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NUMERICAL STUDY OF THE TRANSITION GLASS TEMPERATURE IN TWO-DIMENSIONAL GLASSY CHALCOGENIDES

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In this paper, we investigate the existence of a transition glass temperature in amorphous chalcogenides by means of numerical simulations in the two-dimensional Coulomb glass model. The powerful algorithm we employ enables us to explore greater sizes and lower temperatures than other numerical approaches. From the results we can infer that the 2D system is compatible with a dynamic glassy transition at zero temperature, in agreement with previous preliminary studies.

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1. Introduction

The connection between glassy chalcogenides and electron glasses has been pointed out in some recent publications [1-3]. This relationship describes the amorphous chalcogenide as a strongly correlated system with localized states in which, at very low temperatures, the combined effect of disorder and Coulomb interactions plays an essential role. In this fashion, the system exhibits all the essential electronic properties of Coulomb glasses: loss of ergodicity, memory effects or aging, among others, which are also features common to most glassy systems.

The deep comprehension of the glassy state is still a challenge for scientists, and an open question in the nature of electron glasses is, for example, its critical behavior and the existence of a transition glass temperature. The origin of this kind of studies comes from the early eighties, when Davis, Lee and Rice approached the problem of a regular 3D lattice with diagonal disorder [4]. The analysis of the response functions, i.e., the thermal capacity and the electric susceptibility, gave them evidences of the existence of a phase transition, but the study failed in the characterization of the related order parameter. In the nineties, the importance of the diagonal disorder was pointed out by Grannan and Yu [5], and Votja and Schreiber [6]. The first ones found a transition in 3D systems without diagonal disorder, while the second ones limited the validity of that study precisely to the absence of such disorder. Since there, the question became a common topic in the literature on Coulomb glasses and glassy chalcogenides [7-9].

Modern numerical simulations can shed light on the problem, due to the capability of modern computers and the efficiency of the new simulation algorithms [10, 11]. Among them, the work of Grempel [12] is one of the most representatives, which results are compatible with a glass transition at zero temperature. In our work, we present the numerical results for simulations in 2D systems in samples with diagonal disorder and sites places randomly. We measure the time that the system employs to reach equilibrium for several temperatures. We explore much larger sizes than Grempel and lower temperatures by means of new simulation algorithms, and employ a much easier procedure to determine relaxation time of the system, that is, the time for reaching the

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equilibrium state. As we will see, the results are also compatible with a freezing transition at $T = 0$, which appears as a common result in current numerical simulations for these 2D systems.

2. Mathematical model and numerical details

The standard tight-binding Hamiltonian for describing Coulomb glasses is [2, 13]

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

This classical Hamiltonian neglects the effect of the quantum energies of tunneling between sites, because in the regime of strong localization are much smaller than the rest of relevant energies of the system: disorder, interaction and thermal energies. In expression (1), n_i is the occupancy number of site *i*, which can either be 0 or 1, and r_{ij} is the distance between sites *i* and *j*. Variable ϕ_i is the so called *random site potential*, which represents the random energy of site *i*. We choose its value from a uniform distribution between *–W/2* and *W/2*. To conclude, we define *K* as the compensation, which ensures the electrical neutrality of the system.

We investigate the dynamics of insulating samples doped with impurities randomly placed, in the so called *variable-range hopping* regime [13] at very low temperatures. The system thus behaves as a dielectric sample in which the dynamics is established by jumps of electrons from occupied to empty sites. It is considered that the electrons are placed in the same positions that the impurities, and so the localization length, ξ, is considered very small [14]. We study squared samples of lateral dimension *L* and apply periodic boundary conditions.

The units employed in this work are the same considered in previous works [2, 3]. This time, our range of temperatures is $[0.05, 0.1]$ and the typical size of the samples is $N = 1000$.

We employ two kind of algorithms in our numerical simulations. First of all, specific optimization algorithms, designed to find the lowest energy levels of the system and their corresponding configurations (*i.e.* occupations), for each temperature [15, 16]. In particular, we are interested in the ground state. Secondly, kinetic Monte Carlo algorithms based in the work of Tsigankov *et al*. [17]. The key ingredient of our work is to follow the relaxation of the system through equilibrium by starting with two different initial conditions, that is, the occupancy of the electrons: (a) random occupancy of sites, which represents a typical quench at low temperatures and (b) the ground state, since the specific optimization algorithms also determine the occupancy of the lowest energy levels. So, we perform two different dynamic simulations for each temperature using Tsigankov's algorithm and determine the time at which both data converge, which we call the *relaxation time*, τ_{eq}.

