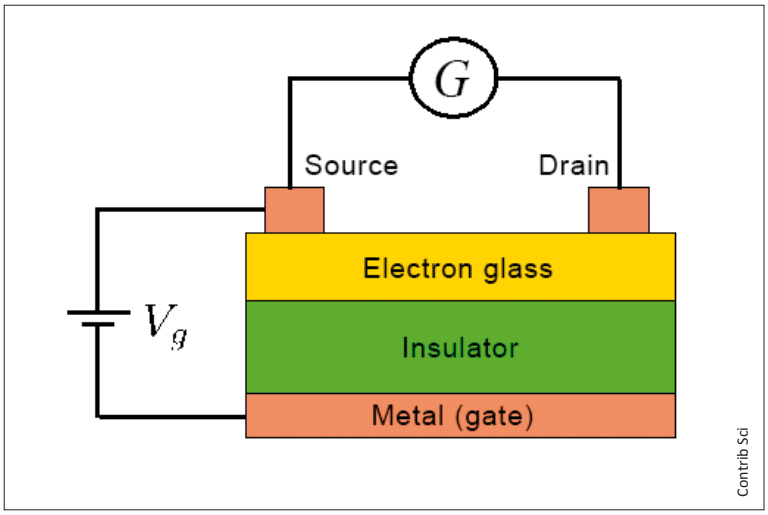

Fig. 3. Scheme of the field effect transistor experimental setup used in conductance experiments. The Anderson insulator under study is deposited on an insulator layer in contact with a metal (the gate). The conductance is measured by applying a voltage between the source and drain electrodes attached to the sample. A voltatge V_g between the gate and the source allows a change in the density of the carriers in the sample. Capacitance experiments use a similar setup except that no source-drain voltage is applied and the current between the gate and the insulator is measured as a function of time.

non-assisted hopping from occupied to empty donor sites can take place.

Amorphous semiconductors. These are covalent structural glasses prepared by rapid cooling of a melt or by deposition. Although they lack translational symmetry, a band structure still exists [18], but tails of localized states appear in the band gap, separated from the extended states by a *mobility edge* (Fig. 2). If the Fermi energy falls within the localized region, the system is an Anderson insulator and conduction takes place via variable-range hopping between localization sites. A metal-insulator transition can be driven by pressure, an increase in the carrier concentration, or thermal annealing.

Granular metals. They are produced by embedding nanometric metallic grains in an insulating material. Electronic transport takes place via tunneling between grains. Below a certain average inter-grain separation, conduction is metallic even if the metal does not percolate the sample. Above this threshold, conduction is of the hopping type. Discontinuous metallic films, consisting of metallic islands deposited on a substrate, behave similarly [30].

Two dimensional systems. In *field effect transistors* (Fig. 3), the charge carriers of a semiconductor can be confined in a narrow 2D layer at an interface with an insulator. The interfacial electronic states display a mobility edge, and by tuning the carrier concentration the Fermi energy can be brought in the localized region, giving rise to a 2D Anderson insulator. Homogeneous ultrathin metallic films, in which localization occurs below a certain thickness, are another example of a 2D Anderson insulator.

Self-assembled arrays of metallic/semiconductor nanocrystals. These consist of nanometric grains coated with an insulating material that self-assemble in a colloidal solution. Variable-range hopping due to tunneling between grains has been experimentally observed [16]. These materials are currently of great interest as an alternative to semiconductor technology for electronic devices.

Glassy behavior

The idea that Anderson insulators with long-range interactions may exhibit glassy dynamics was first put forward in theoretical work by Davies et al. [8], who, by analogy with spin glasses, coined the term “electron glass” for these systems. Spin glasses are disordered magnetic materials in which the interaction between two given magnetic moments (spins) can be, depending on their positions in the solid, either ferromagnetic or antiferromagnetic, favoring a parallel or antiparallel orientation of the spins [34]. Because of the random mixture of signs, the spins cannot satisfy all interactions simultaneously, a fact referred to as frustration.

