

Stretched exponential relaxation in the Coulomb glass

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Abstract. The relaxation of the specific heat and the entropy to their equilibrium values is investigated numerically for the three-dimensional Coulomb glass at very low temperatures. The long time relaxation follows a stretched exponential function, $f(t) = f_0 \exp[-(t/\tau)^\beta]$, with the exponent β increasing with the temperature. The relaxation time follows an Arrhenius behavior divergence when $T \rightarrow 0$. A relation between the specific heat and the entropy in the long time regime is found.

PACS. 61.43.Fs Glasses – 61.43.Dq Amorphous semiconductors, metals, and alloys – 64.70.Pf Glass transition

1 Introduction

The relaxation in most glassy systems such as structural glasses, ionic conductor, supercooled liquids, polymer, colloid, and spin glasses [1] deviates strongly from a single exponential relaxation at some temperature T^* well above the static transition. In the three dimensional Ising spin glass model Ogielski [2] observed that the long time regime of the relaxation functions is well approximated by the non-exponential function

$$f(t) = f_0 t^{-x} \exp[-(t/\tau)^\beta], \quad (1)$$

below $T^* = T_G$, where T_G is the Griffiths temperature [3]. In supercooled liquids [4] the characteristic features of the relaxation processes in the long time regime is captured by a stretched exponential or Kohlrausch-Williams-Watts [5] decay function

$$f(t) = f_0 \exp[-(t/\tau)^\beta], \quad (2)$$

with $0 < \beta \leq 1$. This behavior can be obtained by a superposition of purely exponential relaxation process but subject to a broad distribution of relaxation times [6].

The Coulomb glass [7,8] is a prominent example of glassy systems. In heavily doped crystalline semiconductors, amorphous semiconductor-metal alloys, and granular metals, it plays an important role as a semiclassical model for systems of localized states. The out of equilibrium dynamical behavior of the Coulomb glass has been studied previously by Schreiber et al. [9] and Pérez-Garrido et al.

[10]. They determined numerically the transition probabilities between low-energy many-particle states, and studied the eigenvalues of the transition probability matrix. A broad distribution of relaxation times over several orders of magnitude was found in both cases, which reflects the glassy behavior of this system. This broad distribution bring to a power law relaxation function [10]. Moreover, Wappler et al. [11] used the damage-spreading algorithm to study the temporal evolution of the system in thermal equilibrium, and found evidence for a dynamical phase transition at a temperature T^* which depended of the degree of disorder considered. Yu studied the time development of the Coulomb gap considering a self-consistent equation for the density of states [12]. She also observed that very long time scales are involved. Díaz-Sánchez et al. [13,14], considering the dynamic in the configuration space, studied non-ergodic effects on the specific heat and found very long relaxation times. Pastor et al. [15] predicted the existence of a glass phase for a mean-field model of interacting spinless fermions in the presence of disorder. Experimental measurement in electronic system of an Anderson insulator [16] showed a glass phase with aging phenomenon appearing. There it was found that the common relaxation law can be fitted by a stretched exponential function.

The aim of this paper is to study the shape of the relaxation process of the specific heat and the entropy in the Coulomb glass at very low temperatures and in thermal equilibrium. The paper is organized as follows: Section 2 introduces the Coulomb glass model. Section 3 describes the numerical procedure, i.e. how the low-energy many-particle states are obtained numerically, and the dynamic

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used in the simulation. In Section 4, we use this procedure for the investigation of the relaxation process of the specific heat and the entropy. Finally, in Section 5 we extract some conclusions.

2 The model

A practical model to represent the Coulomb glass problem for simulating an impurity band of localized states in lightly doped semiconductors, when quantum interference can be neglected, was proposed in references [7,8]. Later it was also applied to simulate granular metals [17] and conducting polymers [18]. Following reference [19], we consider a three dimensional cubic lattice with N sites. The sites can be occupied by KN ($0 < K < 1$) electrons which are interacting via an unscreened Coulomb potential. Background charges $-K$ are added at each of the lattice sites, guaranteeing electro-neutrality. The disorder is simulated by a random potential ϵ_i . Their values are uniformly distributed between $-B/2$ and $B/2$. This model is represented by the Hamiltonian

$$H = \sum_i (\epsilon_i - \mu) n_i + \sum_{i < j} \frac{(n_i - K)(n_j - K)}{r_{ij}}, \quad (3)$$

where $n_i \in \{0, 1\}$ denotes the occupation number of site i , r_{ij} is the distance between sites i and j according to periodic boundary conditions [20], and μ is the chemical potential. The lattice spacing is taken as unit of distance.

