Effective Temperature in Relaxation of Coulomb Glasses

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We study relaxation in two-dimensional Coulomb glasses up to macroscopic times. We use a kinetic Monte Carlo algorithm especially designed to escape efficiently from deep valleys around metastable states. We find that, during the relaxation process, the site occupancy follows a Fermi-Dirac distribution with an effective temperature much higher than the real temperature T. Long electron-hole excitations are characterized by T_{eff}, while short ones are thermalized at T. We argue that the density of states at the Fermi level is proportional to T_{eff} and is a good thermometer to measure it. T_{eff} decreases extremely slowly, roughly as the inverse of the logarithm of time, and it should affect hopping conductance in many experimental circumstances.

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Coulomb glasses are Anderson-localized electronic systems with Coulomb electron-electron interactions. Very slow relaxation rates are commonly observed due to the exponential dependence of the transition rates on hopping length and energy and due to the many-valley structure of the phase space. Recent experiments on indium oxides by Ovadyahu's group [1] have shown glassy behavior such as logarithmic relaxation and aging. Similar effects have also been found on granular metals [2], on ultrathin metal films [3], and on a rare earth hydride [4]. All of these systems can be quite well described by the Coulomb glass model, which keeps gaining importance in the study of glassy relaxation.

Slow relaxation in Coulomb glasses has been studied theoretically [5–8] and by Monte Carlo (MC) simulations [9]. Simulations were able only to reach 10^7 MC steps, roughly corresponding to a microsecond, far from times >1 s where the logarithmic relaxation is observed [1–3].

We developed a method [10] which, in contrast to current methods, eliminates irrelevant repetitious fast processes at long times, thus reducing the computation time by integrating such steps without affecting adversely the original dynamics. The method works extremely well at very low temperatures and can be applied to many different systems. We have implemented an improved version of Ref. [10] to study relaxation in Coulomb glasses up to macroscopic times.

Recently, the concept of an effective temperature T_{eff} in glasses out of equilibrium was put forward in the spin glass literature [11]. From the exact solution of a simple spherical model, the fluctuation-dissipation theorem was generalized for systems out of equilibrium, which lead to a definition of T_{eff} for slow modes. While fast modes are equilibrated at the real temperature T, slow modes are characterized by a higher T_{eff}. This proposal has been tested numerically in different systems.

We show here convincingly that an effective temperature T_{eff} in relaxation of Coulomb glasses exists. We find that, out of equilibrium, the site occupation follows a Fermi-Dirac (FD) distribution with a T_{eff} much larger than T. We analyze the role of T_{eff} in single-electron excitations and how to measure it through the density of states. We study the dependence of T_{eff} on time and system size.

We consider two-dimensional (2D) systems in the strongly localized regime with long-range Coulomb interactions, described by the Coulomb gap (CG) Hamiltonian [10]

$$H = \sum_i \phi_i n_i + \sum_{i<j} (n_i - K)(n_j - K)\frac{1}{r_{ij}}, \tag{1}$$

where $n_i = 0, 1$ are occupation numbers, K is the compensation, equal to $1/2$, and ϕ_i are the random site energies chosen from a box distribution with interval $[-W/2, W/2]$. r_{ij} is the distance between sites i and j. We consider square samples of lateral size L with periodic boundary conditions, and N sites placed at random with a minimum separation between them of $0.05 l_0$. $l_0 = L/\sqrt{N}$ is our unit of distance, and $1/l_0$ is our unit of energy and temperature. We study systems ranging from 500 to 2000 sites with a range of disorder $W = 2$.

It should be pointed out that the MC method employed here assumes that the relaxation proceeds by single-electron transitions, while in some previous works [12] we showed that many-electron transitions are crucially important at very low T. But there we dealt with conduction in the linear response regime, while here we deal with relaxation from highly excited systems where many-electron transitions are much less important. Thus we believe that the present work is reliable except perhaps after long relaxation times, where inaccuracies can occur due to the neglect of many-electron transitions. This question will be dealt with in future work. The transition rate from configuration α to β is then

$$\Gamma_{\beta,\alpha} = \tau_0^{-1} \exp\left(\frac{-2r_{ij}}{\xi}\right) \min\left[1, \exp\left(-\frac{E_{\beta,\alpha}}{kT}\right)\right] \quad (2)$$

if α and β are connected by a single electron jump from site i to site j and zero otherwise. τ_0 is the inverse phonon frequency, of the order of 10^{-13} s, and we take it as our unit of time. $E_{\beta,\alpha}$ is the energy difference between configurations α and β. ξ is the localization length, which we choose equal to unity.