The interplay of disorder and interaction gives rise to frustration in electron glasses as well: disorder tends to push the electrons to sites where the random potential is large, while interactions tend to push them away from each other. In both spin and electron glasses, frustration induces a large number of metastable states (i.e., local energy minima) in which the

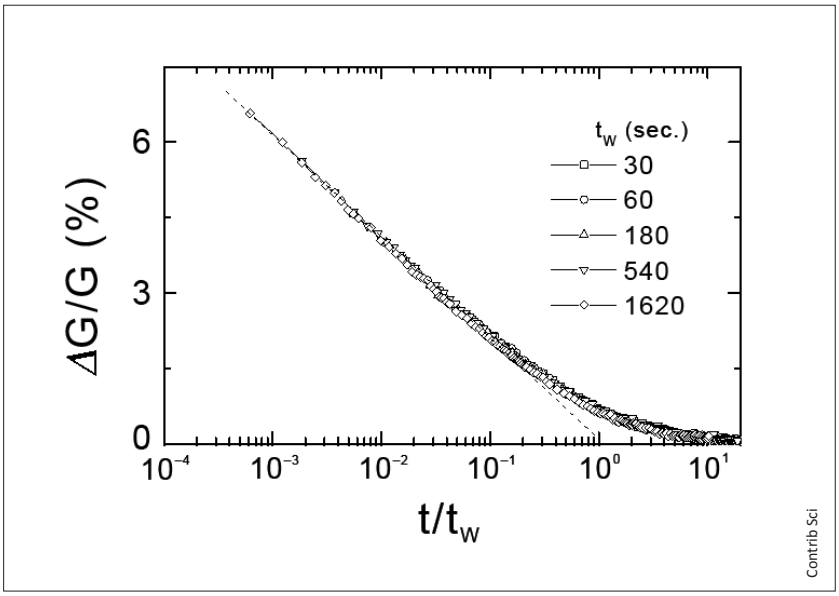

Fig. 4. Experimental results using the protocol described in the text for a film of crystalline indium oxide with a thickness of 5 nm, evaporated on a 140-μm-thick cover-glass coated on the back with a gold film. Here $t_1 = 6$ days, $V_{g1} = 50V$, $V_{g2} = -50V$. The plot shows the relative change in conductance $\Delta G(t)/G(t_w)$ where $\Delta G(t) = G(t+t_w) - G(t_w)$, as a function of t/t_w. Data for several values of t_w are shown together. Figure adapted with permission from [24]. Copyright of the American Physical Society.

system can become trapped for a very long time. In this respect, electron and spin glasses differ from structural glasses, since in the latter there is no external disorder and slow dynamics is generated only by interactions.

Early observations of slow relaxation in semiconductors [17] were followed by the groundbreaking experiments of Ovadyahu and collaborators on amorphous and crystalline indium oxide [6,24]. These experiments typically use a field effect transistor setup (Fig. 3) and monitor the time evolution of the conductance after the system has been pushed out of equilibrium, for example by a sudden change in temperature or gate voltage.

Figure 4 shows representative results obtained with the following protocol [24]: the sample is cooled at liquid helium temperature ($T = 4.11K$) and kept at a gate voltage $V_g = V_{g1}$ for a time t_1. The gate voltage is then switched to $V_g = V_{g2}$ for a "waiting time" t_w, after which it is finally switched back to $V_g = V_{g1}$. At both changes of the gate voltage, the conductance increases abruptly and then slowly relaxes. The figure shows the conductance relaxation after the second change, where t is the time passed since the change. A few observations can be made: (i) The rapid conductance increase occurs regardless of whether V_g was increased or decreased (i.e., whether electrons are injected or removed), unlike in ordinary semiconductors in which the conductance is monotonic in V_g. (ii) The relaxation curve depends on t_w, showing that the system has not equilibrated during the time t_w, even when the latter is large. This is referred to as "aging" in the glass literature. (iii) t_w enters only via the ratio t/t_w ("simple aging" or "full aging.") (iv) the relaxation follows a logarithmic law $\Delta G(t) \approx \log(1 + t/t_w)$ for several decades of time.