3. Results and discussion

In the literature on other glassy systems, such as spin glasses, some authors suggest that the dimension from which a glassy transition can be found is three [18]. Somehow, the existence of disorder would increase in one unit that critical dimension with regard to the model without disorder. Other works on the same field, on the contrary, estimate the critical dimension one less unit [19]. The mapping of the Coulomb glass model to the long interaction range spin glasses one [13] suggests that the results obtained in these will remain valid for those. Nevertheless, as commented, this topic is nowadays and open question. Among all the numerical studies that measure the relaxation time to equilibrium in Coulomb glasses, τ_{eq}, Grempel (2004) is one of the most representatives, as remarked in section 1. Data were obtained by means of Monte Carlo simulations in a two dimensional system without diagonal disorder. To measure the dependence of τ_{eq} on *T*, Grempel used Bhatt and Young's method [18], based in the convergence of two correlation functions. The first one measures the correlation of an observable corresponding to two

samples with the same degree of disorder, at the same time t_0. The second one refers to the same sample and measures the correlation of the observable at the instants t_0 and $2t_0$. Magnitude τ_{eq} is defined as the time at which both curves converge. The quantitative analysis is consistent with a divergence of τ_{eq} at $T = 0$, thus it is obtained that

$$\tau_{\mathrm{eq}} \propto \exp(A/T)$$

where A is a constant. The data from Grempel show an overlap of the curves up to a temperature $T = 0.04$, although such low temperatures are not representative due to finite size effects [13].
In our numerical simulations we have calculated, likewise, the value of τ_{eq}, for the temperature range $T \in [0.05, 0.1]$. The main difference with Grempel's model is that we consider diagonal site disorder, $\phi_i \neq 0$. In principle, Grempel data should be replicable, since strong arguments assert that, in case of a glassy phase transition to occur in two dimensions, it should be reached in an easier way in the problem without diagonal disorder. This is due to the explicit symmetry between electrons and holes for that situation, thereby leading below a critical temperature to a phase with two symmetric energy minima, like an Ising magnet, for example. Nowadays, Grempel's work is considered as a seminal study, but it considers too small samples, tipically $N = 64$, in which, as commented, finite size effects can't be neglected [13]. Besides, some of the temperatures he treated are too high, which implies that the Coulomb gap is filled enough and the system approaches the crossover zone through the non-interacting regime, described by Mott's law. In our simulations, the upper limit for T is 0.1, which guarantees that the gap is only partially filled, as we can see in Figure 1, and so the system is placed inside the pure interacting regime. We show the form of the Coulomb gap in equilibrium for a system of size 1000 at $T = 0.1$ with diagonal disorder. The data were obtained by following the standard procedure described in [11]. Variables ε and $g(\varepsilon - \varepsilon_F)$ are, respectively, the site energy and the normalized density of states, as defined in [3]. Quantity ε_{F} is the site energy obtained from the ground level.

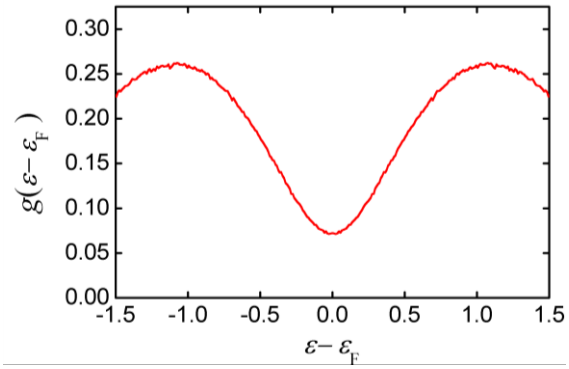

Fig. 1. Partial filling of the Coulomb gap in equilibrium for a system of size 1000 at T = 0.1