In this paper, we focus in the half-filled impurity band $K = 1/2$, where there is particle-hole symmetry and the chemical potential μ equals zero at any temperature in the thermodynamic limit. We take $B = 2$ in this paper, similar results to the ones presented here are found for other values of B [14].

3 Numerical procedure

Our aim is to simulate the temporal evolution of the Coulomb glass at very low temperatures in thermal equilibrium. For that we first obtain a set \mathcal{S} of low-energy many-particle configurations and later define a dynamic between these configurations. We apply this procedure to study the relaxation of the specific heat and the entropy to their equilibrium values.

3.1 Low-energy configurations

We find the low-energy many-particle configurations by means of a three-steps algorithm [14]. This comprises local search [21,22], thermal cycling [23], and construction of “neighboring” states by local rearrangements of the charges [21,22]. The efficiency of this algorithm is illustrated in reference [14]. In the first step we create an initial set \mathcal{S} of metastable states. We start from states chosen at random and relax these states by a local search algorithm which ensures stability with respect to excitations

from one up to four sites. In the second step this set \mathcal{S} is improved by means of the thermal cycling method, which combines the Metropolis and local search algorithms. The third step completes the set \mathcal{S} by systematically investigating the surroundings of the states previously found.

3.2 Dynamic in the configuration space

We now present a method to study the influence of the duration of the measurement, i.e. the observation time τ_m , on the expectation value of the specific heat c and the entropy S [14]. This method defines a dynamic in the configuration space.

During τ_m the state of the sample travels randomly through its configuration space. A transition between configurations I and J is allowed if the transition time τ_{IJ} is shorter than τ_m . So, at finite τ_m only transitions with characteristic time τ_{IJ} shorter than τ_m contribute to the specific heat. Thus, for a given τ_m , we consider two configurations as connected if their τ_{IJ} is shorter than τ_m , and we group the configurations in clusters according to these connections. These clusters correspond to regions of the configurations space being isolated from each other on the time scale τ_m .

The characteristic transition time between configurations I and J is [7,14],

$$\tau_{IJ} = \tau_0 \exp\left(2 \sum r_{ij}/a\right) \exp(E_{IJ}/T) / Z. \quad (4)$$

In this equation, the quantity τ_0 is a constant of the order of the inverse of the phonon frequency, $\tau_0 \sim 10^{-13}$ s. The sum is the minimized sum over all hopping distances between sites which change their occupation in the transition $I \rightarrow J$. a denotes the localization radius, $E_{IJ} = \max(E_I, E_J)$ where E_I is the energy of the state I , T is the temperature, and Z is the partition function.

Provided, at the beginning of the measuring process, the sample is in one of the states of the cluster α , we measure as value of the mean energy in this cluster,

$$\langle E \rangle_\alpha = \sum_{I \in \alpha} E_I P_I, \quad (5)$$

with

$$P_I = \exp\left(\frac{-E_I}{T}\right) Z_\alpha^{-1}, \quad (6)$$

where Z_α denotes the value of the the partition function of this cluster. The entropy of the cluster is given by

$$S_\alpha = \sum_{I \in \alpha} P_I \ln(P_I). \quad (7)$$

Assuming that the clusters are in thermal equilibrium the specific heat for a cluster α of states is:

$$c_\alpha = \frac{\langle E^2 \rangle_\alpha - \langle E \rangle_\alpha^2}{T^2 N}. \quad (8)$$

The expectation value of c , $\langle c \rangle$, that is the mean value of repeated measurements, is given by the weighted average of c_α :

$$\langle c(T, \tau_m) \rangle = \sum_{\alpha} c_{\alpha} P_{\alpha}, \quad (9)$$

where P_{α} is the probability to find the sample in one of the states belonging to the cluster α . Similarly the expectation entropy, $\langle S \rangle$, is given by

$$\langle S(T, \tau_m) \rangle = \sum_{\alpha} S_{\alpha} P_{\alpha}. \quad (10)$$

Thus $\langle c \rangle$ and $\langle S \rangle$ depend via the cluster structure and P_{α} on τ_m , and also on T . Both the cluster structure and P_{α} are influenced by the history of the sample. In our numerical experiments, we presume that the sample has reached thermal equilibrium before the measurements; thus $P_{\alpha} = Z_{\alpha}/Z$.