A similar behavior has been observed in amorphous indium oxide, thin beryllium films, granular aluminum [12], and discontinuous metallic films [13], also for much longer waiting times. (Recent reviews can be found in [1,22,30]). A rapid increase of the conductance followed by logarithmic relaxation has been reported with other types of perturbations, such as rapid cooling, the application of strong electric fields, and exposure to infrared radiation [23,30]. More complex protocols have been used as well [12,24], in particular to investigate how the system "remembers" its previous history over long time scales (the so called memory effect) [1].

An important observation from these studies is that the relaxation time τ (defined, for example, as the time necessary for $G(t)$ to decrease to half its peak value after a perturbation) increases rapidly with the carrier concentration [33]. This fact provides strong evidence that glassiness is an intrinsic property of the electron dynamics and is not due to extrinsic relaxing elements, such as slowly moving charges in the substrate or structural rearrangements. It might also explain [22] why glassy effects of the type described above have not been observed in lightly doped semiconductors, which have a much lower carrier density than all the other Anderson insulators discussed above.

Another crucial finding is that τ does not depend appreciably on temperature, in contrast to both structural glasses and spin glasses, in which this dependence is strong[4]. A likely explanation lies in the fact that in structural and spin glasses

the dynamics is thermally activated (i.e., the transition rate between two configurations is proportional to $\exp(-\Delta E / k_B T)$), while in electron glasses it depends on both quantum tunneling and thermal activation. The temperature independence of τ thus suggests that quantum tunneling plays a key role in glassy relaxation.

Expert readers may note that the time protocol discussed above is very similar to the "isothermal remanent magnetization" protocol used in spin glass experiments [7]. Aging is observed in spin glasses as well, but the relaxation is not logarithmic and the dependence on t and t_w is more complicated. The reason for this different behavior of spin and electron glasses is not fully understood.

Recent theoretical work

The origin of slow non-equilibrium relaxation in electron glasses is still actively debated, and it is beyond the scope of this introductory paper to discuss the many theoretical ideas that have been put forward [30]. Only a few aspects are mentioned below.

Many-electron transitions. Because the Coulomb gap strongly affects the stationary conductivity, the slow non-equilibrium relaxation of the conductivity is sometimes attributed to the slow formation of the Coulomb gap after having been disrupted by a perturbation. Recently, by means of kinetic Monte Carlo simulations of transport in a model electron glass in which only single-electron transitions are allowed, we found [10] that after an abrupt temperature quench the conductivity reaches a stationary value $\sigma_{eq}(T)$ on a time scale of order $\tau_M \approx (T^2 \sigma_{eq}(T))^{-1}$ (the Maxwell relaxation time), and that indeed the Coulomb gap forms on the same time scale. However, the measured τ_M is at most $10^{-6}\,\mathrm{s}$, many orders of magnitude smaller than the relaxation time observed experimentally in a comparable temperature range. This suggests that glassy dynamics depends on the simultaneous tunneling of more than on electron [1,15]. Multi-electron transitions are slow processes, as the transition rate decays exponentially with the total distance traveled by the electrons. The most probable transitions involve small clusters of sites, thus they do not participate directly in conduction, which occurs via long single-electron transitions. However, it has been conjectured [1,15] that when these clusters relax, they lower the conductivity by affecting the conduction path via the Coulomb interaction.

Search for a thermodynamic phase transition. A long-debated question in spin glass physics concerns the existence of a thermodynamic transition to a low temperature "spin glass phase" [34], in which each spin points in a preferential orientation (unlike in the paramagnetic phase in which spins spend, on average, the same time in all orientations), but the pattern of orientations, dictated by the interactions, has a random appearance. It is now well established [3,26] that in three dimensions and in the absence of an external magnetic field the spin glass phase exists, although its precise nature is still debated.

