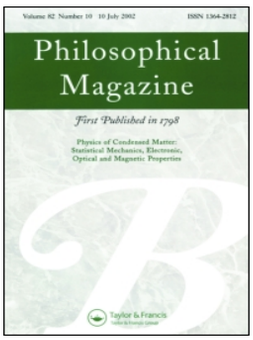

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Clare C. Yu

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Slow dynamics in glassy systems

CLARE C. YU†

Department of Physics and Astronomy, University of California, Irvine, California 92697, USA

ABSTRACT

We describe similarities in the features of various glassy systems where interactions and randomness compete. For example, Coulomb glasses have a gap in their single-particle density of states centred at the Fermi energy. This gap is analogous to the hole in the distribution of local fields of spin glasses and of ordinary glasses with dipolar interactions between two-level systems. When the field or energy where these holes are centred is suddenly shifted by the application of an external field, a new hole or gap develops roughly logarithmically in time. Such slow relaxation is characteristic of glassy systems. If we assume that this logarithmic behaviour applies for small perturbations, then thermal fluctuations will lead to fluctuations in the density of states and $1/f$ noise.

§1. INTRODUCTION

It is a great pleasure to contribute an article in honour of Professsor Michael Pollak's seventy-fifth birthday. Professor Pollak has made seminal contributions to our understanding of Coulomb glasses. He has drawn me into the field via a number of stimulating conversations over the years. I first met Mike on a visit to the University of California, Riverside, where he explained that a Coulomb glass is an insulator with randomly placed electrons that have Coulomb interactions. Lightly doped semiconductors and disordered metals are examples of such systems. I could see an analogy between Coulomb glasses and spin glasses and realized that Monte Carlo techniques combined with finite-size scaling could be used to revisit the question of whether a Coulomb glass has a second-order phase transition (Lee *et al.* 1982, 1984). Soon afterwards Grannan and I showed that there was indeed a second-order phase transition in three dimensions when the disorder arose solely from the random placement of the sites where electrons could sit (Grannan and Yu 1993). A few years later, Mike came to my office and described some non-equilibrium experiments that he, Ovadyahu and Vaknin were working on. He thought there could be a relation to experiments being done on ordinary window glasses at low temperatures by Osheroff's group for which my group had been providing some theoretical support (Carruzzo *et al.* 1994). Mike was right about this and this is what I would like to discuss here.

One of the points that I would like to make is that a variety of different systems in which interactions and randomness compete behave in similar ways because of the physics that they have in common. Examples of such systems are spin glasses, Coulomb glasses, ordinary window glasses at low temperatures, and

†Email: cyu@uci.edu

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vortex glasses in superconductors. Let me give a few words of introduction about the last two. At low temperatures the thermal and acoustic properties of insulating glasses at low temperatures tend to be universal, independent of the chemical make-up of the particular material (Phillips 1981, 1987, Hunklinger and Raychaudhuri 1986). This includes materials as diverse as amorphous SiO_2, polymers, sol–gels and varnishes. Their physical properties also differ qualitatively from crystalline solids. Below 1 K the specific heat is roughly linear in temperature, a property which contrasts strongly with the universal T^3 behaviour in insulating crystals. The thermal conductivity is quadratic in temperature (Zeller and Pohl 1971). Traditionally this low-temperature behaviour has been attributed to the existence of tunnelling two-level systems (Anderson *et al.* 1972, Phillips 1972). In this picture an atom or small group of atoms tunnels between the minima of a double-well potential. However, the true microscopic nature of the two-level systems remains a mystery. Two-level systems limit the thermal conductivity by scattering phonons. Since two-level systems couple to phonons, they can interact with each other by exchanging phonons. Another way to say this is to say that two-level systems interact with one another via the elastic strain field and this interaction goes as $1/r^3$. Some fraction of them have electric dipole moments and so these can also interact with each other via an electric dipolar interaction which also goes as $1/r^3$. It is the two-level systems with electric dipole moments which contribute to the dielectric constant, as we shall discuss later.

Vortex glasses refer to magnetic vortices in a superconductor which has randomly placed pinning sites. The repulsive interaction between the vortices is described by a Macdonald function which goes as $\ln(r/\lambda)$ for $r \ll \lambda$ where λ is the magnetic penetration depth (Lifshitz and Pitaevskii 1980). At large distances ($r \gg \lambda$), the interaction is screened and falls off exponentially. Without disorder, the vortices form a vortex lattice. In the presence of randomly placed pinning centres, the position of the vortices is somewhat random and the result is a vortex glass. There are a variety of vortex glasses, one of which is the so-called Bose glass which draws on the analogy between vortices pinned by columnar defects and the world lines of two-dimensional quantum-mechanical bosons (Nelson and Vinokur 1992, 1993).

These systems where interactions and randomness compete exhibit common features such as a Coulomb gap in the density of single-particle states or a hole in the local field distribution. This feature is independent of the particular form of the interaction and of the way that randomness is introduced, although the actual function describing the gap or hole will depend on the form of the interaction. Let us use a simple model of Ising spins to illustrate these general features. Assume that spins S_i and S_j on sites i and j can have values of $+1$ and -1 and that J_{ij} describes the interaction. There are a number of different ways in which we can introduce disorder into this model. If the spins reside on a lattice and J_{ij} is a random number, then we have an ordinary Ising spin glass. This corresponds to off-diagonal disorder. Alternatively the spins could be placed randomly with $J_{ij} = J(r_{ij})$ a well-defined function of the separation r_{ij} between sites i and j, for example $J_{ij} \sim r_{ij}^{-1}$ would describe a Coulomb interaction. Yet another way would be to put the spins on a lattice with $J_{ij} = J(r_{ij})$. In this case we introduce a random field ϕ_i on each site. This corresponds to diagonal disorder. So in general we can write

$$H = \tfrac{1}{2}\sum_{i\neq j} J_{ij}S_iS_j + \sum_i \phi_i S_i$$
$$= \sum_i h_i S_i, \tag{1}$$