At low T, a standard MC algorithm that avoids rejections consists in forcing at every step to change from the present configuration α to a new one β with probability $P_{\beta,\alpha} = \Gamma_{\beta,\alpha}/\Sigma_\alpha$, where $\Sigma_\alpha = \sum_\beta \Gamma_{\beta,\alpha}$ with $\Gamma_{\alpha,\alpha} = 0$ [13]. The physical time associated with this new step is $\tau_\alpha = 1/\Sigma_\alpha$ [13,14]. However, the program still needs to perform many intravalley transitions before escaping to another valley in phase space. To leave a valley, it is generally necessary to wait for the system to undergo a particular combination of very improbable excitations. We can generalize the previous method [13] so that every step transfers the system to a new configuration, outside the current set of configurations. Let us assume a set of "internal" configurations and focus on when and to which external configuration the system first moves, without paying attention to the evolution of the system within the internal configurations. We will denote with roman letters the internal and with Greek letters the external configurations. Let us call $\mathcal{M}$ the internal transition matrix, whose elements are $P_{b,a}$, where obviously $P_{a,a} = 0$, and $\mathcal{I}$ the identity matrix. The probability $\tilde{P}_{\alpha,a}$ that, starting in configuration a, the system ends up in an external configuration α after having visited any internal configuration any number of times is [10]

$$\tilde{P}_{\alpha,a} = \sum_b P_{\alpha,b}[(\mathcal{I} - \mathcal{M})^{-1}]_{b,a}, \quad (3)$$

where b ranges over all internal configurations (including a). The average time that the system takes to escape from the internal set is [10]

$$\tilde{\tau}_a = \sum_{\alpha,b} \tilde{P}_{\alpha,b} \tau_b [(\mathcal{I} - \mathcal{M})^{-1}]_{b,a}. \quad (4)$$

We have implemented Eqs. (3) and (4) in our MC algorithm in the following way. Initially, the internal set consists of the single initial configuration. We choose a new configuration with a probability $P_{a,b}$ and incorporate it in the internal set. Every time a new configuration is visited, we add it to the internal set and recalculate the external probabilities according to Eq. (3). The size of the set keeps increasing up to a certain maximum number. The entire procedure is then started with the last configuration serving as the initial set. When we add a configuration to the internal set, we can calculate the new inverse matrix $(\mathcal{I} - \mathcal{M})^{-1}$ efficiently from the old inverse matrix [15]. The maximum size of the internal set is chosen so that the computer time spent in the inversion process is similar to the time for the rest of the tasks. This depends on T and N, and it is of the order of a few hundred. This procedure is similar to one used in the context of spin glasses, where thermal equilibrium was assumed for the internal set [16].

We start from a random configuration and follow the relaxation dynamics at a given T, monitoring all relevant magnitudes. We can reach $t = 2 \times 10^{13}$, corresponding to macroscopic relaxation times, for sizes of up to 1000 sites for $T = 0.001$. For larger sizes, we can reach configurations with low enough energy to be able to continue the dynamics to macroscopic times with the method presented in Ref. [17].

The formation of the CG is a very slow process [5]. To try to understand the relation between the excess energy in the system and the shape of the CG, we have studied the site occupation. Let us define the occupation function $n(\epsilon)\Delta\epsilon$ as the number of occupied sites per unit area with energy $\epsilon_i \in [\epsilon, \epsilon + \Delta\epsilon]$, with respect to the Fermi level. Taking into account the symmetry of the system, the density of states is $g(\epsilon) = n(\epsilon) + n(-\epsilon)$, and the occupation probability $f(\epsilon) = n(\epsilon)/g(\epsilon)$. At equilibrium, $f(\epsilon)$ is a FD distribution characterized by the phonon bath temperature T [18]. The slow formation of the CG suggests a higher degree of excitation for very long times. Surprisingly, we found that the site occupation probability also follows very well a FD distribution, but with a T_{eff} much higher than the real T. In Fig. 1, we show $f(\epsilon)$ at three times $t = 3 \times 10^4$, 7×10^6, and 10^{10}, for a size $N = 2000$ and for $T = 0.001$. The continuous curves are FD distributions with $T_{\text{eff}} = 0.078$, 0.040, and 0.022, respectively.