In their seminal paper, Davies et al. [8] conjectured the existence of a similar phase in electron glasses. A mean-field theory [20,28], inspired by a mathematical similarity between electron glass models and long-range spin glass models, predicted a transition to a "marginally stable" low temperature phase in three dimensions. A consequence of marginal stability is that the density of levels goes to zero quadratically at the Fermi energy, saturating the Efros-Shklovskii bound [9]. By means of large-scale equilibrium Monte Carlo simulations, however, we found convincing evidence against the existence of a phase transition down to very low temperatures [11]. Furthermore, using energy minimization computations with large system sizes we determined that density of levels in the Coulomb gap vanishes faster than quadratically [27], confirming some earlier findings (references can be found in [27]).

Avalanches and nonlinear screening. Screening in a system of localized electrons is very different than in a metal [4]. Because charge is discrete, weak electric fields can penetrate over large distances, a phenomenon referred to as nonlinear screening. Capacitance experiments using a metal-insulator-semiconductor structure (Fig. 3) found evidence that the charge injected in an Anderson insulator through a gate voltage moves very slowly from the "top" to the "bottom" of the sample [17].

Motivated by these observations, we recently investigated numerically the "avalanches" created by inserting or displacing a charge in an electron glass [27]. Both types of per-

4 The relaxation time can be measured in structural glasses from the relaxation of the viscosity after a temperature quench or a stress, and in spin glasses from the response of the magnetization to a change in temperature or magnetic field.

turbation cause some electrons to tunnel to other sites, which in turn can create further hops, in a cascade process. We found that the avalanche size has a scale-free probability distribution $p(S) \approx S^{-\tau} \exp(-S / S_c)$, with a cutoff diverging with the linear size of the sample as $S_c \approx L / r_0$, where r_0 is the characteristic size of the soft dipoles (i.e., electron-hole pairs with low excitation energy). Furthermore, we showed that the avalanche is well described by a branching process in which each electron hop induces in average another hop. Recent experiments on 3D indium-oxide samples are consistent with the hypothesis that a change in gate voltage induces avalanche-like rearrangements in the whole sample [23].

Scale-free avalanches have been observed before in a wide variety of systems ranging from earthquakes to paper crumpling [31]. Recently, it was argued that, under certain conditions, they are a universal consequence of the existence of a Coulomb gap (or similar gaps in related systems) stemming from the long-range interaction [21].

Conclusions

Electron glasses show a remarkable non-equilibrium phenomenology resulting from the interplay of disorder and electron-electron interactions. Conductance and capacitance experiments allow to measure the responses of electron glasses to a variety of excitations (temperature, electric field, charge injection, electromagnetic radiation), thus offering an ideal testing ground to investigate the properties of glasses in general. The basic mechanism leading to slow relaxation in electron glasses is as yet unknown. Its elucidation will probably require theoretical efforts combining concepts from the statistical physics of disordered systems, quantum condensed-matter physics, and non-equilibrium statistical physics, as well as powerful computational algorithms. Quantum phenomena beyond tunneling, such as Anderson orthogonality catastrophe and many-body localization, not discussed here, may play an important role.

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Competing interests. None declared

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About the image on the first page of this article. This photograph was made by Prof. Douglas Zook (Boston University) for his book *Earth Gazes Back* [www.douglaszookphotography.com]. See the article "Reflections: The enduring symbiosis between art and science," by D. Zook, on pages 249-251 of this issue [http://revistes.iec.cat/index.php/CtS/article/view/142178/141126]. This thematic issue on "Non-equilibrium physics" can be unloaded in ISSUU format and the individual articles can be found in the Institute for Catalan Studies journals' repository [www.cat-science.cat; http://revistes.iec.cat/contributions].