where the effective field on site i is given by

$$h_i = \phi_i + \sum_{i\neq j} J_{ij}S_j. \tag{2}$$

We can map this system on to that of a Coulomb glass by identifying an up spin with a site occupied by an electron and a down spin with an unoccupied site. If the spins are on a lattice, ϕ_i would correspond to a random on-site energy. If the spins are placed randomly, then we have enough randomness and it is easiest to set ϕ_i to be zero everywhere. For a Coulomb glass we can identify the single-particle energy ε_i on site i with h_i in equation (1) with $J_{ij} = e^2/\kappa r_{ij}$, where e is the electron charge and κ is the dielectric constant. We can also use equation (1) as a simplified model of interacting two-level systems in glasses. In this case, $J_{ij} \sim r_{ij}^{-3}$ (Burin 1995). One can also use equation (1) as a model of a Bose glass (Taüber *et al.* 1995, Taüber and Nelson 1995) with $S_i = 1$ if site i is occupied by a vortex and $S_i = 0$ if there is no vortex on site i. For a vortex glass, ϕ_i corresponds to a random pinning potential and $J(r_{ij})$ is the Macdonald function. In all these systems, randomness and interactions produce a Coulomb gap-like feature.

§2. Coulomb gap

It was Pollak (1970) who first pointed out that in a Coulomb glass there must exist a Coulomb gap in the single-particle density of states in order for the ground state to be stable with respect to single-electron hopping. We can explain this gap using a spin-glass analogy. Suppose that we have a spin glass with a fixed total spin which corresponds to a fixed number of electrons in a Coulomb glass. In order for the ground state of the spin glass to be stable with respect to single spin flips, there must be a gap in the local field distribution $P(h)$ centred at $h = 0$ (Kirkpatrick and Varma 1978). For long-range interactions, the local field distribution goes to zero, that is $P(h = 0) = 0$ at $T = 0$. One way to understand this gap is as follows. Suppose that the ground-state spin configuration is found and suppose that the distribution of local magnetic fields $P(h)$ is finite at $h = 0$. This means that those spins with zero local field can flip without changing their energy. However, if they do so, other spins have their field altered and so some of them will flip. This in turn causes others to flip and so on. This avalanche means that the supposed ground state is not stable. In order to have a stable ground state, the distribution at zero field must go to zero as $h \to 0$. Such a gap has been found for a variety of interacting systems with randomness. For a Coulomb glass, the Coulomb gap is centred on the Fermi energy μ. Éfros and Shlovskiĭ (1975) and Shklovskiĭ and Éfros (1984) gave a more formal argument in which they showed that the gap vanishes as $(E - \mu)^2$ in three dimensions and as $|E - \mu|$ in two dimensions for a $1/r$ Coulomb interaction.

Such a gap also exists for dipoles such as those associated with two-level systems in ordinary glasses at low temperatures. This has been shown both theoretically (Carruzzo *et al.* 1994, Burin 1995) and experimentally (Salvino *et al.* 1994). For a $1/r^3$ interaction, an Ising spin glass on a lattice with a random field and fixed total

spin has a gap that is centred at $h = 0$ and goes as $P(h) \sim 1/\ln(1/h)$ (Baranovskiĭ *et al.* 1980).

A Coulomb gap centred at the chemical potential also exists for flux lines interacting in a superconductor in which there are random pinning sites. This has been shown using Monte Carlo simulations of a Bose glass model where the interaction between the vortices is the Macdonald function (Taüber *et al.* 1995, Taüber and Nelson 1995). Technically speaking the vortex–vortex interaction is screened and falls off exponentially for distances larger than the magnetic penetration depth, so the interaction is not truly long range. When the interaction is short range, $P(h)$ does not go all the way to zero at $h = 0$. This agrees with Monte Carlo simulations of the Bose glass (Taüber *et al.* 1995, Taüber and Nelson 1995). It also explains why there is a dip and not a gap at the Fermi energy in the single-particle density of states for a dirty metal on the metallic side of the metal–insulator transition where the Coulomb interactions are screened and fall off exponentially (Lee and Ramakrishnan 1985).

So far we have been concentrating on single-particle hopping or single spin flips. However, Pollak has been a strong proponent of the view that, in Coulomb glasses, it is the correlated motion of many particles that is important in the physics. No doubt this is correct but, to obtain a quantitative measure of the role played by many-particle hopping versus single-particle hopping, we should work out what effects we can expect from single-particle hopping and then try to compare the results with experiment. The discrepancies can probably be attributed to the effects of multiple-particle hopping. Such a comparison with experiment has been made for two-level systems in glasses (Carruzzo *et al.* 1994) and this showed that interactions between two-level systems must be taken into account. Comparison with experiment has not yet been made for Coulomb glasses, but the purpose here is to motivate our considerations of single-particle hopping.

§ 3. Non-equilibrium effects

We have established that interactions and randomness in spin glasses and dipolar glasses produce a hole centred at $h = 0$ in the local field distribution $P(h)$. This is analogous to a Coulomb gap centred at the chemical potential in a Coulomb glass. We now consider what happens if the centre of the hole is shifted suddenly. For example, if an external dc magnetic field h_{dc} is suddenly applied to a spin glass, then each spin finds itself in a new effective field $h_i = h_{i,old} + h_{dc}$. So effectively $h = 0$ is suddenly shifted. The system must dig a new hole in $P(h)$ centred at the new $h = 0$ and remove the old hole centred at $h_{old} = 0$. As we shall see, because the system is glassy, it makes this adjustment roughly logarithmically in time.