The procedure followed to determine τ_{eq} in our simulations differs from that used by Grempel. It explores larger sizes and lower valid temperatures. As remarked above, we perform two simulations at each temperature for the same sample built. In one of them we start in a totally random configuration and measure the energy relaxation towards equilibrium, while in the other one we start in the ground state, monitoring the same magnitude. The time for which the two curves converge is our value of τ_{eq}. We perform the simulations up to a maximum time of 10^7 Monte Carlo steps using the mentioned Tsigankov *et al.* algorithm [17], and we achieve an overlap of both curves up to a temperature $T = 0.05$. Below it, for our conditions, $\tau_{\mathrm{eq}} < t_{\mathrm{sim}}$, where t_{sim} is the total simulation time.

In Figure 2 we show the typical overlap of the two curves to determine the value of τ_{eq}. The black line represents the quench experiment, that is, relaxation of the energy E from a random

configuration. The red line corresponds to the relaxation from the ground state configuration. In our simulations, the magnitude of τ_{eq} has been determined by assuring that both curves only differ a fixed small percent.

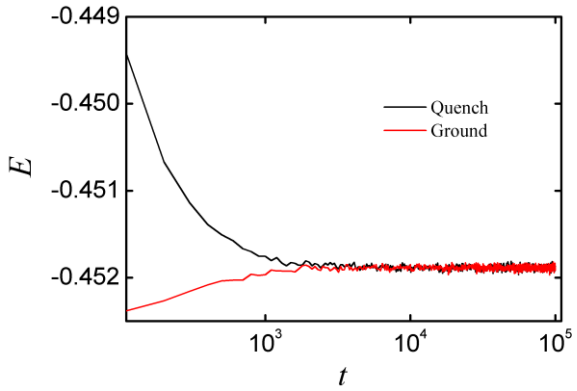

Fig. 2. Relaxation of the energy, E, as a function of time, t, for a system of size 1000 at T = 0.08, starting by two different initial conditions of occupancy: random initial configuration (quench, black line) an d ground state configuration (red line). The convergence of both curves defines the equilibrium time.

In figure 3 we represent ln τ_{eq} vs $1/T$ for a system of size 1000. A good linear dependence is shown, which is compatible with a dynamic glassy transition at $T = 0$. The linear fit has been extrapolated to $1/T = 0$, and it shows an intersection with the vertical axis equal to 1.6. This fact suggests that, at infinite temperature, the equilibration time is not zero, as might be expected a priori. This fact is associated to the dynamic of the program, since it needs at least one Monte Carlo step to carry out the relaxation.

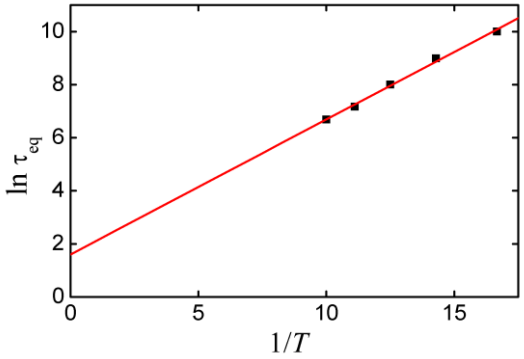

Fig. 3. Natural logarithm of time vs 1/T for a system of size 1000. The good linear fit of the data is compatible with a dynamic glassy transition at T = 0. The extrapolation of the fitted line results in a value of ln $\tau_{eq} = 1.6$ for infinite temperature.

4. Conclusions

In this paper we show a simple and illustrative numerical method to determine the presence of a glass transition temperature in two-dimensional glassy chalcogenides. The glass transition is still a challenge for scientists and new numerical approaches have to be developed to achieve a full comprehension of the glassy state. Compared with other preliminary studies, our procedure explores greater sizes and lower temperatures. The analysis of the range of temperatures relevant for the study is also an advantage in our approach, because extreme values have to be controlled. Lowest temperatures should lead to finite size effects, while highest ones may put the

system into the crossover zone, approaching the non-interacting regime. Besides, the procedure for determining the equilibrium time is easier to perform than in previous simulations.

The versatility of the method can be employed to study pseudo two-dimensional or three dimensional systems, which are more related to the real experiments, topics in which we are working, nowadays.

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