4 Relaxation functions

We study the process of relaxation of the specific heat and the entropy according to the methods presented in the previous section. To make the influence of τ_m directly visible, we consider the rate of the values of the specific heat and the entropy for finite and infinite (equilibrium) duration of measurement, respectively:

$$q_c(T, \tau_m) = \frac{\langle c(T, \tau_m) \rangle}{\langle c(T, \infty) \rangle} = \frac{\langle c(T, \tau_m) \rangle}{c_{\text{eq}}}, \quad (11)$$

$$q_s(T, \tau_m) = \frac{\langle S(T, \tau_m) \rangle}{\langle S(T, \infty) \rangle} = \frac{\langle S(T, \tau_m) \rangle}{S_{\text{eq}}}, \quad (12)$$

where c_{eq} and S_{eq} are the equilibrium values. Moreover, in order to characterize the shape of the relaxation process in the long time regime we study the following functions,

$$R_c(T, \tau_m) = 1 - q_c(T, \tau_m), \quad (13)$$

$$R_s(T, \tau_m) = 1 - q_s(T, \tau_m), \quad (14)$$

which go to zero at the equilibrium.

In our simulations we consider the localization radius a equals always 0.2 [24]. Investigating the physical properties of macroscopic systems, we have calculated ensemble averages, and have compared the results for different sample sizes. In order to ensure the convergence of the results on the number of low-energy many-particle configurations in the set \mathcal{S} , we have taken from 25000 to 75000 configuration into account for the cases studied here (the number of configurations to consider depends on the temperature and system size). This election ensures that the width of the related energy interval exceeds the temperature by at least a factor of 25. As in reference [14] for $N \gtrsim 512$ the result are free of finite-size effects. Here we present the simulations for $N = 512$ taking into account 1000 samples for ensemble averages. In order to simplify the notation we consider the variable $t = \tau_m/\tau_0$, i.e. our unit of time is τ_0 .

We first study the shape of the relaxation functions R_c and R_s . Figure 1 shows R_c and R_s as a function of t , for five different temperatures. Here we can see that the long time regime, $R_c \lesssim 0.5$ and $R_s \lesssim 0.5$, may be reasonably fitted by a stretched exponential function. In the worse case, $T = 0.014$, the 75% of the stretched exponential function is compared with the data although for $T = 0.006$ it becomes the 80%. For all temperatures considered here R_c is equal to R_s in the long time regime (within the errors). This form of the relaxation functions brings us to think in a broad distribution of relaxation times in the system, as it has been found in references [9,10]. We have also attempted a fit of our results with equation (1) but the better fit is made with $x \simeq 0$, i.e. we recover equation (2).

The influence of the temperature on the relaxation process is studied. The values of β , obtained by fitting R_c and R_s with equation (2), are presented in Figure 2. We see that β increases with the temperature and it is the same for both relaxation functions, R_c and R_s , at each temperature (within the errors). This behavior of β is also found in other glassy systems [25]. We could expect some temperature T^* , higher than the temperatures studied here, where $\beta = 1$, recovering the exponential relaxation process at $T > T^*$. Unfortunately our method does not let to investigate higher temperatures because we must take a finite number of configurations into account in the set \mathcal{S} (we have taken the maximum of configurations that we have been able in order to make the calculations in a reasonable time).

For an estimate of the long time relaxation we use τ from the fit of equation (2) of R_c and R_s . As we can see in Figure 3, τ_r is reasonably fitted by an Arrhenius behavior divergence as $T \rightarrow 0$

$$\tau_r(T) = a \exp[T_0/T], \quad (15)$$

with $a = 1.4 \times 10^8$ and $T_0 = 0.1106$ for R_c ; for R_s we have $a = 1.2 \times 10^9$ and $T_0 = 0.0946$. The differences in a and T_0 from both relaxation functions are because of the sensitivity of τ on the details of the fit.