This non-equilibrium behaviour was first seen by Osheroff's group at Stanford in window glasses at low temperatures in which there are electric dipoles (Salvino *et al.* 1994). They put a thin film (1–3 μm) of glass between two capacitor plates which they cooled to low temperatures (20–1000 mK). Then they applied a large dc electric field (about 10^6–10^7 V m^{-1}). Using an ac capacitance bridge, they watched the capacitance jump up and then decay roughly logarithmically with time after the dc field had been applied. The dipoles which are easiest to polarize are those with a small local field, that is those in the vicinity of $h = 0$. It is these dipoles which contribute most to the dielectric constant and to the capacitance. So the jump corresponds to what happens to $P(h = 0)$ as the external field is applied (Carruzzo *et al.* 1994). When the dc field is applied, a new set of defects find themselves in zero local field. The applied dc field effectively shifts the local field distribution along the

local field axis. Immediately after the dc field is switched on, $P(0)$ will be large and finite. Thus the polarizability and hence capacitance suddenly increases. The size of the jump increases with the depth of the old hole out of which the system jumps. Since the hole is smeared by thermal effects, the size of the jump will increase as the temperature decreases. Once the dc field is switched on, a new hole develops and the capacitance decreases. After applying a fixed voltage for some time (about a day), Salvino *et al.* (1994) subsequently swept the dc bias field and found a hole in the capacitance at the previously applied bias voltage. This corresponds to sweeping h without allowing the system to equilibrate and, hence, the sweeps in field map out the hole in $P(h)$ versus h which is mirrored in the capacitance versus voltage V. Theoretical calculations including Monte Carlo simulations of the nearest-neighbour Ising spin glass (Carruzzo *et al.* 1994) and analytic calculations with $1/r^3$ interactions (Burin 1995) confirmed this interpretation and showed that the capacitance would relax roughly logarithmically in time.

Similar effects have been seen in Cu–Mn spin glasses by Fenimore and Weissman (1994). They have performed the magnetic analogue of the Stanford dielectric experiments and found similar results. For example, they found a hole in the imaginary part of the susceptibility χ'' versus magnetic field H with the minimum being at the field in which the sample was cooled. They also find that χ'' relaxes logarithmically in time when they change the applied field from $+\mathbf{H}$ to $-\mathbf{H}$.

§4. Forming the Coulomb gap

Pollak suggested that the physics underlying these experiments could help to explain the slow electron relaxation that he, Ovadyahu and Vaknin were seeing in their non-equilibrium experiments on disordered semiconducting indium oxide films (Ovadyahu and Pollak 1997, Vaknin *et al.* 1998). Similar results have been found in metallic films (Martinez-Arizala *et al.* 1998). The films were grown on insulating substrates which separated them from a gate electrode that regulated the electron density, and hence the chemical potential, of the film. The conductance G was measured as a function of the gate voltage V_G. If V_G had a particular value V_0, for a long time and then was varied over a range of voltages, there was a dip in the conductance centred at V_0. We identify this dip with the Coulomb gap in the density of states because the value of the conductance depends on the density of states at the Fermi energy (Vaknin *et al.* 1998, Yu 1999). (A similar scenario was qualitatively discussed by Martinez-Arizala *et al.* (1998).) In Mott's picture of variable-range hopping, the hopping conductivity increases when the density of states at the Fermi energy increases, since there are then more states to which an electron at the Fermi energy can hop (Shklovskiĭ and Éfros 1984). We identify sweeping V_G with varying the chemical potential without allowing time for equilibration. In effect the sweeps scan the density of states. Thus we expect the conductance to increase with increasing density of states and hence as the gate voltage V_G moves away from V_0.

In the experiments, if the gate voltage was changed suddenly from, say, V_0 to V_1, the conductance had a very fast initial rise, followed by a period of rapid relaxation, which in turn was followed by a long period of very slow relaxation. In some cases the relaxation was logarithmic in time. Our interpretation of this is that, when the gate voltage is changed, the Fermi energy changes, and time-dependent relaxations arise because the system must dig a new hole in the density of states at the new Fermi energy and remove the old hole at the old Fermi energy. Indeed, Vaknin *et al.* (1998)

found that subsequent sweeps of the gate voltage revealed that the old dip in the conductance at V_0 fades with time while a new dip centred at V_1 increased with time. The dip in the conductance and the long time relaxation were present only at very low temperatures and not at higher temperatures ($T \gtrsim 20\,\mathrm{K}$).

Our explanation of these experiments is confirmed by the following calculations (Yu 1999). Our model of the Coulomb glass follows that of Baranovskiĭ *et al.* (1980). In this model, the electrons occupy the sites of a periodic lattice, and the number of electrons is half the number of sites. Each site has a random on-site energy ϕ_i chosen from a uniform distribution extending from $-A$ to A. Thus, g_0 (the density of states without interactions) is flat. A site can contain 0 or 1 electron. In order to follow the time development of the Coulomb gap, we assume that the Coulomb interactions are turned on at time $t = 0$. The Hamiltonian can be written as

$$H = \sum_i \phi_i n_i + \sum_{i>j} \frac{e^2}{\kappa r_{ij}} n_i n_j \theta(t), \tag{3}$$

where the occupation number n_i equals $\frac{1}{2}$ if site i is occupied and $-\frac{1}{2}$ if site i is unoccupied, e is the electron charge, κ is the dielectric constant and the step function $\theta(t)$ is 0 for $t < 0$ and 1 for $t \geqslant 0$.

The Coulomb gap arises because the stability of the ground state with respect to single-electron hopping from an occupied site i to an unoccupied site j requires (Shklovskiĭ and Éfros 1984)

$$\Delta_i^j = \varepsilon_j - \varepsilon_i - \frac{e^2}{\kappa r_{ij}} > 0, \tag{4}$$

where the single-site energy $\varepsilon_i = \phi_i + \sum_j (e^2/\kappa r_{ij}) n_j$. Δ_i^j is the change in energy that results from hopping from i to j. So we need to subtract from the density of states those states which violate this stability condition. This leads to (Baranovskiĭ *et al.* 1980, Burin 1995)

$$g(\varepsilon, t) = g_0 \prod_{j>i} \left[1 - a_0^3 \int_{-A}^{A} \mathrm{d}\varepsilon' g(\varepsilon', t) \theta\left(\frac{e^2}{\kappa r_{ij}} + \varepsilon - \varepsilon' \right) F(n_i' = 1, n_j' = 0) \theta(t - \tau_{ij}(\varepsilon', \varepsilon, r_{ij})) \right], \tag{5}$$