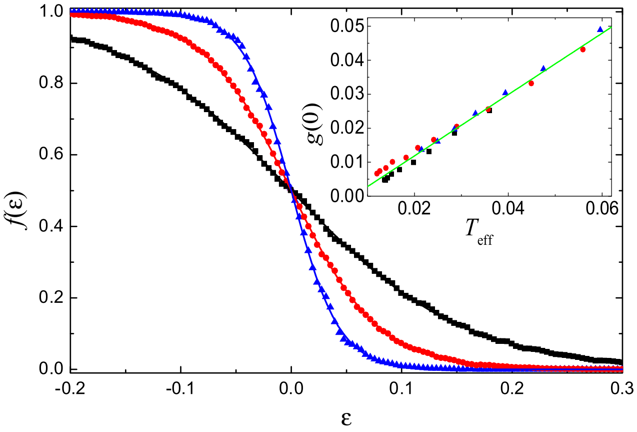

FIG. 1 (color online). Site occupation probability as a function of energy at $t = 3 \times 10^4$ (black squares), 7×10^6 (red circles), and 10^{10} (blue triangles). The continuous curves are fits by FD distributions with $T_{\text{eff}} = 0.078$, 0.040, and 0.022, respectively. Inset: $g(0)$ versus T_{eff} for several t and sizes $N = 500$ (black squares), 1000 (red circles), and 2000 (blue triangles).

After roughly $t \approx 100$, a proper FD distribution is observed, independently of the initial configuration. We have checked that starting from an electron-hole asymmetric initial configuration, designed to reproduce experiments where the sample is excited with light [4], basically does not change the results.

The fit of $f(\epsilon)$ to a FD distribution is so good for all relaxation times, system sizes, and initial conditions that we conclude that there must be some thermalization procedure for slow modes, different from the phonon mechanism for fast modes. Then $T_{\rm eff}$ should affect other slowly relaxing quantities, and it should be measurable in different ways. In equilibrium, a good thermometer for Coulomb glasses is the density of states at the Fermi level $g(0)$, which for 2D systems is proportional to T, due to the linearity of the CG [18]. We have found that in our relaxation processes $g(0)$ is also proportional to $T_{\rm eff}$ as can be seen in the inset in Fig. 1 for different system sizes and relaxation times. Thus, $g(0)$ constitutes an appropriate thermometer to measure $T_{\rm eff}$ experimentally (e.g., by tunneling experiments [19]).

We have a rough argument for the time dependence of $T_{\rm eff}$. The logarithmic time dependence of the relaxation procedure ensures that occupied sites with positive energies cannot have holes at sites with negative energies at distances closer than $r_t = (a/2)\ln(\nu_0 t)$ [5,20]. By an argument similar to the one used to obtain the form of the CG, we expect that this electron-hole distance is proportional to the typical distance between electrons with positive energies. Then $n(\epsilon)$ should obey

$$\int_0^\infty n(\epsilon)d\epsilon \lesssim \frac{1}{r_t^2}. \tag{5}$$

Near the Fermi level, we found that $g(\epsilon)$ is roughly constant in the range $\epsilon \approx T_{\rm eff}$, so that the previous integral is proportional to $g(0)T_{\rm eff}$. We assume that $n(\epsilon)$ approaches the maximum value allowed by Eq. (5), since electron hops, other than very short ones, change significantly the site energies, due to the long-range character of the interaction. Then both $T_{\rm eff}$ and $g(0)$ decrease as

$$g(0) \approx T_{\rm eff} \propto \frac{1}{r_t} = \frac{2}{a\ln(\nu_0 t)}. \tag{6}$$

This behavior is approximately satisfied by our numerical simulations, as we will see later.

As fast degrees of freedom must be locally equilibrated at the real T, and we have seen that site occupancy is governed by $T_{\rm eff}$, we expect the coexistence of two characteristic temperatures, relevant for fast and slow modes, respectively, as in spin glasses [21]. Coulomb glasses are especially suitable to study this subject, since their electron-hole excitations cover an extremely large range of relaxation times and can be readily classified through their hopping distance at high enough T and/or short times. For this reason, we have studied the role of $T_{\rm eff}$ on electron-hole excitations for several hopping distances. At equilibrium, these excitations also follow the FD statistics for the quantity $F(E) = P(E)/[P(E) + P(-E)]$, where $P(E)$ is the number of excitations of energy E. If a configuration has a transition with energy E, the configuration resulting from performing the transition contains the inverse excitation, with energy $-E$. The relative probability is $P(E)/P(-E) = e^{-\beta E}$, which implies FD statistics for $F(E)$.

We have studied the distribution of excitations $F(E)$ for different ranges of the hopping length at several relaxation times, and we have found that short excitations are distributed according to T, while long excitations are distributed according to a time-dependent $T_{\rm eff}$. In Fig. 2, we plot $F(E)$ for excitations with hopping length smaller than 1 (empty symbols) and larger than 10 (solid symbols) for $t = 7 \times 10^6$ (circles) and 10^{10} (squares). The continuous curves are FD distributions for the real T (for $r < 1$) and different $T_{\rm eff}$ (for $r > 10$). For intermediate distances, $F(E)$ does not follow this distribution. $T_{\rm eff}$ obtained for long excitations agrees within 15% with that from site occupation. As there are many more excitations than sites, we have much better statistics, and, at the same time, we avoid finite size problems with the estimation of the chemical potential.