We now consider the relation between the specific heat and the entropy in the long time regime. At the equilibrium both are connected by the relation

$$\frac{\partial \langle S \rangle}{\partial T} = \frac{N}{T} \langle c \rangle. \quad (16)$$

As we will see below due to the temperature dependence of R_c and R_s this thermodynamic relation only hold at the equilibrium, i.e. when $t \gg \tau_r$. We have seen that $R_c \approx R_s = R$ in the long time regime, so that $q_c \approx q_s = q$ and we can write from equations (11) and (12)

$$\langle c \rangle = q(T, t) c_{\text{eq}}, \quad (17)$$

and

$$\langle S \rangle = q(T, t) S_{\text{eq}}, \quad (18)$$

after we have substituted $\langle c \rangle$ and $\langle S \rangle$ in equation (16) and regrouped terms we have

$$\left(\frac{\partial S_{\text{eq}}}{\partial T} - \frac{N}{T} c_{\text{eq}} \right) q = -S_{\text{eq}} \frac{\partial q}{\partial T}. \quad (19)$$

The left term of this equation is zero while the right term is different of zero when the system is not at the equilibrium. So that we can write a new relation for the entropy and the specific heat. It follows as

$$\frac{\partial \langle S \rangle}{\partial T} - S_{\text{eq}} \frac{\partial q}{\partial T} = \frac{N}{T} \langle c \rangle. \quad (20)$$

At the equilibrium $\partial q / \partial T$ vanishes and we recover equation (16). In order to check this expression in Figure 4 we compare the specific heat calculated from this equation, and from the values of the entropy, with the specific heat calculated from equation (9). The derivatives are approximated with finite differences. We can see that both agree very well in the long time regime.

5 Conclusion

We have studied the relaxation process of the specific heat and the entropy for the three-dimensional Coulomb glass at very low temperatures and in thermal equilibrium. The long time relaxation regime follows a stretched exponential function, with the exponent $\beta < 1$ increasing with the temperature, which is an indication of a broad distribution of relaxation times in the system. From these results, we could expect a dynamical transition at some temperature T^* (above of the temperatures studied here) where $\beta = 1$. The exponential relaxation process would be recovered for $T > T^*$. In reference [11] it was found $T^* > 0.03$ for the two-dimensional Coulomb glass although the effects of distance-dependent transition probabilities were not taken into account. Moreover, the relaxation functions found here in the thermal equilibrium are very different from the power law relaxations found when the system is out of equilibrium [10]. Nevertheless, in out of equilibrium experiments stretched exponential relaxation functions are found [16]. The relaxation time diverges as an Arrhenius law when $T \rightarrow 0$. Finally, we have found a relation between the specific heat and the entropy in the long time regime.

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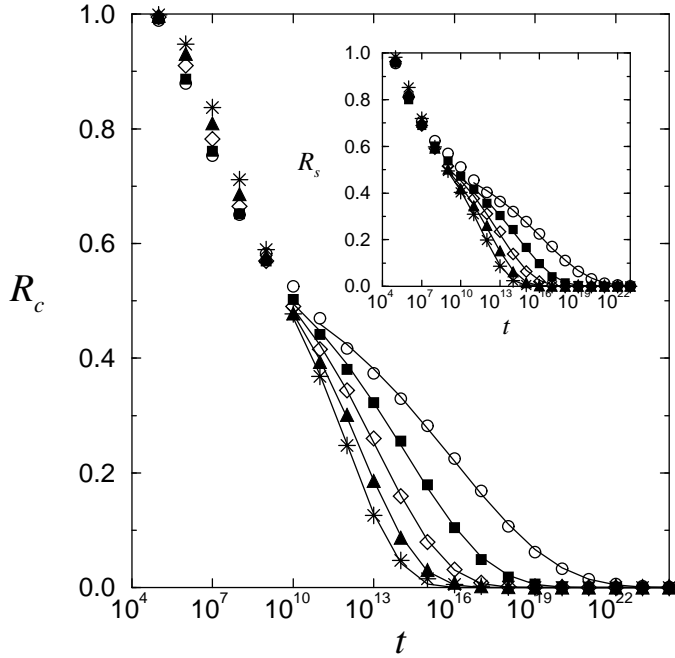


Fig. 1. Relaxation functions of the specific heat as a function of t , for the temperatures $T = 0.006$ (\circ), 0.008 (\blacksquare), 0.01 (\diamond), 0.012 (\blacktriangle) and 0.014 ($*$). Inset the relaxation functions of the entropy for the same temperatures. The solid lines correspond to fits with the function $f(t) = f_0 \exp[-(t/\tau)^\beta]$.