where the single-site energy $\varepsilon_i = \varepsilon$, $\varepsilon_j = \varepsilon'$ and a_0 is the lattice constant. $n_i' = n_i + \frac{1}{2}$; so $n_i' = 1$ if site i is occupied and $n_i' = 0$ if site i is unoccupied. $F(n_i', n_j')$ is the probability that donors i and j have occupation numbers n_i' and n_j' respectively, while all other sites have their ground-state occupation numbers $\tilde{n}_k'$. τ_{ij}^{-1} is the number of electrons which jump from site i to site j per unit time. $\theta(t - \tau_{ij})$ represents the fact that at time t, the primary contributions to the change in the density of states will be from those hops for which $\tau_{ij} < t$. (The exact form of the cut-off is not important; for example, replacing $\theta(t - \tau_{ij})$ with $[1 - \exp(-t/\tau_{ij})]$ affects our results only negligibly.) In writing equation (5), we assume that these hops together with phonons have equilibrated the system as much as is possible at time t. The hopping rate τ_{ij}^{-1} is given by (Shklovskiĭ and Éfros 1984)

$$\tau_{ij}^{-1} = \gamma_{ij}^{0} \exp\left(-\frac{2r_{ij}}{a}\right)[1 + N(\Delta_i^j)]F(n_i' = 1, n_j = 0). \tag{6}$$

where $a = \kappa a_B$ is the effective Bohr radius of a donor and a_B is the usual Bohr radius ($a_B = \hbar^2/me^2$). We shall set the mass m equal to the electron mass so that $a_B = 0.529$ Å. $N(\Delta_i^j)$ is the phonon occupation factor and reflects the contribution of phonon-assisted hopping. We are also allowing for spontaneous emission of phonons since we are considering a non-equilibrium situation in which electrons hop in order to lower their energy. The coefficient γ_{ij}^0 is given by (Shklovskiĭ and Éfros 1984)

$$\gamma_{ij}^{0} = \frac{E_1^2|\Delta_i^j|}{\pi d s^5 \hbar^4}\left(\frac{2e^2}{3\kappa a}\right)^2 \frac{r_{ij}^2}{a^2}\left[1 + \left(\frac{\Delta_i^j a}{2\hbar s}\right)^2\right]^{-4}, \tag{7}$$

where E_1 is the deformation potential, s is the speed of sound and d is the mass density. Following Baranovskiĭ *et al.*, we can derive a self-consistent equation for the density of states, $g(\varepsilon, t)$:

$$g(\varepsilon, t) = g_0 \exp\left(-\tfrac{1}{2}\int_{-A}^{A} d\varepsilon' g(\varepsilon', t) \int_{a_0}^{\infty} dr 4\pi r^2 F(n(\varepsilon) = 1, n(\varepsilon') = 0)\right.$$
$$\left. \times \theta\left(\frac{e^2}{\kappa r} + \varepsilon - \varepsilon'\right)\theta(t - \tau(\varepsilon', \varepsilon, r))\right). \tag{8}$$

At low energies, large distances play an important role and so we have replaced the sum by an integral over r in the exponent. The origin is at site i. $n(\varepsilon)$ is the occupation probability of a site with energy ε. $\tau(\varepsilon', \varepsilon, r)$ is given by equation (6) with r_{ij} replaced by r, ε_i replaced by ε, and ε_j replaced by ε'.

Since it is not clear how the stability condition of equation (4) can be applied to finite temperatures, we shall confine our calculations to the case of $T = 0$. In this case the phonon occupation factor $N(\Delta_i^j) = 0$ and the electron occupation factor $F(n_i = 1, n_j = 0) = 1$ if $\varepsilon_i < 0$ and $\varepsilon_j > 0$. Otherwise $F(n_i = 1, n_j = 0) = 0$. We set the Fermi energy $\mu = 0$. We can solve equation (8) iteratively on the computer. For the first iteration we start with $g(\varepsilon', t) = g_0$, and calculate $g(\varepsilon, t)$. This is then used as the input for $g(\varepsilon', t)$ in the next iteration. After a few iterations the typical difference between successive iterations is typically less than 1 part in 10^5. Because there is particle–hole symmetry, we only need to calculate $g(\varepsilon, t)$ for $\varepsilon < 0$. Figure 1 (*a*) shows the density of states, $g(\varepsilon, t)$, as a function of energy at different times, while figure 1 (*b*) shows $g(\varepsilon, t)$ as a function of time at different energies. Note the development of the Coulomb gap occurs over many decades in time. The functional form of the time dependence of $g(\varepsilon, t)$ varies with the energy ε and with g_0. For example, at the Fermi energy $g(\mu, t) \sim A - B \ln t$ for $g_0 = 2 \times 10^5$ states $K^{-1} Å^{-3}$ (A and B are constants) and $g(\mu, t) \sim t^{-0.05}$ for $g_0 = 6.25 \times 10^5$ states $K^{-1} Å^{-3}$. After an infinite amount of time, the density of states at the Fermi energy μ goes to zero and $g(\varepsilon) \sim \varepsilon^2$. For finite times, $g(\varepsilon) \sim |\varepsilon|$ in the vicinity of the Fermi energy, though there will be thermal smearing at finite temperatures.

The temporal development of the Coulomb gap is qualitatively consistent with the experimental observation of the long-time relaxation of the conductance after the gate voltage V_G has been changed. The exact relation between the conductance and the density of states is difficult to ascertain in this case because the system is not in equilibrium. However, it is reasonable to assume that the conductance reflects the