In Fig. 3, we represent $T_{\rm eff}$ as a function of $1/\log t$ for four system sizes $N = 500$, 750, 1000, and 2000, at $T = 0.001$. We first note that, even at macroscopic times, $T_{\rm eff}$ is at least an order of magnitude larger than T. We also recognize a relatively good linear behavior, in agreement with our prediction. The dependence of $T_{\rm eff}$ with system size is fairly systematic. All four curves are roughly parallel and displaced by an amount proportional to $1/L$. In the inset in Fig. 3, we plot the crossing r_c of the fitting lines in the main part of the figure with the horizontal axis as a function of $1/L$. The data fit pretty well a straight line passing through the origin and with a slope α of the order of 1. Then

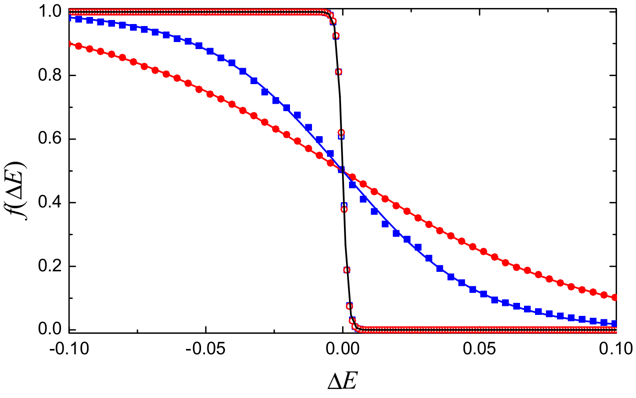

FIG. 2 (color online). $F(E)$ as a function of energy for $t = 7 \times 10^6$ (circles) and 10^{10} (squares). Empty symbols correspond to $r < 1$ and solid symbols to $r > 10$. Continuous lines represent FD distributions.

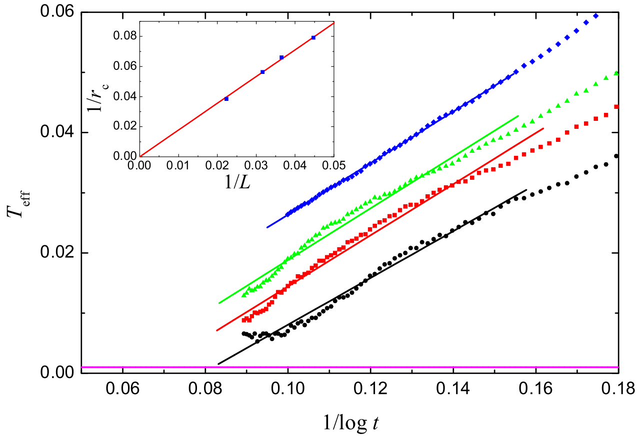

FIG. 3 (color online). $T_{\rm eff}$ as a function of $1/\log t$ for $N = 500$, 750, 1000, and 2000 from bottom to top. The horizontal line corresponds to T. Inset: r_c versus $1/L$.

$$T_{\rm eff} \propto \frac{1}{r_t} - \frac{\alpha}{L}. \tag{7}$$

The determination of $T_{\rm eff}$ from the fluctuation-dissipation ratios [11] is very noisy in the regime considered here. In order to compare this technique with our method, we have studied the CG model considered by Grempel, i.e., without diagonal disorder and at higher T [9]. We found that our main conclusions remain valid, i.e., $g(0) \propto T_{\rm eff}$. The two results for $T_{\rm eff}$ differ by roughly 25%, partly due to the limitations inherent in involving two time variables in the fluctuation-dissipation technique [22].

The resistances of long jumps in hopping conduction, crucial to establish a percolation path, should be calculated by using $T_{\rm eff}$ for the occupation probabilities and T for the phonon excitations. Thus a very important ramification of this Letter is that, having shown the existence of $T_{\rm eff}$ and its time dependence, it allows for a derivation of an analytic expression for the relaxation of the conductance over a large domain of time. A stationary conduction at high enough currents will also produce a $T_{\rm eff}$ that can explain some experimental results [23].

We verified the existence of an effective temperature in relaxation in Coulomb glasses, compatible with recent predictions for spin glasses. We propose that Coulomb glasses are excellent systems to measure $T_{\rm eff}$ through the density of states. The determination of $T_{\rm eff}$ through the excitations can be used in a wide variety of systems and may constitute a practical alternative to the use of the fluctuation-dissipation theorem. $T_{\rm eff}$ is roughly proportional to $1/\ln(\nu_0 t)$. A similar behavior was found in a different system with slow dynamics [24].

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