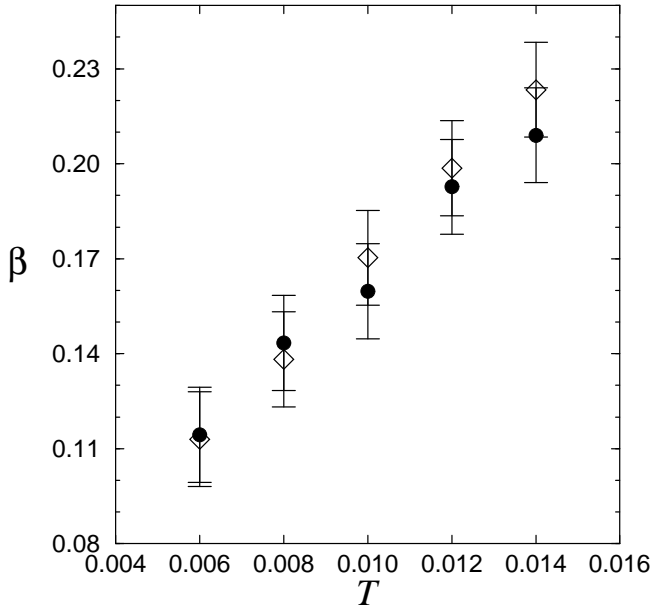


Fig. 2. Parameter β as a function of the temperature, obtained by fitting R_c (\bullet) and R_s (\diamond) with the function $f(t) = f_0 \exp[-(t/\tau)^\beta]$.

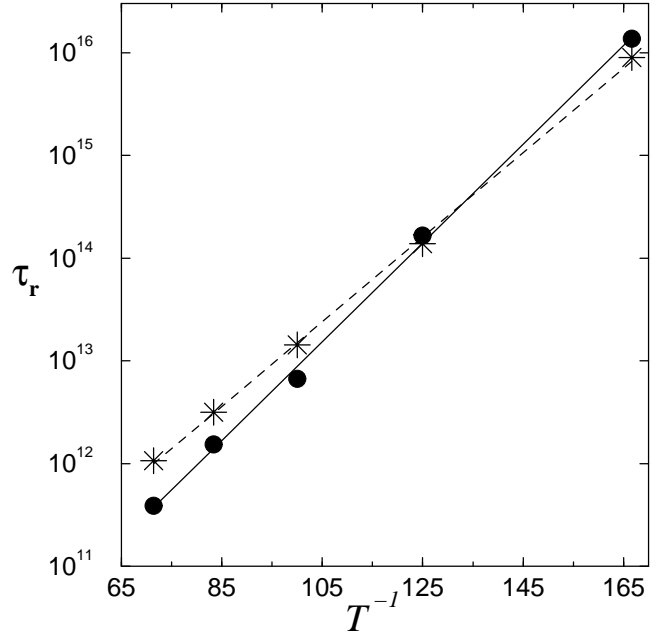


Fig. 3. Relaxation time τ_r as a function of the inverse of the temperature, obtained by fitting R_c (\bullet) and R_s ($*$) with the function $f(t) = f_0 \exp[-(t/\tau_r)^\beta]$. The lines are fits with the function $\tau_r(T) = a \exp[T_0/T]$. The values of τ_r correspond to the values obtained from the fit in Figure 1.

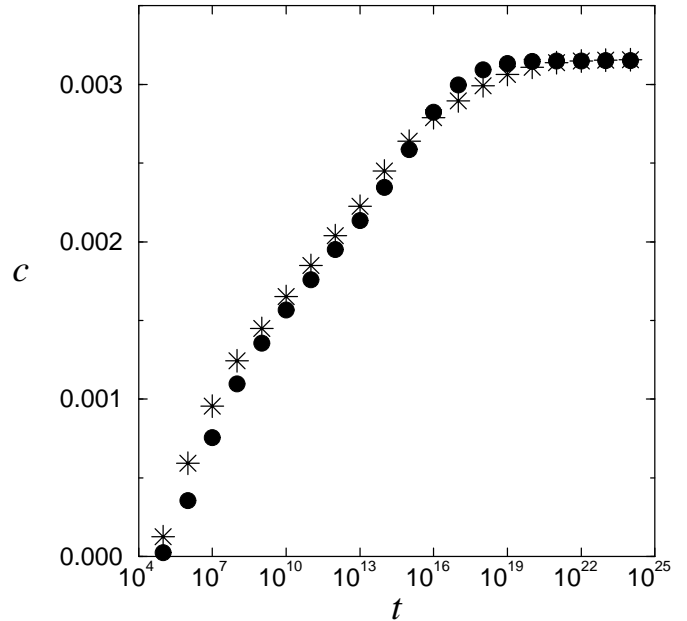


Fig. 4. Specific heat as a function of time obtained from equation (20) and the values of the entropy ($*$) and from equation (9) (\bullet), for $T = 0.008$.