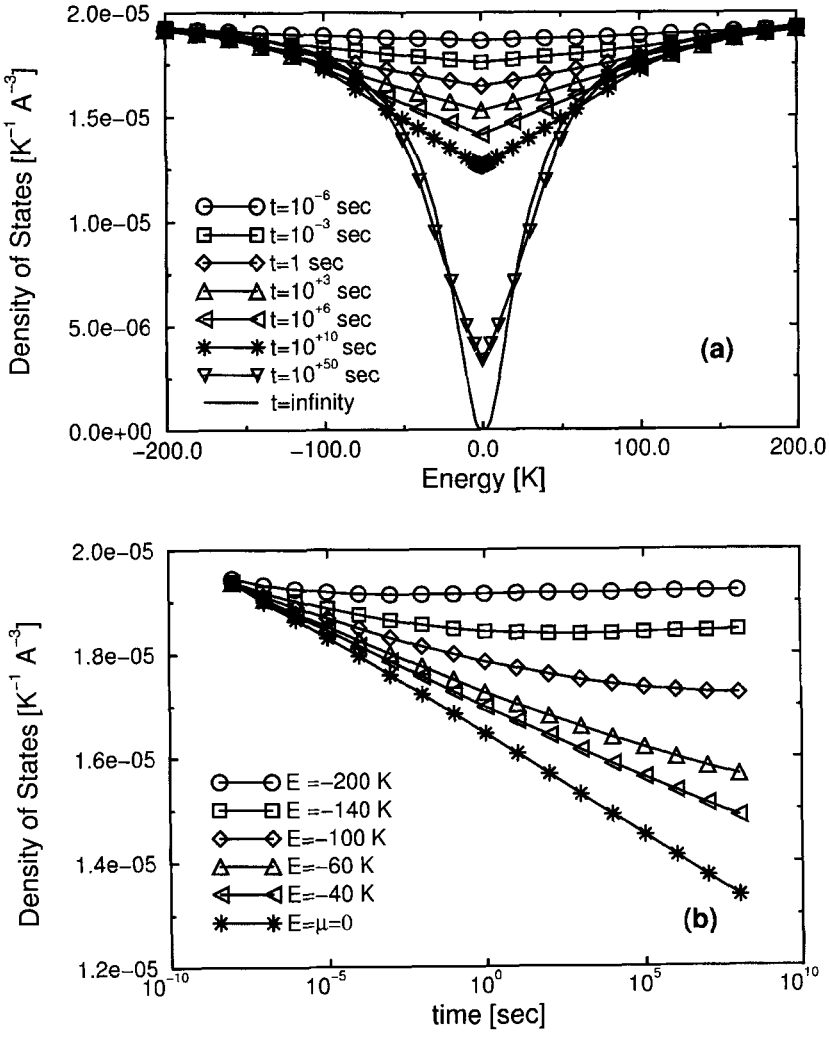

Figure 1. (*a*) Density of states, $g(\varepsilon)$, as a function of energy for different times. (*b*) Density of states as a function of time for various energies. The parameters used are $g_0 = 2 \times 10^5$ states $K^{-1}\,Å^{-3}$, $T = 0$, $A = 10^4\,K$, $\kappa = 10$, $d = 7.18\,g\,cm^{-3}$, $s = 5.0 \times 10^5\,cm\,s^{-1}$, $E_1 = 5 \times 10^3\,K$, and $a_0 = 4\,Å$. The density d is chosen to be that of In_2O_3. The energy is measured from the Fermi energy $\mu = 0$.

density of states at the Fermi energy. A well-known example is Mott's formula for conductivity σ due to variable-range hopping (Shklovskiĭ and Éfros 1984):

$$\sigma = \sigma_0 \exp\left[-\left(\frac{T_0}{T}\right)^{1/4}\right] \tag{9}$$

where $T_0 = c_0/k_B g(\mu) a^3$, c_0 is a numerical constant and $g(\mu)$ is the density of states at the Fermi energy. While strictly speaking this equilibrium formula does not apply to our non-equilibrium situation, we see qualitatively that an increase (decrease) in $g(\mu)$ leads to an increase (decrease) in the conductivity. In the experiments, rapidly sweeping the gate voltage V_G varies the chemical potential without allowing time for

equilibration. Relating the conductance to the density of states means that the sweeps over V_G scan the density of states. To obtain a qualitative feel for this connection, we shall use equation (9). Let us assume that our zero-temperature density of states continues to be valid at low temperatures. Then we identify $g(\mu)$ with $g(\varepsilon)$ and substitute the $g(\varepsilon)$ shown in figure 1 into $T_0 = c_0/(k_B g(\varepsilon)a^3)$. Here we treat ε as the instantaneous chemical potential μ. For most of the scan the density of states has the linear form $g(\varepsilon) = g(\varepsilon_0) + \alpha(\varepsilon - \varepsilon_0)$, where ε_0 and the slope α are constants. The exponent of 1/4 is appropriate for this case. The experiments on indium oxide (Ovadyahu and Pollak 1997) were carried out at 4.11 K; so we set $T = 4$ K. The result is shown in figure 2.

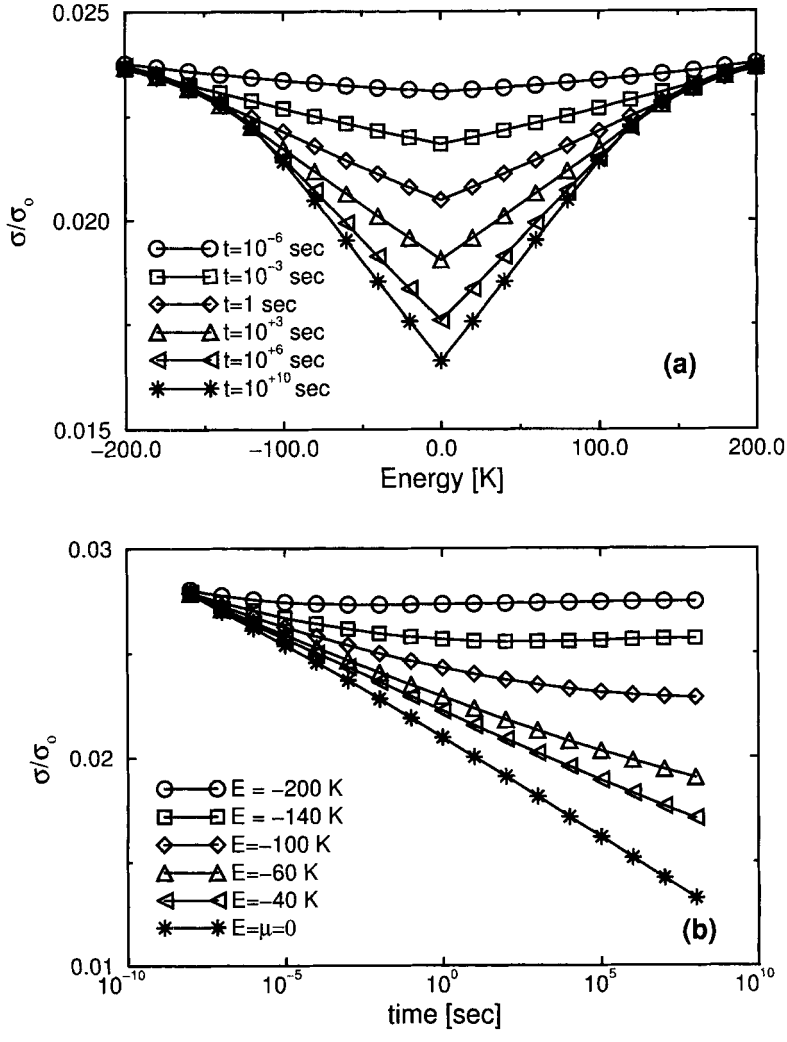

Figure 2. (*a*) Dimensionless conductivity σ/σ_0 as a function of energy for different times. (*b*) Dimensionless conductivity σ/σ_0 as a function of time for various energies. The conductivities for both (*a*) and (*b*) are calculated using Mott's formula (9) with $T = 4$ K, $a = 5.291\,77$ Å and $c_0 = 2.23$. The rest of the parameters are the same as in figure 1.

The experiments found that the dip in the conductance as a function of gate voltage V_G, and the long relaxation times of the conductance following a change in V_G were present only at low temperatures. These features were not observed at $T \gtrsim 20\,\mathrm{K}$. From equations (5) and (6), we see that an increase in temperature will affect $g(\varepsilon, t)$ in two ways. First the thermal smearing of the occupation factor $F(n_i', n_j')$ will fill in the Coulomb gap to some extent (Levin *et al.* 1987, Mogilyanskiĭ and Raĭkh 1989, Hunt 1990, Grannan and Yu 1993, Vojta *et al.* 1993, Li and Phillips 1994, Sarvestani *et al.* 1995). Secondly, as the number of phonons increases with increasing temperature, there is an increase in the phonon assisted hopping of electrons. We expect that this leads to a rapid rearrangement of electrons on time scales that are too short to observe experimentally. As a result, no dips in the conductance and no long time relaxation were seen experimentally at higher temperatures. It is difficult to calculate these effects because it is not clear how to generalize the stability condition (4) to finite temperatures, although Mogilyanskiĭ and Raĭkh (1989) have suggested one possible way. In addition the system is not in equilibrium and hence temperature is not well defined for the electrons. However, the absence of the conductance dips at higher temperatures is consistent with this scenario.

To summarize our calculations in this section, we have shown that the time development of the Coulomb gap in a Coulomb glass can involve very long time scales owing to electron hopping and rearrangement. These results are consistent with conductance experiments on disordered semiconducting and metallic films. Although we have only considered single-electron hops, these hops are dependent upon previous hops of other electrons through their cumulative effect on the single-particle density of states. We expect multiple-electron processes also to contribute to the conductance, particularly at long time scales.

§ 5. $1/f$ NOISE

In the last section we saw that a sudden shift in the centre of the Coulomb gap or of the hole in the local field distribution leads to a new hole forming roughly logarithmically in time. In this section we argue that such slow relaxation times lead to $1/f$ noise. Low-frequency $1/f$ noise (Dutta and Horn 1981, Weissman 1988, Kogan 1996) is ubiquitous; it is found in a wide variety of conducting systems such as metals, semiconductors, tunnel junctions (Rogers and Buhrman 1984) and even superconducting quantum interference devices (Koch 1983, Koelle *et al.* 1999). Yet the microscopic mechanisms are still not well understood. For the moment let us focus on $1/f$ noise in Coulomb glasses. Experimental studies on doped silicon inversion layers have shown that low-frequency $1/f$ noise is produced by hopping conduction (Voss 1978). More recent experiments have observed $1/f$ noise down to 0.1 Hz in boron-doped silicon (Massey and Lee 1997) and in doped germanium (Shlimak *et al.* 1995). Because the systems are glassy, electron hopping can occur on very long time scales and this produces low-frequency noise. In this section we show that the resulting noise spectrum goes as $f^{-\alpha}$ where f is the frequency and the exponent $\alpha \approx 1$.

Shklovskiĭ developed the first theory of $1/f$ noise in Coulomb glasses. He suggested that it is produced by fluctuations in the number of electrons in an infinite percolating cluster (Shklovskiĭ 1980). These fluctuations are caused by the slow exchange of electrons between the infinite conducting cluster and small isolated donor clusters. A more rigorous calculation combined with numerical simulations

(Kogan and Shklovskiĭ 1981) of Shklovskiĭ's model found a noise spectrum that went as $f^{-\alpha}$ where α was considerably lower than 1. Furthermore, below a minimum frequency of the order of 1–100 Hz, the noise spectral density saturated and became a constant independent of frequency. A similar conclusion holds for a model suggested by Kozub (1996) in which electron hops within finite clusters produce fluctuations in the potential seen by hopping conduction electrons that contribute to the current. Hunt (1998) used a similar model and included the effect of the size of the finite clusters on the polarization currents in these clusters. By taking into account the effect of these currents on the conduction through the percolating network, he again found $1/f$ noise. Kogan (1998) has argued that transitions between valleys in the energy landscape produces $1/f$ noise because high barriers result in slow fluctuations in hopping conduction.

We take a different approach in which we focus on fluctuations in the single-particle density of states rather than on the percolating network. Electron hopping shifts the single-particle energies ε because they depend on Coulomb interactions with other sites. This leads to fluctuations in the single particle density of states, $g(\varepsilon)$, which, in turn, produces fluctuations in the conductivity. The conductivity depends on the density of states, $g(\varepsilon \approx \mu)$, in the vicinity of the Fermi energy μ. Note that $g(\varepsilon \approx \mu)$ can be affected by hops between sites i and j even if the energies on these sites are not near the Fermi energy because an electron or hole on site i or j can interact with other sites whose energy is (or was) near the Fermi energy.

We shall use Mott's argument for variable-range hopping (Mott 1968, Ambegaokar *et al.* 1971, Shklovskiĭ and Éfros 1984) to relate fluctuations in the density of states to fluctuations in the resistivity. One can regard a Coulomb glass as a random resistor network (Miller and Abrahams 1960) with a transition between sites i and j associated with a resistance R_{ij} given by

$$R_{ij} = R_{ij}^0 \exp(\xi_{ij}), \tag{10}$$

where the prefactor $R_{ij}^0 = kT/e^2\gamma_{ij}^0$ with γ_{ij}^0 given by equation (7). In equation (10), the exponent is given by

$$\xi_{ij} = \frac{2r_{ij}}{a} + \frac{\varepsilon_{ij}}{kT}. \tag{11}$$

The exponent reflects the thermally activated hopping rate between i and j as well as the wavefunction overlap between the sites. ε_{ij} is given by (Shklovskiĭ and Éfros 1984)

$$\varepsilon_{ij} = \begin{cases} |\varepsilon_j - \varepsilon_i| - \dfrac{e^2}{\kappa r_{ij}}, & (\varepsilon_i - \mu)(\varepsilon_j - \mu) < 0, \\ \max[|\varepsilon_i - \mu|, |\varepsilon_j - \mu|], & (\varepsilon_i - \mu)(\varepsilon_j - \mu) > 0. \end{cases} \tag{12}$$

At both high and low compensations, electron hopping usually occurs on one side of the Fermi level μ and the lower expression applies. At intermediate compensations and in the regime of variable-range hopping, hopping electrons often cross the Fermi level and the upper expression applies.

In the regime of variable-range hopping, Mott pointed out that hopping conduction at low temperatures comes from states near the Fermi energy. Let $\tilde{\varepsilon} = \varepsilon - \mu$. If we consider states within ε_0 of the Fermi energy, then the concentration of states in this band is $N(\varepsilon_0) = \int_{-\varepsilon_0}^{\varepsilon_0} g(\tilde{\varepsilon})\,\mathrm{d}\tilde{\varepsilon}$, where $g(\tilde{\varepsilon})$ is the density of states with energy $\tilde{\varepsilon}$

measured from the Fermi energy. So the typical separation between sites is $R = [N(\varepsilon_0)]^{-1/3}$. To estimate the resistance corresponding to hopping between two typical states of the band, we replace r_{ij} with R and $|\varepsilon_j - \varepsilon_i|$ with ε_0 in equations (11) and (12) to obtain $\xi(\varepsilon_0)$. Minimizing $\xi(\varepsilon_0)$ yields $\bar{\varepsilon}_0$. Substituting this into equations (11) and (10) yields the variable-range hopping formula for the resistivity: $\bar{\rho}(T) = \rho_0(T) \exp[\xi(\bar{\varepsilon}_0)]$.

In our model the noise results from electron hopping which produces fluctuations in the density of states $g(\varepsilon) = \bar{g}(\varepsilon) + \delta g(\varepsilon)$, where $\bar{g}(\varepsilon)$ is the average density of states. This in turn creates fluctuations in $N(\varepsilon_0)$, $\xi(\varepsilon_0)$, $\bar{\varepsilon}_0$ and $\rho(T)$. We can calculate these fluctuations by applying perturbation theory (Marion 1970) to the derivation of the variable-range hopping formula. We begin by letting $\xi(\varepsilon_0) = \xi_0(\varepsilon_0) + \lambda\,\delta\xi(\varepsilon_0)$. λ is just used to keep track of the various orders of perturbation theory. As before, we want to find ε_0 such that $\mathrm{d}\xi(\varepsilon_0)/\mathrm{d}\varepsilon_0 = 0$. We use a trial solution $\varepsilon_0 = \overline{\varepsilon_0} + \lambda\,\delta\varepsilon_0$, and expand $\mathrm{d}\xi(\varepsilon_0)/\mathrm{d}\varepsilon_0$ in powers of λ. Solving $\mathrm{d}\xi(\varepsilon_0)/\mathrm{d}\varepsilon_0 = 0$ to first order in λ leads to $\delta\xi(\varepsilon_0) = \delta\rho(T)/\bar{\rho}(T) = -[2kTg(T,\bar{\varepsilon}_0)]^{-1} \int_{-\bar{\varepsilon}_0}^{\bar{\varepsilon}_0} \delta g(T,\tilde{\varepsilon})\,\mathrm{d}\tilde{\varepsilon}$. We have included the temperature dependence of the density of states because at finite temperatures the Coulomb gap fills in and the density of states no longer vanishes at the Fermi energy (Levin *et al.* 1987, Mogilyanskiĭ and Raĭkh 1989, Hunt 1990, Grannan and Yu 1993, Vojta *et al.* 1993, Li and Phillips 1994, Sarvestani *et al.* 1995). The autocorrelation function for the fluctuations in the resistivity is

$$\frac{\langle \delta\rho(T,t_2)\,\delta\rho(T,t_1)\rangle}{\bar{\rho}^2(T)} = \frac{1}{4k^2T^2g^2(T,\bar{\varepsilon}_0)} \int_{-\bar{\varepsilon}_0}^{\bar{\varepsilon}_0} \mathrm{d}\tilde{\varepsilon} \int_{-\bar{\varepsilon}_0}^{\bar{\varepsilon}_0} \mathrm{d}\tilde{\varepsilon}' \langle \delta g(T,\tilde{\varepsilon},t_2)\,\delta g(T,\tilde{\varepsilon}',t_1)\rangle. \tag{13}$$

We assume that there is no correlation between the fluctuations in the density of states at different energies; so

$$\langle \delta g(T,\tilde{\varepsilon},t_2)\,\delta g(T,\tilde{\varepsilon}',t_1)\rangle = E\langle \delta g(T,\tilde{\varepsilon},t_2)\,\delta g(T,\tilde{\varepsilon},t_1)\rangle \delta(\tilde{\varepsilon} - \tilde{\varepsilon}'), \tag{14}$$

where E is an energy of the order of $2\bar{\varepsilon}_0$. Furthermore we assume that the time and energy dependences of the density-of-states autocorrelation function are separable, allowing us to write

$$\int_{-\bar{\varepsilon}_0}^{\bar{\varepsilon}_0} \mathrm{d}\tilde{\varepsilon} \langle \delta g(T,\tilde{\varepsilon},t_2)\,\delta g(T,\tilde{\varepsilon},t_1)\rangle = C(\bar{\varepsilon}_0,T) f(T,t_2 - t_1), \tag{15}$$

where we are assuming translational invariance in time (stationary processes). $C(\bar{\varepsilon}_0,T)$ is a function of $\bar{\varepsilon}_0$ and temperature. The function $f(T,t)$ characterizes the time dependence of the return to equilibrium by the system after it is perturbed by a fluctuation in the density of states. Inserting equations (14) and (15) into equation (13) yields

$$\frac{\langle \delta\rho(T,t_2)\,\delta\rho(T,t_1)\rangle}{\bar{\rho}^2(T)} = \frac{EC(\bar{\varepsilon}_0,T)}{4k^2T^2g^2(T,\bar{\varepsilon}_0)} f(T,t_2 - t_1). \tag{16}$$

To relate this to the spectral density $S(\omega)$ of the noise, let $\psi_\rho(t_2 - t_1) = \langle \delta\rho(T,t_2)\,\delta\rho(T,t_1)\rangle$ and let $\psi_\rho(\omega)$ be the Fourier transform of $\psi_\rho(t_2 - t_1)$. According to the Wiener–Khintchine theorem (Kogan 1996), for a stationary process the spectral density of fluctuations is given by

$$S_\rho(\omega) = 2\psi_\rho(\omega)$$
$$= \frac{E\bar{\rho}^2(T)C(\bar{\varepsilon}_0, T)}{2k^2T^2g^2(T, \bar{\varepsilon}_0)} f(T, \omega). \tag{17}$$

This equation for $S_\rho(\omega)$ is valid for both cases in equation (12).

We do not know the temperature dependence of $f(T, t)$; so for the moment we shall suppress this and just refer to $f(t)$. As we showed in the last section, after large deviations from equilibrium, the density of states returns to equilibrium with a time dependence given by $g(\mu, t) \sim -\ln t$ or $g(\mu, t) \sim t^{-\theta}$ where $\theta \ll 1$ (Yu 1999). Let us assume that these functional forms also hold for $f(t)$ in the linear response regime at low temperatures. If a fluctuation $\delta g(\mu, t = 0)$ at $t = 0$ pushes the density of states away from its mean equilibrium value at the Fermi energy, then this perturbation will decay according to $f(t)$ which enters into equations (15) and (16). Our non-equilibrium calculation indicates that $f(t)$ can have the form

$$f_1(t) = B_1 \ln\left(\frac{t_0}{t}\right), \tag{18}$$

where $t < t_0$, and t_0 is of the order of the age of the Universe or longer, or

$$f_2(t) = B_2 t^{-\theta}, \tag{19}$$

where $\theta \ll 1$, and B_1 and B_2 are positive constants. In both cases, t is greater than some $t_{\min}$ of the order of 10^{-8} s, say. The time dependence is a function of the energy, so here we set $\varepsilon = \mu$. Fourier transforming $f_1(t)$ and keeping the real part, we find that

$$f_1(\omega) \approx \frac{\pi B_1}{2} \frac{1}{\omega}. \tag{20}$$

This implies that the noise spectral density $S(\omega) \sim 1/\omega$. Fourier transforming $f_2(t)$ and keeping the real part yields

$$f_2(\omega) \approx \frac{\pi B_2 \theta}{2} \frac{1}{\omega^{1-\theta}} \tag{21}$$

for $\theta \ll 1$. This implies that $S(\omega) \sim 1/\omega^{1-\theta}$.

Let us summarize our argument for $1/f$ noise in Coulomb glasses. Electron hopping leads to fluctuations in the density of states that relax back to equilibrium roughly logarithmically in time. This leads to $1/f$ noise in the spectral density $S(\omega)$ of the noise in the resistivity. In particular we find that $S(\omega) \sim 1/\omega^\alpha$ where $\alpha = 1$ if the relaxation is logarithmic in time, and $\alpha = 1 - \theta$ if the relaxation is a power law that goes as $t^{-\theta}$ where $\theta \ll 1$. In general, α depends on temperature (Massey and Lee 1997) and is weakly dependent on the non-interacting density of states, g_0, and on the time scales. As equation (17) indicates, the noise amplitude also depends on the temperature. Unfortunately we cannot ascertain these temperature dependences because we do not know the temperature dependence of the fluctuations $\delta g(T, \tilde{\varepsilon}, t)$ in the density of states. However, we believe that our mechanism for $1/f$ noise should be valid at low temperatures ($T \lesssim 20$ K) where the logarithmic time dependence of the conductance is observed after the Coulomb glass has been pushed out of equilibrium by the sudden application of a gate voltage (Ovadyahu and Pollak 1997, Martinez-Arizala *et al.* 1998, Vaknin *et al.* 1998).

So far we have been discussing $1/f$ noise in Coulomb glasses but we believe that these arguments can be generalized to a variety of systems where interactions and randomness compete. As we discussed earlier, ordinary glasses and spin glasses recover roughly logarithmically in time when pushed out of equilibrium. If we assume that this logarithmic behaviour also applies in the linear response regime where there are small perturbations, then it is reasonable to expect that the autocorrelation function $\psi_P(t)$ for fluctuations in the local field distribution will have a logarithmic time dependence:

$$\psi_P(t) = \langle \delta P(h=0,t)\, \delta P(h=0,t=0) \rangle \sim -\ln\left(\frac{t}{t_0}\right). \tag{22}$$

(The subscript P in $\psi_P(t)$ refers to the distribution $P(h)$.) These fluctuations can be thermal fluctuations of the spins in a spin glass or the dipoles in a dielectric glass or the electron occupation of the sites in a Coulomb glass. Fourier transforming $\psi_P(t)$ yields a noise spectral density that goes as $1/f$:

$$S_P(\omega) = 2\psi_P(\omega) \sim \frac{1}{\omega}, \tag{23}$$

Thus we expect a variety of glassy systems to exhibit $1/f$ noise in a measurable quantity.

To summarize, systems which are glassy owing to the competition between disorder and interactions exhibit some very similar features. Coulomb glasses have a gap in their single-particle density of states which is analogous to the hole in the distribution of local fields of spin glasses and ordinary glasses with dipolar interactions between two level systems. When the field or energy where these holes are centred is suddenly shifted by the application of an external field, a new hole or gap develops roughly logarithmically in time. Such slow relaxation is characteristic of glassy systems. If we assume that this logarithmic behaviour applies for small perturbations, then thermal fluctuations will lead to fluctuations in the density of states and $1/f$ noise in a measurable quantity.

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